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On The Variability of Mineral Dust and the Intertropical Convergence Zone Over West Africa: A synthesis of observations, climate models and geochemical analysis

A Dissertation Presented

by

Owen Doherty

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Abstract of the Dissertation

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Mineral dust in the atmosphere is of great importance through its impacts on climate, atmospheric chemistry and biogeochemical processes. The year-to-year and season-to-season variability of mineral dust transport are not well understood. This work focuses on investigating and explaining the inter-annual variability of mineral dust transport by an approach that combines data analysis from in-situ observations, global climate models, multiple reanalysis products and geochemical laboratory analysis. Linking a 38-year record of mineral dust concentration at Barbados to variations in position and intensity of the zone of nearsurface convergence over West Africa, a part of the Intertropical Convergence Zone (ITCZ) a relationship between the latitude of the ITCZ over West Africa and mineral dust at Barbados is demonstrated during winter (r = -0.69) and summer (r = -0.47). This finding represents an improvement over previous studies, which related mineral dust transport to changes in the NAO and ENSO. Southward displacement of the ITCZ leads to favorable winds for dust emission near the surface in both seasons, and in summer also leads to reductions in precipitation over the Sahel, which in turn increases aridity. The newly developed climate index that quantifies the variability of the ITCZ is then used to assess the performance of a suite of 11 global climate models (from CMIP3) in both hindcasts of the 20th century and predictions of the 21st century. A northward trend in the position of the ITCZ is observed in the summer of the 20th century, and predicted in both summer and winter of the 21st century. Northward migration of the ITCZ in the 21st century is expected to reduce dust load by $-1.9 \ \mu g m^{-3}$ to $-3.8 \ \mu g m^{-3}$ in summer and by $-1.5 \ \mu g m^{-3}$ to $-2.2 \ \mu g m^{-3}$ in winter. To establish provenance of mineral dust the mineralogy and elemental composition of dust samples collected from four continents are analyzed and via principle component analysis it is demonstrated that continental sources can be separated.

Dedication Page

Churn, churn.

Forward and onward!

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¹ Chapter 1

² Introduction

3 1.1 Background

4 1.1.1 Importance of Mineral Dust

Mineral dust, particles of Earth derived material suspended in the atmosphere, has been 5 of interest to the scientific community since the 19th century. The record of mineral dust 6 in modern scientific literature begins with Darwin's observations aboard the H.M.S. Beagle 7 [Darwin, 1846]. Mineral dust transport did not receive attention again until the 1960's when 8 Joseph Prospero began his seminal work on Saharan Mineral Dust in the Tropical North 9 Atlantic Ocean [Prospero and Carlson, 1972]. Recently, as the importance of mineral dust 10 to many natural systems (climate, chemical, geological and biological) has become clear, the 11 number of papers published on mineral dust has grown exponentially as seen in Figure 1.1. 12

¹³ Implications for Climate

Recent IPCC reports have identified mineral dust aerosols as a major source of uncertainty
in climate predictions [Solomon et al., 2007]. Much of this uncertainty extends from mineral
dust exerting both a direct radiative forcing on climate as it interacts with solar and longwave
radiation [Andreae, 1996; Sokolik and Toon, 1996; Tegen et al., 1996; Miller et al., 2004] and

an indirect forcing on climate as dust particles play an important role for various aspects of 18 the microphysics of clouds [Wurzler et al., 2000; Rosenfeld et al., 2001; Mahowald et al., 2003; 19 Levin et al., 2005], including mixed-phase clouds [DeMott et al., 2003], thin ice clouds [Sassen 20 et al., 2003; DeMott et al., 2003] and warm clouds [Levin et al., 2005]. By affecting cloud 21 microphysics, mineral dust may impact climate indirectly, as it alters the cloud lifetime and 22 their reflectivity. In summary, depending on the conditions, these forcing mechanisms can 23 impart a positive (warming) or a negative (cooling) forcing on the climate system as mineral 24 dust both absorbs and reflects radiation, and by changing the properties and lifetime of 25 clouds can either warm or cool the planet. The overall effect of mineral dust on the climate 26 system varies with a number of factors, including the composition, size distribution and 27 shape of the dust particles, composition of underlying surface, the quantity and the vertical 28 profile of the mineral dust and the strength of solar insolation. 29

The largest source of mineral dust globally is the Sahara Desert [Prospero et al., 2002; 30 Washington et al., 2003]. Mineral dust originating from this region, so called "Saharan 31 mineral dust", impacts the adjacent tropical climates. Saharan mineral dust has been linked 32 with the frequency and intensity of Atlantic hurricanes [Dunion and Velden, 2004; Evan 33 et al., 2006; Lau and Kim, 2007b,a; Braun, 2010]. Radiative forcing from mineral dust has 34 been shown to shift the location and strength of the tropical rainband over the Atlantic 35 Ocean [Huang et al., 2009] and may impact the ITCZ-monsoon circulation over Africa [Lau 36 et al., 2009a; Kim et al., 2010; Wilcox et al., 2010]. Changes in the conditions of the tropics 37 have been shown to impact the mid-latitudes of the globe, thus a forcing from the presence 38 of mineral dust in the tropics may have global impacts. As mineral dust abundance impacts 39 regional climate, and regional climate in turn impact the abundance of mineral dust, there 40 is potential for a dust feedback loop [Rosenfeld et al., 2001]. 41

⁴² Implications for Atmospheric Chemistry

Mineral dust particles in the atmosphere have a large impact on atmospheric chemistry. 43 Dust particles provide a large surface area on which heterogeneous chemical reactions may 44 occur [Dentener et al., 1996; Grassian, 2002; Bian and Zender, 2003; Krueger et al., 2004]. 45 Heterogeneous reactions involving sulfur dioxide on mineral dust may result in an important 46 sink for atmospheric sulfur. Sulfur dioxide and sulfides often become deposited on the surface 47 of mineral dust particles, increasing the hygroscopicity of the dust particles and reducing the 48 sulfur load of the atmosphere [Bauer et al., 2004; Bian and Zender, 2003]. This reduction 49 in atmospheric sulfur, and thus atmospheric scattering, acts as a positive radiative forcing 50 [Bauer et al., 2004]. Atmospheric chemistry also impacts mineral dust, by coating mineral 51 dust particles in turn reducing the ability of mineral dust to act as an ice nucleus. Photolysis 52 rates, important in many chemical reactions in the atmosphere are affected by the presence 53 of mineral dust [Dickerson et al., 1997]. 54

55 Biogeochemistry

As discussed, mineral dust has large impacts on the atmosphere while it is airborne. Once 56 deposited, mineral dust has similarly large impacts on biological, chemical and geological 57 systems. Mineral dust is rich in silicon, calcium, iron and aluminum [Prospero and Savoie, 58 1989; *Jickells*, 1999] as well as other trace nutrients, which may be important in many 59 regions of the global ocean that are considered to be nutrient-limited [Fung et al., 2000; 60 Meskhidze et al., 2003, 2005]. These regions feature a physical environment that is conducive 61 to planktonic growth but lack essential macro or micro nutrients [Jickells, 1999]. Iron has 62 been shown to be a common limiting micronutrient that is prevalent in mineral dust, and in 63 turn mineral dust deposition onto the global ocean has been shown to be an important source 64 of iron to the surface ocean [*Jickells*, 1999]. As a result, understanding the atmospheric 65 distribution of mineral dust is important to understanding primary production in the ocean. 66 Encountering suitable circumstances the deposition of mineral dust can lead to algal blooms 67

[Walsh and Steidinger, 2001]. The long-range transport of mineral dust from the Sahara
across the Atlantic is significant enough to be responsible for the formation of the soils of
many Caribbean islands and the Amazon Basin [Borg and Banner, 1996; Herwitz et al.,
1996; Muhs et al., 2007; Muhs et al., 2012]. Saharan mineral dust is an important supplier of
both phosphate and potasium for the Amazon Basin [Swap et al., 1992; Koren et al., 2006].

⁷³ 1.1.2 Global Sources and Transport Routes of Mineral Dust

This study focuses on mineral dust reaching the North American continent. Mineral dust
reaches North America primarily from two major sources, multiple deserts in Asia and the
Sahara Desert in North Africa.

Arid and semi-arid regions represent potential sources for mineral dust. The majority of mineral dust emitted into the atmosphere originates from the arid Sahara Desert and the surrounding semi-arid Sahel [*Prospero et al.*, 2002; *Washington et al.*, 2003], with the highest concentrations of mineral dust occurring in the summer season of June through September. Mineral dust from Asian deserts is the second largest source of mineral dust with emissions maximized in spring [*Massie et al.*, 2004]. Arid regions in North America, South America, South Africa and Australia represent lesser sources of mineral dust to the atmosphere.

Saharan mineral dust in summer is transported long distances, often traveling at least 84 5,000 km and persisting in the atmosphere for periods in excess of a week [Prospero et al., 85 1970 before reaching the Americas. Saharan mineral dust is able to persist in the atmosphere 86 for long periods of time due to the formation of Saharan Air Layer. The Saharan air layer 87 forms as hot, dry air from the Sahara moves over the comparatively cool, damp air of the 88 Tropical North Atlantic [Carlson and Prospero, 1972]. The elevated Saharan airmass has 89 very high static stability, resulting in little or no vertical mixing and reduced probabilities of 90 precipitation [Carlson, 1979]. As such, mineral dust aerosols in this layer tend to be longer 91 lived than aerosols emitted from other deserts, allowing for increased transport downwind. 92 The occurrence of Saharan air layers is found dominantly in the summer months, occurring 93

less frequently in the spring and fall, and infrequently in the winter [*Chiapello and Moulin*,
2002; *Karyampudi and Pierce*, 2002]. This layer can extend to about 5000 m vertically,
resulting in elevated transport [*Petit et al.*, 2005].

While summer is the peak season for mineral dust transport from the Sahara, dust 97 emissions form the deserts in central Asia are maximized in spring when meteorological 98 conditions are dry and windy [Sun et al., 2001]. Strong westerlies during spring across 99 the Pacific allow for inter-continental transport of dust [Merrill et al., 1989]. However, as 100 generally less dust is emitted in the Gobi, Taklamakan and other Asian sources the distance 101 between the Asian deserts to North America is greater, and the atmospheric stability over 102 the Pacific is lower than in the Saharan case, thus smaller quantities of dust are expected to 103 reach other continents. 104

¹⁰⁵ 1.2 Mineral Dust Variability

Using both sediment and ice cores, reconstructions of mineral dust over geologic time indi-106 cates variability on the order of 2 to 4 fold increases [$McGee \ et \ al., 2010$] from interglacial to 107 glacial periods. Some of this variability could be tied to additional sources of mineral dust 108 emerging during glacial periods over such regions as North America, Europe and Siberia 109 [Mahowald et al., 2006]. Recently McGee et al. [2010] show that wind gustiness controls the 110 quantity of mineral dust emitted to the atmosphere over geologic time. Changes in precipita-111 tion between glacial and interglacial periods over semi-arid and arid regions like the Sahara 112 have also been shown to be important in controlling mineral dust emission over geologic time 113 [Stuut et al., 2002]. 114

¹¹⁵ More recently, during the instrumental record mineral dust has been estimated or mea-¹¹⁶ sured via satellite and by surface based stations, respectively. For example, much attention ¹¹⁷ has been paid to a long-term record of mineral dust at Barbados where dust concentrations ¹¹⁸ have been measured from 1965 through present [*Rydell and Prospero*, 1972; *Prospero and*

Nees, 1977; Glaccum and Prospero, 1980; Prospero and Carlson, 1980; Prospero and Nees, 119 1986; Savoie et al., 1987; Moulin et al., 1997; Chiapello et al., 1999; Prospero and Lamb, 120 2003; Ginoux et al., 2004; Chiapello et al., 2005; Prospero et al., 2008; Trapp et al., 2010]. 121 Long-range transport of mineral dust, such as that to Barbados, is subject to considerable 122 inter-annual and intra-annual variability due to a number of physical factors. Winds over 123 the Sahara and Sahel control emissions of mineral dust [Washington et al., 2003; Washington 124 and Todd, 2005, and winds downstream of the emission region affect subsequent transport 125 [Chiapello et al., 1995; Riemer et al., 2006; Doherty et al., 2008]. Precipitation and vege-126 tation in the Sahel have been suggested as key factors in the dust emission process *Evan* 127 et al., 2006a]. Precipitation over the source region can modulate dust emission, and precip-128 itation over the transport region can remove dust from the atmosphere [Tegen and Funq, 129 1994; Zender et al., 2003]. While this system has been studied in detail in the past 50 years, 130 two key factors, wind and precipitation, have emerged as being critical in both geologic and 131 contemporary time. 132

¹³³ Understanding the year-to-year variability of this dust load is critical as mineral dust ¹³⁴ originating from the Sahara is the dominant aerosol type over much of West Africa and ¹³⁵ the Tropical North Atlantic [*Formenti et al.*, 2008] and in turn has complex impacts on the ¹³⁶ climate and biogeochemistry of the region. The focus of this dissertation is in explaining ¹³⁷ the year-to-year variability of mineral dust reaching North America over the recent past and ¹³⁸ near future, focusing on the Caribbean and Southeastern United States.

While the long-term average quantity of mineral dust emitted in Africa and transported over the Tropical North Atlantic Ocean is at its peak in summer, this quantity varies greatly from year to year. Much effort has gone into improving our understanding of the processes that are responsible for this variability. Observations of Saharan mineral dust over the Atlantic Ocean and at Barbados have been related to variability in the North Atlantic Oscillation (NAO) [Moulin et al., 1997], particularly in the winter season [Chiapello and Moulin, 2002]. Similar relationships have been diagnosed from global chemistry and transport model results [Ginoux et al., 2004]. Prospero and Lamb [2003] related mineral dust in Barbados and an El Niño-Southern Oscillation (ENSO) index. Dust transport to the Americas has been liked to the position of the Azores High in winter [Riemer et al., 2006] and in summer to the position of the Azores High and Hawaiian High [Doherty et al., 2008]. Schwanghart and Schütt [2008] found no connection between regional weather systems and dust emission in summer, instead suggesting that dust emission in West Africa is connected to regional convection.

In this work we show that the mechanisms causing variability of mineral dust concen-153 tration at Barbados depend on the season and are linked to the migration of the ITCZ over 154 West Africa. During winter the variability can be attributed to changes in emissions [Do-155 herty et al., 2012a]. In contrast, during summer the variability can be explained by increased 156 aridity driven by reductions in precipitation in the Sahel, leading to increased emissions. In 157 both seasons it is the southward migration of the ITCZ that leads to increased dustload at 158 Barbados through changes in near surface wind and precipitation, respectively. To obtain 159 these results it was necessary to develop a framework to quantify the movements of the ITCZ 160 over West Africa. 161

¹⁶² 1.3 Intertropical Convergence Zone Over West Africa

In this work we present a new perspective on mineral dust transport in the region by fo-163 cusing on the role of the Intertropical Convergence Zone (ITCZ) over Africa rather than 164 on global teleconections indices as has previously been done. The ITCZ is a global belt 165 of inclement weather that generally occurs where the trade winds converge and form the 166 ascending branch of the Hadley Circulation. Annually the ITCZ moves north and south 167 following the progression of annual solar maximum [Folland et al., 1991]. Over the ocean the 168 ITCZ circulation is relatively simple whereas over land the structure of the ITCZ becomes 169 more complicated. When the ITCZ occurs over land it can be referred to as a monsoon. For 170
example over Africa parts of the ITCZ are referred to as the West Africa Monsoon (WAM). Much ambiguity exists in terminology regarding the ITCZ [*Nicholson*, 2009], owing to the increased complexity of the system over land. Over West Africa the annual northward migration of the ITCZ-WAM brings critical seasonal rainfall to areas of the Sahel and southern Sahara [*Nicholson*, 2009]. As such the location and strength of the ITCZ has implications for seasonal soil and vegetative conditions, and likely dust emission and transport.

It has been previously noted that a relationship between the ITCZ and mineral dust 177 likely exists. The tradewind circulation associated with the ITCZ blows through the area 178 downwind of the Sahara and Sahel where mineral dust loads are the highest *Prospero and* 179 Carlson, 1972; Moulin et al., 1997]. Prospero and Carlson [1972] first showed in a field 180 experiment that the dust belt was located directly to the north of the ITCZ. Moulin et al. 181 [1997] and later Evan et al. [2006b] showed using satellite observations of mineral dust that 182 the dust belt moves north blowing dust into the Caribbean ahead of the ITCZ in summer, 183 and returns south blowing dust into South America in winter. Precipitation in the tropical 184 North Atlantic basin and surrounding locations have been shown to be dependent on the 185 location of the ITCZ [Kapala et al., 1998] and multiple studies have linked mineral dust in 186 Barbados with precipitation in the Sahel region of Africa [Prospero and Nees, 1986; Chiapello 187 et al., 2005]. Although work has been done linking the movement of the ITCZ to changes in 188 dust load locally in Africa [Engelstaedter and Washington, 2007; Schwanghart and Schütt, 189 2008; Sunnu et al., 2008; Lau et al., 2009b; Wilcox et al., 2010], this is the first study that 190 addresses how the movement of the ITCZ has implications for the amount of dust that 191 extends over the Atlantic and reaches the Americas. Specifically we examine how changes in 192 the ITCZ effect wind and precipitation patterns over both the North Africa and the Tropical 193 North Atlantic and in turn control the amount of mineral dust that reaches Barbados. 194

195 1.4 Statement of Problem

This work addresses the question of what controls the annual transport of mineral dust. 196 Here we hypothesize that the position of the ITCZ over West African controls the quantity 197 of mineral dust reaching the Caribbean and North America. To test this hypothesis we ask 198 and answer four major questions. (1) How does the movement of the ITCZ over West Africa 199 impact mineral dust emission and transport in winter and summer? (2) How do global 200 climate models recreate the ITCZ over West Africa over the 20th century? (3) Do global 201 climate models predict major changes to the ITCZ over West Africa in the 21st century and 202 how would this impact mineral dust emission and transport? (4) Can we use geochemical 203 tracers of mineral dust source on transported mineral dust samples to confirm our ITCZ 204 over West Africa hypothesis? To answer these questions we utilize multiple data-sources 205 including in situ observations, climate reanalysis products, satellite observations and a novel 206 geochemical analysis of mineral dust samples taken from five continents. 207

²⁰⁸ 1.5 Approach

²⁰⁹ 1.5.1 Quantification of the ITCZ and the Center of Action Ap ²¹⁰ proach

Before quantitative analysis involving the position of the ITCZ can be made, first we must 211 develop an index to track the movement of the ITCZ. To accomplish this we utilize the 212 "Center of Action" approach. Centers of Action (COA) refer to seasonal large-scale sys-213 tems with occur annually in nearly the same geographic regions [Rossby, 1939; Angell and 214 Korshover, 1974, 1982; Hurrell, 1995; Kapala et al., 1998; Mächel et al., 1998]. In the mid-215 latitudes examples of such systems include the Azores High and the Icelandic Low. Over 216 West Africa, the ITCZ can be thought of as a "center of action" as annually it appears in 217 nearly the same position, moving with solar heating over the course of the year. The Center 218

of Action approach identifies where the center of mass of the semi-permanent action center
is position, yielding a latitude and longitude position. Similarly, an intensity of the center is
calculated. We can then relate the positions of the Center of Action to geophysical quantities
[Christoforou and Hameed, 1997; Croke et al., 1999; Piontkovski and Hameed, 2002; Hameed
and Piontkovski, 2004; Riemer et al., 2006; Kolker and Hameed, 2007; Bakalian et al., 2007;
Doherty et al., 2008; Jordi and Hameed, 2009; Doherty et al., 2012a; Taylor et al., 2012], in
this case mineral dust, to establish relationships.

1.5.2 Application: Relating the ITCZ to mineral dust and atmo spheric variables

Using the COA approach and reanalysis data, latitude, longitude and intensity of the ITCZ 228 over West Africa can be calculated from 1948 to the present. We compare values of latitude, 229 longitude and intensity of the ITCZ over West Africa to surface based measurements of 230 mineral dust at Barbados [Prospero and Carlson, 1972] and satellite measurements of mineral 231 dust to note how changes in the position of the ITCZ over West Africa changes mineral dust 232 loads. It is desirable and necessary to physically explain observed relationships between the 233 ITCZ over West Africa and mineral dust. To do this we examine winds over West Africa 234 and Tropical Atlantic in reanalysis products, as well as precipitation estimates, vegetation 235 and drought indices to assess changes in aridity. 236

²³⁷ 1.5.3 Application: GCM hindcast of ITCZ in 20th century

Global climate models (GCM) are now ubiquitous in atmospheric science. In order to use GCM models to make future predictions, first the models must show they have skill to recreate past climate. Here we compare GCM output from the Climate Model Intercomparison Project 3 (CMIP3) to multiple reanalysis datasets to quantify the errors from GCM using the newly defined ITCZ index as a metric. Differences from reanalysis are calculated for ²⁴³ spatial fields and temporal trends of the ITCZ over West Africa.

²⁴⁴ 1.5.4 Application: GCM predictions of ITCZ in 21st century

Next the climate models are evaluated in the 21st century. Deviations from the 20th century are noted, and trends in the 21st century are calculated. Changes in the positions of the ITCZ over West Africa in the 21st century are used to estimate changes in mineral dust emission and transport.

²⁴⁹ 1.5.5 Geochemical Analysis

Changes in precipitation and wind patterns driven by the ITCZ over West Africa lead to key 250 mineral dust hot-spots over West Africa becoming more or less active. Each such hot-spot 251 has a unique geochemical fingerprint, a combination of mineralogy, elemental composition 252 and isotopic ratios. Here we attempt to use these fingerprints on both samples acquired close 253 to mineral dust source regions and filter samples of unknown sources to differential source 254 regions. To accomplish this samples representative of source regions are acquired from four 255 continents, and sensitive analysis techniques are developed to be applied trace quantities of 256 dust. 257

Source regions important to mineral dust emissions have unique geological histories, for 258 example the Bodele Depression is a dried lakebed. In turn this geological history dictates 259 the mineral composition of the surface soils which once eroded are lifted and transported by 260 the atmosphere. The extent of prevalence of which a mineral or a group of minerals appear 261 in a given location can be used as a proxy [Formenti et al., 2011]. Elemental ratios also vary 262 from location to location, for example the Ca to Al ratio has been used to apportion aerosol 263 provenance in the Southeastern United States [Perry et al., 1997]. Lastly the isotopic ratios 264 of Pb, Nd, Sr can be used to determine the source of mineral dust and other crustal materials 265 [Grousset and Biscaye, 2005]. 266

267 1.5.6 Application: Apportionment of Unknown Samples in the 268 Southeastern United States

As we seek to use such fingerprints to detect changes in source regions, we must fully char-269 acterize source regions. To this end samples have been acquired and analyzed from four 270 continents; Europe, Asia, Australia and Africa. Additionally to test our hypothesis sam-271 ples of unknown provenance were acquired from Pensacola, FL and are analyzed for two 272 purposes; first to demonstrate if we can clearly separate continental sources and secondly 273 for samples with African sources if changes in the position of the ITCZ over West Africa 274 drives is reflected in the mineralogy, elemental composition or isotopic ratios of the samples. 275 Geochemical analysis produces a large quantity of data, signals in which may not be clearly 276 evident. For example, the elemental composition of mineral dust in this analysis are calcu-277 lated for 26 elements. To parse signals out of large datasets we apply principle component 278 analysis. Using a simple framework identified from the mineralogy, we construct a framework 279 by which continental source apportionment can easily be made. 280

²⁸¹ 1.6 Unique Contributions of the Work

This dissertation consists of two complimentary components advancing our understanding of 282 the controls of mineral dust transport by combining atmospheric sciences and geochemistry. 283 (1) A synergistic data analysis component: Central to this is the development of a new 284 climate index quantifying the movement and intensity of the ITCZ. This new climate index 285 is then applied in three novel ways: to explain dust transport mechanisms where the new 286 framework proves to be superior in comparison to the ones previously used; to assess skill of 287 GCM in the 20th century; and to predict future trends in the ITCZ in the 21st century. (2) 288 A lab component developing geochemical techniques, which allow for the differentiation of 289 mineral dust sources. Geochemical tracers are then applied to unknown samples taken from 290 the Southeastern United States. In sum, this work produces a new conceptual model for 291

explaining year-to-year variability of mineral dust concentration and a novel multidisciplinary
approach to apportioning mineral dust sources.

²⁹⁴ 1.7 Dissertation Outline

The structure of the dissertation is as follows. The role of the ITCZ over West Africa in 295 controlling mineral dust transport in winter is explored in Chapter 1. These results are 296 presented as published in *Doherty et al.* [2012a]. Chapter 2 presents the results of a similar 297 experiment, here we asses the role of the ITCZ over West African in controlling mineral 298 dust transport in summer. These results are presented as a manuscript in preparation for 290 submission [Doherty et al., 2012b]. The ability of global climate models to accurately capture 300 the position of the ITCZ over West Africa is assessed and presented in Chapter 3. Model 301 trends and projections for the ITCZ over West Africa and the implications for mineral dust 302 transport in the coming century are presented in Chapter 4. The geochemical analysis and 303 framework for continental apportionment are presented in Chapter 5. Chapter 5 is presented 304 here as a dissertation chapter but is anticipated to be published with co-authors A. Beck, 305 J.K. Cochran and E.T. Rasbury. The dissertation is concluded in Chapter 6 by summarizing 306 the results, discussing the results in context of previous work, the implications of this study 307 and possible continuation of this work in future studies. 308



Figure 1.1: Number of publications related to keyword search "Saharan Mineral Dust" in Google Scholar by year, 1965 to present. After *Kaufman et al.* [2005].

³⁰⁹ Chapter 2

³¹⁰ Control of Saharan Mineral Dust ³¹¹ Transport to Barbados in Winter by ³¹² the Intertropical Convergence Zone ³¹³ over West Africa

$_{_{314}}$ 2.1 Introduction

The largest source of mineral dust globally is the Saharan desert [Prospero et al., 2002; 315 Washington et al., 2003]. Dust from this region is transported long distances over the North 316 Atlantic Ocean [Darwin, 1846; Prospero et al., 1970] and plays an important role in the 317 climate system [Arimoto, 2001]. It exerts a direct radiative forcing on climate as it interacts 318 with solar and longwave radiation [Sokolik and Toon, 1996; Tegen et al., 1996]. Moreover, 319 dust particles play an important role for various aspects of the microphysics of both mixed-320 phase clouds [DeMott et al., 2003] and warm clouds [Levin et al., 2005] and hence are an 321 important contributor to the aerosol indirect effect. Saharan dust has been linked with 322 the frequency and intensity of Atlantic hurricanes [Dunion and Velden, 2004; Evan et al., 323

³²⁴ 2006; *Lau and Kim*, 2007b,a] and has been found to fertilize ocean waters with iron and ³²⁵ other micronutrients [*Jickells*, 1999; *Baker et al.*, 2006], to promote algal blooms [*Walsh* ³²⁶ *and Steidinger*, 2001], and to provide phosphate and potassium to the Amazon Basin [*Swap* ³²⁷ *et al.*, 1992]. The long-range transport of mineral dust from the Sahara across the Atlantic is ³²⁸ significant enough to be responsible for the formation of the soils of many Caribbean islands ³²⁹ and the Amazon Basin [*Herwitz et al.*, 1996; *Muhs et al.*, 2007].

This long-range transport of mineral dust is subject to considerable inter-annual and 330 intra-annual variability due to a number of physical factors. Winds over the Sahara and 331 Sahel control emissions of mineral dust [Washington et al., 2003; Washington and Todd, 332 2005, and winds downstream of the emission region affect subsequent transport [Chiapello 333 et al., 1995; Riemer et al., 2006; Doherty et al., 2008]. Precipitation and vegetation in the 334 Sahel has been suggested as a key factor in dust emission processes [Evan et al., 2006a]. 335 Precipitation over the source region can modulate dust emission, and precipitation over the 336 transport region can remove dust from the atmosphere [Tegen and Fung, 1994; Zender et al., 337 2003]. 338

The variability of mineral dust transport from Africa across the Atlantic is documented 339 in the long-term mineral dust record measured at Barbados. Dust concentrations have been 340 measured and published at this locations from 1965 through 2003 and have been studied 341 extensively by the community [Rydell and Prospero, 1972; Prospero and Nees, 1977; Glaccum 342 and Prospero, 1980; Prospero and Carlson, 1980; Prospero and Nees, 1986; Savoie et al., 1987; 343 Moulin et al., 1997; Chiapello et al., 1999; Prospero and Lamb, 2003; Ginoux et al., 2004; 344 Chiapello et al., 2005; Prospero et al., 2008; Trapp et al., 2010]. Several studies focused 345 on explaining this variability by linking the observed dust load with climate indices. For 346 example, a qualitative link relating mineral dust in Barbados and an El Niño-Southern 347 Oscillation (ENSO) index has been suggested by Prospero and Lamb [2003]. Moulin et al. 348 [1997] demonstrated a statistical relationship between mean annual dust load at Barbados 349 and the mean value of the North Atlantic Oscillation (NAO) index, while Chiapello and 350

Moulin [2002] linked variability in satellite observations of dust over the tropical North Atlantic Ocean off the coast of Africa to variability of the NAO during winter. *Ginoux et al.* [2004] showed a weak statistical relationship between the winter NAO index and the observed surface dust at Barbados as well as in simulated mineral dust concentration in the region. *Doherty et al.* [2008] demonstrated an improvement upon the relationship of NAO and mineral dust by showing an even stronger relationship between the location of the Azores High in the Atlantic and the Hawaiian High in the Pacific and dust in the Caribbean.

In this work we present a new perspective on mineral dust transport in the region by 358 focusing on the role of the Intertropical Convergence Zone (ITCZ) over Africa rather than 359 on global teleconections indices as has previously been done. Here we consider the ITCZ to 360 represent the zone of near-surface convergence over continental West Africa that is sometimes 361 referred to as the West African monsoon trough [Sultan et al., 2003]. Year-to-year and 362 decade-to-decade variability of the ITCZ has been observed over Africa and the Atlantic 363 [Mächel et al., 1998; Kapala et al., 1998], with differences in the intensity and location noted. 364 Some variability of the ITCZ can likely be ascribed to changes in sea surface temperature 365 [Folland et al., 1986; Hoerling et al., 2006] as precipitation in the Sahel has been linked to 366 sea surface temperature changes [Folland et al., 1986; Giannini et al., 2003]. Tomas et al. 367 [1999] present theoretical evidence for the location of the ITCZ also being a function of the 368 cross-equatorial pressure gradient. 369

It has been previously noted that a relationship between the ITCZ and mineral dust 370 likely exists. The tradewind circulation associated with the ITCZ blows through the area 371 downwind of the Sahara and Sahel where mineral dust loads are the highest *Prospero and* 372 Carlson, 1972; Moulin et al., 1997]. Prospero and Carlson [1972] first showed in a field 373 experiment that the dust belt was located directly to the north of the ITCZ. Moulin et al. 374 [1997] and later Evan et al. [2006b] showed using satellite observations of mineral dust that 375 the dust belt moves north blowing dust into the Caribbean ahead of the ITCZ in summer, 376 and returns south blowing dust into South America in winter. Precipitation in the tropical 377

North Atlantic basin and surrounding locations have been shown to be dependent on the
location of the ITCZ [Kapala et al., 1998] and multiple studies have linked mineral dust in
Barbados with precipitation in the Sahel region of Africa [Prospero and Nees, 1986; Chiapello
et al., 2005].

Although work has been done linking the movement of the ITCZ to changes in dust 382 load locally in Africa [Engelstaedter and Washington, 2007; Schwanghart and Schütt, 2008; 383 Sunnu et al., 2008; Lau et al., 2009b; Wilcox et al., 2010, this is the first study that addresses 384 how the movement of the ITCZ has implications for the amount of dust that extends over 385 the Atlantic and reaches the Americas. Specifically we examine how changes in the ITCZ 386 effect wind and precipitation patterns over both the North Africa and the Tropical North 387 Atlantic and in turn control the amount of mineral dust that reaches Barbados. We focus 388 in this paper on the winter season to compare to more well known teleconnections such as 380 the NAO and ENSO. Although less dust reaches Barbados in winter than does in summer 390 [Kaufman et al., 2005], dust continues to reach Barbados in winter in appreciable quantities 391 as Barbados lies on the northern edge of this dust plume. The dustier summer season will 392 be the focus of a forthcoming paper. 393

The structure of this paper is as follows. In Section 2.2 we describe how we quantify the variability of the ITCZ. We will introduce the various data sets used in this study in Section 2.3. In Section 2.4 we will then explore the long-term relationship between mineral dust in Barbados and the ITCZ, and we will compare the physical mechanisms by which the ITCZ impact the mineral dust concentrations in Barbados. Section 2.5 concludes our findings.

³⁹⁹ 2.2 The ITCZ over West Africa as a Center of Action

Year-to-year and decade-to-decade [*Mächel et al.*, 1998; *Kapala et al.*, 1998] variability of the
ITCZ has been observed, with differences in the intensity and location noted. To compare
to the record of mineral dust at Barbados, a time series of the position and intensity of the

ITCZ is necessary. Here we develop such a time series by utilizing the "Center of Action"
(COA) approach. This provides us with three indices, one for the latitudinal position, one
for the longitudinal position, and one for the intensity of the system.

The concept of an atmospheric "Centers of Action" (COA) was first suggested by Rossby 406 [1939] and used by others since [Angell and Korshover, 1974, 1982; Hurrell, 1995; Kapala 407 et al., 1998; Mächel et al., 1998]. A COA is a seasonal large-scale system which occurs 408 annually in nearly the same geographic region. Examples of such COA are the Azores High 409 COA or the Icelandic Low COA. While each of these COA have a well established and well 410 understood long-term mean location, on a monthly or seasonal basis there is a large amount 411 of variability in its exact location. As each of these COA are often the major climatic feature 412 of a given region for a given season, this short-term location variability can be extremely 413 important in terms of dictating the weather for a given season. Similarly each COA can vary 414 in terms of its strength or intensity from month to month or season to season. 415

Once the position and intensity is quantified by the COA indices, those indices can be 416 related to geophysical variables to investigate their relationships. Such an approach in the 417 mid-latitudes has been successfully applied to many geophysical systems, e.g. solar cycle 418 variability [Christoforou and Hameed, 1997], cloud cover and global temperature [Croke 419 et al., 1999], zooplankton in the Gulf of Maine [Piontkovski and Hameed, 2002], location of 420 the Gulf Stream northwall [Hameed and Piontkovski, 2004], mineral dust transport [Riemer 421 et al., 2006; Doherty et al., 2008], variability in sea level [Kolker and Hameed, 2007], frequency 422 of Greenland tip jet events [Bakalian et al., 2007] and sea-surface temperatures in the Gulf 423 of Lion [Jordi and Hameed, 2009]. As we will describe shortly, in this paper we apply this 424 framework to the ITCZ over West Africa. 425

In the literature there is some ambiguity regarding the definition of the term ITCZ [*Nicholson*, 2009]. The term ITCZ has been applied to both the tropical rain belt over West Africa [*Sultan et al.*, 2003] and the region of near-surface convergence, sometimes referred to at the Inter Tropical Front (ITF). As explained below, we define our index of convergence to ⁴³⁰ be the center of mass of the low level (925 hPa) convergence as described in NCEP Reanalysis
⁴³¹ over West Africa. We refer to this as the West Africa Convergence Zone (WACZ) to avoid
⁴³² any confusion with terminology associated with the ITCZ in the literature. Defined in this
⁴³³ way, the WACZ is observed to be north of the African Easterly Jet as described in *Mohr*⁴³⁴ and Thorncroft [2006], consistent with the conceptual model presented in *Nicholson* [2009].
⁴³⁵ It is shown to be associated with large scale changes in circulation and precipitation both
⁴³⁶ locally in West Africa and regionally over the Tropical Atlantic.

While pressure has previously been used as the field to quantify the COA [Hameed and 437 *Piontkovski*, 2004; *Riemer et al.*, 2006; *Doherty et al.*, 2008], in the tropics the gradient of 438 sea level pressure is very small, and so we follow after Kapala et al. [1998] and use divergence 439 as the reference field. Specifically, we define the WACZ to be the center of mass of the 440 low level (925 hPa) convergence as described in NCEP Reanalysis [Kalnay et al., 1996] over 441 West Africa (15° S to 20° N and 20° W to 20° E). Figure 2.1 shows the climatological 442 mean of divergence at 925 hPa from NCEP Reanalysis from 1965-2003 during the boreal 443 winter including the months December to March. The three WACZ COA indices identify 444 the position and intensity of the center of mass of the band of convergence extending between 445 5° N and 10° N from the West African coast east across the continent. 446

The intensity index W_i is defined as an area-weighted divergence departure from a threshold value over the domain (I, J)

449

$$W_{i} = \frac{\sum_{i,j=1}^{I,J} (D_{i,j} - D_{t}) \cos \phi_{i,j} (-1)^{M} \delta_{i,j}}{\sum_{i,j=1}^{I,J} \cos \phi_{i,j} \delta_{i,j}},$$
(2.1)

where $D_{i,j}$ is the divergence value at 925 hPa at a grid point (i, j), D_t is the threshold divergence value $(D_t = -1.8 \times 10^{-6} \text{ s}^{-1})$, $\phi_{i,j}$ is the latitude of grid point (i, j), M = 0 for divergent systems and 1 for convergent systems, $\delta = 1$ if $(-1)^M (D_{i,j} - D_t) > 0$, and $\delta = 0$ divergent $(D_{i,j} - D_t) < 0$. 454 The latitudinal index W_{ϕ} is defined as:

455

$$W_{\phi} = \frac{\sum_{i,j=1}^{I,J} (D_{i,j} - D_{t}) \phi_{i,j} \cos \phi_{i,j} (-1)^{M} \delta_{i,j}}{\sum_{i,j=1}^{I,J} (D_{i,j} - D_{t}) \cos \phi_{i,j} (-1)^{M} \delta_{i,j}}.$$
(2.2)

The longitudinal index W_{λ} is defined analogously. The location indices thus give divergenceweighted mean latitudinal and longitudinal positions of the WACZ.

т **т**

As a result we obtain time series of monthly values for latitude (W_{ϕ}) , longitude (W_{λ}) and 458 intensity (W_i) that characterize the WACZ. The indices are available from January 1948 to 459 the present. Results were not significantly sensitive to changes in box size, the box location 460 or the divergence threshold. This work focuses on the seasonal variability of mineral dust 461 and the WACZ in boreal winter, and to represent this we use the average of four months 462 December (Y - 1), January (Y), February (Y) and March (Y) where Y represents the year 463 of the winter season. Our choice of DJFM is motivated by the work of Ben-Ami et al. [2011] 464 who find that dust transport of Africa enters its southerly or winter mode from the end of 465 November through the end of March. Values of the WACZ presented in this work are for 466 the period December 1965 to March 2003, concurrent with the mineral dust record at the 467 surface. 468

$_{469}$ 2.3 Datasets

In this study we utilize the mass concentration of mineral dust as recorded in Barbados. 470 Barbados is an ideal location for observing mineral dust as it is the first land mass encoun-471 tered by mineral dust traveling over the North Atlantic from Africa. Mineral dust at the 472 surface at Barbados has been the subject of previous studies *Prospero et al.*, 1970; Carlson 473 and Prospero, 1972; Prospero and Nees, 1986; Moulin et al., 1997; Prospero and Lamb, 2003; 474 Ginoux et al., 2004; Chiapello et al., 2005] and is the longest continuous record of surface 475 mineral dust concentrations. The record of monthly mineral dust at Barbados is available 476 from August 1965 through December 2003. Data is collected near the surface at Barbados, 477

 $_{478}$ located at 13° 10' N and 50° 30' W.

We use NCEP/NCAR Reanalysis wind data from NOAA/ESRL PSD [Kalnay et al., 1996] to calculate the WACZ COA indices. Data is available from January 1948 to present, although we only consider the period of 1965–2003 to be consistent with the mineral dust record at Barbados. We compared the WACZ COA index from NCEP/NCAR Reanalysis to one calculated from ERA-40 (not shown), and for the winter season we found the two were not statistically different. NCEP/NCAR Reanalysis winds were also used to compute composite images, as described in Section 2.4.2.

Two traditional teleconnection climate indices are used in this study for comparison purposes, the NAO and ENSO. Monthly values of the NAO are from the Climate Prediction Center of NCEP/NOAA (http://www.cpc.ncep.noaa.gov). ENSO monthly values are also from the Climate Predictions Center of NCEP/NOAA who computes the ENSO indices from NOAA OI SST product over multiple regions in the Pacific.

The GPCP Dataset (v 2.1) [*Adler et al.*, 2003] is used for precipitation data in this study. Data is available from January 1979 - April 2008, however data after 2003 is not used in this study to be consistent with the mineral dust record at Barbados.

We use TOMS Aerosol Index (AI, v 8.0) as a measure of aerosol load [Herman et al., 1997]. Data is available for the period 1979-1993 and 1996-2005. As described in Kiss et al. [2007] some calibration and instrument errors exist for this instrument. To this end we do not use the AI in any quantitative way, but present it as a qualitative aid in understanding how dust load changes with variation in the WACZ.

499 2.4 Results

⁵⁰⁰ 2.4.1 Climatology of West Africa Convergence Zone

As shown in Figure 2.2 the WACZ (as defined in Section 2.2) displays a clear seasonal latitudinal migration from its southernmost point in January to its northernmost point in August. The latitudinal index W_{ϕ} is thus consistent with the conventional understanding that the WACZ's location is seasonal and that the WACZ moves to follow the solar heating [Folland et al., 1991]. The index W_{ϕ} reaches its northernmost position over Africa in July and August, trailing the movement of the solar heating maxima by one to two months. *Nicholson* [2009] notes that August also represents the period of maximum rainfall in the Sahel region and the period of maximum variability associated with the WACZ.

The movement of the WACZ is not a simple north-to-south migration, but rather is a twodimensional migration. W_{λ} reaches its most westward point in the spring and moves towards the east in the fall. Coupled with the annual latitudinal migration, this means the WACZ moves on a NW to SE axis from spring/summer into fall/winter. Maximal convergence occurs in the spring and decreases to a minimum in fall, and is thus more intense in its north and west mode than its south and east mode.

While the long-term mean of the indices shows a clear annual cycle with an obvious 515 seasonal migration, both the inter- and intra-annual variability of the WACZ is large. In 516 the summer, when W_{ϕ} reaches its northernmost maximum there are large differences in the 517 latitudinal position from year to year, ranging from 16 to 19 degrees north. In semi-arid 518 locations such as the Sahel, a large shift in the location of the WACZ can be the difference 519 between flooding and drought, and in turn can greatly effect soil conditions and ultimately 520 dust mobilization. Additionally the latitudinal intra-annual variability plays a major role 521 in controlling regional climate. Some years the WACZ remained in a northward phase for 522 multiple consecutive months, leading to an extended rainy season for the Sahel, while in 523 other years the northward phase and hence the rainy season is very short. The duration of 524 the northward rainy phase may be critical in determining soil characteristics for the semi-arid 525 Sahel, particularly in the summer when strong solar insolation can quickly dry out soils. 526

⁵²⁷ 2.4.2 Relationship of Mineral Dust at Barbados and WACZ and ⁵²⁸ ENSO during winter

To establish a relationship we correlated mean winter (DJFM) dust load at Barbados with 529 the mean winter (DJFM) WACZ COA indices $(W_{\phi}, W_{\lambda} \text{ and } W_{i})$. Winter season mineral 530 dust load at Barbados is significantly correlated with the location of the WACZ in terms of 531 both the latitude (W_{ϕ}) and longitude (W_{λ}) of the WACZ at an α of at least 0.01 as shown in 532 Table 2.1. No statistical relationship between the intensity of the WACZ (W_i) and mineral 533 dust at Barbados was observed. Since the W_{ϕ} index increases in value as the WACZ moves 534 north, the negative correlation observed between dust and W_{ϕ} suggests that as the WACZ 535 moves south dust load at Barbados increases. Likewise the positive correlation between dust 536 and W_{λ} suggests that as the WACZ moves eastward dust load at Barbados increases. 537

Traditionally a Pearson product-moment correlation coefficient is used to represent co-538 variance between two datasets. The Pearson product-moment correlation coefficient is not 539 robust as it requires a normal distribution for both datasets nor resistant to outliers [Wilks, 540 1995]. Both the seasonally mean DJFM mineral dust at Barbados and ENSO climate in-541 dices used in this work are not normally distributed, and in the case of the ENSO dataset 542 include multiple outliers. To reduce the impacts of outliers between datasets we use the 543 Spearman rank correlation as shown in *Wilks* [1995]. It should be noted that using either 544 the Pearson or the Spearman correlation coefficients the correlations between dust and W_{ϕ} , 545 W_{λ} and E_{1+2} are statistically significant, but the Spearman correlation coefficient gives a 546 more representative reflection of the covariance between datasets. 547

The indices W_{ϕ} and W_{λ} are not independent of one another as shown in Table 2.2. The WACZ is located further east in winters when it is in its south phase, and further west in winters when it is in its north phase, suggesting that in the winter season the WACZ varies along a NW to SE axis. This NW to SE axis is clearly visible in Figure 2.3a where during seasons in which the WACZ is north (blue diamonds) it also tends to be west and vice versa. Hence the NW location correlates to a reduction of mineral dust in Barbados and the SE location correlates to an increase in mineral dust at Barbados. Figure 2.3b shows the magnitude of change in divergence at 925 hPa that accompanies the NW to SE shift, and a clear dipole pattern emerges. To produce this figure we subtracted the seasonal winter mean (DJFM) of divergence of the northernmost WACZ seasons (75th percentile) from the seasonal winter mean (DJFM) of the southernmost WACZ seasons (25th percentile). The black contour indicates a significant difference at 10%. The box over which the WACZ COA is computed is shown in black.

Figure 2.4 shows the temporal relationship between W_{ϕ} and mineral dust load at Barbados. Mineral dust load at Barbados reached its lowest value during the mid-1960's when W_{ϕ} is found to be at its most northward location, and it reached its peak in the mid-1980's when W_{ϕ} was in its most southward location.

Also shown in Table 2.1 is that the south and east migration of the WACZ explains more 565 of the variability of the mineral dust than traditional climate indices such as the NAO or 566 ENSO do. The ENSO Region 1+2 (E_{1+2}) is located along the coastline of Ecuador and Peru, 567 ENSO 3 (E_3) is located just to the north and west of ENSO 1+2 spanning the central Pacific, 568 and ENSO 4 (E_4) is the westernmost box approaching Papua New Guinea. We observe that 569 the largest correlation between mineral dust and ENSO occurs for the ENSO index closest to 570 South America (E_{1+2}) , with the correlation coefficient attenuating moving into the Central 571 Pacific. The relationship between ENSO and mineral dust is statistically significant at 5%572 close to the coast of South America (E_{1+2}) and not significant for the eastern Pacific (E_3) , 573 central Pacific $(E_{3,4})$ and central-west Pacific (E_4) . This is consistent with Mahowald et al. 574 [2003] who did not find widespread correlations in and around West Africa between dust and 575 $E_{3.4}$ using a combination of modeling results and station observations. Our results suggest 576 that statistical correlations between mineral dust transport and ENSO are sensitive to choice 577 of ENSO domain. 578

As E_{1+2} and W_{ϕ} are not independent (r = 0.80), we apply a partial correlation analysis to account for their covariance and assess the relationship between each and mineral dust

as if they were independent [*Wilks*, 1995]. There is precedent for using a partial correlation 581 analysis to remove an ENSO signal. Sankar-Rao et al. [1996] use partial correlation to 582 evaluate the signal of the Indian Monsoon on Eurasian seasonal snowfall without the influence 583 of ENSO. More recently partial correlation was used similarly in a paper on mineral dust, 584 tropical cyclone activity and ENSO [Evan et al., 2006]. If we evaluate the rank correlation 585 between mineral dust and W_{ϕ} with E_{1+2} held constant, the correlation coefficient decreases 586 only slightly from r = -0.69 to r = -0.66, remaining significant at α of <0.001. The rank 587 correlation between mineral dust and E_{1+2} with W_{ϕ} held constant decreases from r = 0.30588 to r = 0.06, which is statistically insignificant. Much of the statistical relationship between 580 E_{1+2} and mineral dust is a mathematical artifact caused from E_{1+2} varying in time with 590 W_{ϕ} . We confirmed this in a separate composite analysis and find no physical relationship 591 between E_{1+2} and wind or precipitation over emission, transport and deposition regions (not 592 shown). 593

To corroborate our findings on the statistical relationships of Barbados mineral dust and 594 the WACZ, we present the appropriate composite figures of the TOMS Aerosol Index (AI) 595 data from NIMBUS-7 (1979-1993) and Earth Probe (1996-2005) as a semi-quantitative proxy 596 for dust load over the entire Tropical North Atlantic. We analyze AI data to assess if the 597 relationship between the WACZ and mineral dust at Barbados is reflective of larger scale 598 changes in mineral dust load over the North Atlantic and Caribbean. AI measures absorbing 599 particles in the atmosphere by comparing observed UV measurements to an idealized UV 600 profile of the atmosphere. TOMS AI is not able to differentiate between types of absorb-601 ing aerosols, in particular particles from biomass burning register similarly to mineral dust 602 aerosols. During boreal winter biomass burning is at a maximum [Formenti et al., 2008] in 603 North Africa, and a mix of mineral dust and biomass burning aerosols are likely. TOMS 604 AI has known biases due to the height of the aerosol layer [Herman et al., 1997; Mahowald 605 and Dufresne, 2004, calibration drift errors [Kiss et al., 2007] and cloud contamination is-606 sues [Torres et al., 1998, 2002]. Here we only seek to verify the relationships of mineral 607

dust surface observation and WACZ index, and so a qualitative use of the TOMS data is appropriate.

We use composites to represent seasonal conditions in which an index is in a positive or 610 negative phase. The index W_{ϕ} is ranked in ascending order, the lowest quartile (q25) of data 611 is identified as the negative phase and the highest quartile (q75) of data is identified as the 612 positive phase. The years used to compute the seasonal composites are shown in Table 2.3. 613 For W_{ϕ} the negative phase (q25) corresponds to periods in which the WACZ is in a south 614 mode, and the positive phase (q75) corresponds to periods in which the WACZ is in a north 615 mode. All seasons of AI data that have been selected as negative (q25) or positive (q75)616 phases are then averaged together to form a composite. 617

Figure 2.5 shows composites of TOMS AI with respect to the northward (minimum dust 618 in Barbados) and southward phase (maximum dust at Barbados) of W_{ϕ} . We restrict our 619 analysis to the period 1979-1993, 1996-2003 to included all AI data that is concurrent with 620 the dust record at Barbados. Barbados is observed to be in a plume of dust entering the 621 Caribbean in the southward phase of the WACZ (Figure 2.5a). When the WACZ is south 622 (Fig. 2.5a), the greatest values of AI occur between 15° and 20° N, from the Atlantic 623 Coastline east to 20° E. A very large plume of mineral dust is also evident over the Tropical 624 North Atlantic centered between 15° and 20° N, which extends into the Caribbean Sea. The 625 largest AI values of greater than AI = 3.0 occur near the Bodele Depression, a known hot-626 spot for mineral dust emission in Africa [Washington et al., 2003]. Along the Gulf of Guinea 627 values of AI are modest, near AI = 1.0 and likely reflect a mix of mineral dust and biomass 628 burning aerosols. 629

In contrast, when W_{ϕ} is in its northward phase, the highest values of AI occur along the Gulf of Guinea, extending westward into the Tropical North Atlantic, centered at about 7.5° N. While some of this plume may be the result of biomass burning, *Formenti et al.* [2008] found that more than 70% of this material is mineral dust. A peak in AI (AI= 2.5) is observed near the Bodele Depression. Over much of the Sahara values of AI are modest, 635 coming in around 1.0.

⁶³⁶ Comparing the north and south phases of the WACZ, a clear dipole in AI is evident in ⁶³⁷ the composite difference image (Fig. 2.5c), similar to the dipole pattern in the composite ⁶³⁸ difference for divergence as shown in Figure 2.3b. As the WACZ moves southward a large ⁶³⁹ increase in AI is seen over the Sahara Desert, with increasing value of AI extending over ⁶⁴⁰ a broad geographical area northward to the Mediterranean and eastward to the Red Sea, ⁶⁴¹ suggesting a robust response in dust emission to southward movement of the WACZ.

⁶⁴² Higher values of AI are observed across nearly all of the Tropical North Atlantic north of
⁶⁴³ 10° N suggesting a large increase in westward transport of mineral dust. Increases in AI are
⁶⁴⁴ visible into the Caribbean. This analysis supports the conclusion drawn from the observed
⁶⁴⁵ correlations, that mineral dust at the surface at Barbados increases as the WACZ moves
⁶⁴⁶ southward.

Over Africa south of the Sahel and in particular south of 12° N a decrease in AI is observed as the WACZ moves southward. It is not clear if this is due to a decrease in mineral dust transport into this region or a reduction in biomass burning aerosols as TOMS AI cannot differentiate between mineral dust and biomass burning aerosols. This reduction in AI over this region is not likely due to precipitation as DJFM is the dry season in the Sahel, where precipitation is very low.

Our results are similar to those of Engelstaedter and Washington [2007] who find a spatial 653 correlation between 10 m-divergence in ERA-40 and TOMS-AI over much of North Africa. 654 Our results differ from the findings of *Ben-Ami et al.* [2011] who argue that during the boreal 655 winter season that the dust plume across the Atlantic is constrained to a band centered on 656 4° N. However when the WACZ is in its north phase our results support the finding of Ben-657 Ami et al. [2011] as maximum aerosol load does appears to be near 4° N. We find that the 658 location of the dust belt in TOMS-AI measurements is sensitive to the latitudinal position 659 of the WACZ during the boreal winter, shifting northward to include Saharan sources when 660 the WACZ is south phase. We find a plume of most likely mixed dust-biomass smoke is 661

found near to 4° N only when the WACZ is in a north phase. It should be noted that the studies focus on different time periods. This analysis includes years over the period 1979-1993/1996-2004, whereas the Ben-Ami data is from the 21st century only.

665 2.4.3 Physical Mechanisms for Observed Correlation

The strong correlation between W_{ϕ} and mineral dust at Barbados and mineral dust at Barbados must be explained physically. Next we will examine how changes in W_{ϕ} impact wind and precipitation in the region, which in turn control the emission and transport of mineral dust from the Sahara.

Strong northeast winds over the Sahel have been shown to increase mineral dust emissions 670 [Engelstaedter and Washington, 2007; Schwanghart and Schütt, 2008; Doherty et al., 2008]. 671 Figure 2.6 shows the composite difference in winds for the 25th percentile W_{ϕ} (southern 672 most) winter (DJFM) seasons minus the 75th percentile W_{ϕ} (northern most) winter (DJFM) 673 seasons over the period December 1965 - March 2003. This represents the difference in 674 flow from the southern mode in which mineral dust is maximized at Barbados and the 675 northern mode in which such dust is minimized. Figure 2.6a shows a strengthening of near-676 surface northeasterly winds during these conditions. This strengthening occurs over much 677 of the Sahel and the Eastern Sahara, which are important source regions for mineral dust 678 [Washington and Todd, 2005; Engelstaedter and Washington, 2007]. We hypothesize that 679 the location of these strengthening winds is important in increasing dust emissions. 680

Northeastern winds in this region are typical in winter [Engelstaedter and Washington, 2007]. These winds known as the Harmattan winds transport mineral dust from the Sahara and Sahel regions southwards into sub-Saharan Africa in winter. The Harmattan winds are effective at emitting dust not only because of their strength but because of where they occur. The area around Lake Chad is a major source of mineral dust emissions [Washington and Todd, 2005] and just downwind of this region the largest increase in northeastern flow (Figure 2.6a and b) is observed as the WACZ changes phase. Ben-Ami et al. [2011] suggest that the Bodele Depression, which lies just north and east of Lake Chad, is the primary source of mineral dust during the boreal winter, where dust emissions are highly dependent on near-surface wind speeds [*Koren and Kaufman*, 2004]. Again near to the Bodele Depression we note increases in NE winds as the WACZ changes phase.

Our results support the hypothesis of *Schwanghart and Schütt* [2008] who noted the role of Harmattan winds which blow over the Bodele Depression, emitting large quantities of mineral dust in the boreal winter. In their study *Schwanghart and Schütt* [2008] observed that the strength of NE winds at 850 and 925 hPa near the Bodele was tightly correlated to the amount of mineral dust emitted in winter. *Schwanghart and Schütt* [2008] suggested that northward movement of the ITCZ had the capacity to diminish the NE winds. Our results here confirm their hypothesis.

In summary the strength of the NE Harmattan winds are an important control on the amount of mineral dust that is emitted in the boreal winter. We hypothesize that shifts in the WACZ controls dust emission not only by increasing wind speeds over much of West Africa, but does so by focusing these increases in critical locations such as the Sahel, Lake Chad and the Bodele Depression. Similar near-surface wind increases are observed from the composites for W_{λ} (not shown).

Wind increases in the Sahel, Lake Chad and Bodele Depression, known dust hot spots, 705 are a means of relating changes in position of the W_{ϕ} to a third key physical parameter, 706 soil texture. Mineral dust originating in dried lake beds (e.g. areas surrounding Lake Chad, 707 Bodele Depression) are composed of vastly different minerals than that of sand (e.g. Sahara 708 Desert) [Cole et al., 2009]. Such lake bed derived minerals are known to be more easily 709 emitted as windborne mineral dust [Koren and Kaufman, 2004]. In turn incorporation 710 of soil texture databases (such as http://daac.ornl.gov/cgi-bin/dataset_lister_new. 711 pl?p=19) in climate models is known to be of importance to accurately predict mineral dust 712 emission. As the location of wind augmentation is shown to be critical in this work, we 713 successfully link W_{ϕ} to soil texture. 714

Steering winds aloft at the base of the Saharan Air Layer show an increase in east to 715 west flow, increasing trans-Atlantic transport of emitted particles (Figure 2.6b). The height 716 of the Saharan Air Layer depends on the season [Carlson and Prospero, 1972], and satellite 717 observations suggest that mineral dust transport is constrained to heights below 700 hPa in 718 boreal winter over the Tropical North Atlantic [Ben-Ami et al., 2009]. Figure 2.6b both show 719 an increase in trans-Atlantic transport, although this increase is not statistically significant. 720 Figure 2.7a shows the winter (DJFM) mean precipitation for years in which the WACZ 721 is in its south phase. Maximum precipitation rates are observed south of the equator, with 722 the highest values observed in Brazil south of the Amazon River. In Africa precipitation is 723 highest in Central Africa, south of the equator. Moderate precipitation is observed between 724 the equator and 9° N over West Africa. No precipitation occurs over the Sahel or Sahara, as 725 winter is the dry season. Differences in precipitation between the south phase conditions of 726 the WACZ (Figure 2.7a) and the north phase conditions are shown in Figure 2.7a. Composite 727 differences are calculated in such a way to show conditions that lead to maximized dust load 728 at Barbados. We do not see a shift in the location of precipitation as the phase of W_{ϕ} 729 changes, but rather a large scale drying across the Central Africa, the equatorial Atlantic 730 and the Amazon Basin. 731

Precipitation over mineral dust source regions is not affected by changes in W_{ϕ} . No change in the amount of precipitation over the Sahel or Sahara regions occurs as W_{ϕ} changes phase from south to north, as no precipitation falls in this region during either phase. Wet deposition, vegetation, soil moisture in source regions are therefore not related to W_{ϕ} in winter.

In contrast to the Sahel and Sahara, over the tropical North Atlantic Ocean precipitation is non-zero. Mineral dust reaching Barbados and the Caribbean encounters precipitation in these regions as it is transported. Figure 2.7b suggests that dust maximization at Barbados is consistent with drier conditions in the tropical North Atlantic region, however. Although dust transport to the Americas could be increased in seasons when W_{ϕ} is in its south phase

as less precipitation and therefore less wet deposition occurs, the observed change in pre-742 cipitation here is too small to support this conclusion. Based on changes in precipitation in 743 source and transit regions we conclude that changes in precipitation driven by variability W_{ϕ} 744 are not important for mineral dust emission and transport to Barbados. Interestingly, Figure 745 2.7 shows significant precipitation occurring during the winter season across the equatorial 746 Atlantic into South America, suggesting the wet deposition may be an important removal 747 process in this region. As the W_{ϕ} changes to the southern phase, significant drying is ob-748 served across the equatorial Atlantic into South America (Figure 2.7b). While this is not 749 relevant for the dust transport processes that are the focus of this paper, we hypothesize that 750 this drying would reduce wet deposition of mineral dust that is transported to the Amazon 751 Basin during winter from the Sahara and Sahel by way of the equatorial Atlantic (Figure 752 2.5). 753

Table 2.1 shows the near-zero correlation coefficient between intensity of WACZ (W_i) and 754 mineral dust at Barbados (r = 0.13). This finding may seem surprising and counterintuitive, 755 hence deserves further analysis. For this "null case" we produced composite images as was 756 done for the latitude index W_{ϕ} . The weak (q25 of W_i) and intense (q75 of W_i) years used 757 in these composite images are listed in Table 2.4. Here we note that since the correlation 758 coefficient between W_i and dust at Barbados is near zero, the direction of the difference has 759 no physical meaning. This is in contrast to the composites based on W_{ϕ} , which were built 760 to highlight conditions that lead to dust maximization at Barbados. 761

The differences in the wind field at both 925 hPa (Figure 2.8a) and 850 hPa (Figure 2.8b) are not statistically significant between intense and weak WACZ seasons over source regions of the Sahel or Sahara. Over the Tropical North Atlantic a few patches of increased east to west flow are observed, suggesting a slight increase in transport when the WACZ is weak compared to the strong phase. These patches of increased flow do not appear to be large enough in scale to increase mineral dust transport to Barbados. We attribute a lack of change in wind over source regions as an explanation for why the intensity of the WACZ is ⁷⁶⁹ not correlated with mineral dust load at Barbados.

As no precipitation falls in the source regions of the Sahel and Sahara and winter, the 770 strength of the WACZ does not impact precipitation in these regions. Mineral dust emission 771 is thus not impacted via precipitation as a function of WACZ intensity. Over the Tropical 772 North Atlantic a general increase in precipitation is observed in years in which the WACZ 773 is weak, with significant changes observed near to the African coastline. Mineral dust being 774 transported to the Caribbean and Barbados could encounter more precipitation, and thus be 775 impacted by wet deposition. Large changes in precipitation are observed over the equatorial 776 Atlantic and Amazon Basin. Here precipitation is increased in the weak WACZ phase, 777 potentially increasing removal of mineral dust via wet deposition. 778

To summarize we observe no changes in near surface flow or precipitation over the source 779 regions of the Sahel and Sahara as the WACZ changes phase from weak to strong. Hence no 780 physical mechanisms for increased dust emission are tied to changes in the WACZ intensity. 781 We do observe some changes in the transport region of the tropical North Atlantic Ocean. 782 specifically favorable winds for transport are increased while removal by precipitation also 783 increases in the weak phase of the WACZ. We conclude that (1) processes in the source 784 region are more important than processes in the transport region for mineral dust transport 785 to Barbados and the Caribbean and (2) increased transport via enhanced advection is com-786 pensated for by increased removal in the weak phase resulting in no significant change in 787 transport. 788

In summary, we note that increases in dust at Barbados during the winter season are associated with the WACZ moving south and that the underlying physical process is a change in winds in particular over the source region, not a change in precipitation. This is in agreement with the findings of *Sunnu et al.* [2008] who performed a field experiment and observed increased concentrations of mineral dust at the surface in Ghana when the ITCZ was south of the observing station in boreal winter.

795 2.5 Conclusions

The relationship between convergence over West Africa, referred to as the ITCZ or West Africa Monsoon, and mineral dust in the Tropical North Atlantic has long been discussed, and in this paper we quantified the relationship for the winter season. We constructed three climate indices that quantify the variability of the convergence over West Africa by applying the Centers of Action approach to wind divergence at 925 hPa from NCEP Reanalysis. We demonstrated the utility of such indices by relating them to the quantity of mineral dust reaching Barbados over the period from 1965 to 2003.

Southward and eastward movement of the WACZ is associated with an increase of the 803 quantity of mineral dust reaching the surface at Barbados. This increase can be explained by 804 increases in near-surface NE Harmattan winds over the Sahel. The location of the increase 805 in wind is critical with increases in NE flow observed near the key Lake Chad and Bodele 806 Depression region. Combined this leads to increases in emission of mineral dust when the 807 WACZ is in its south phase. Once emitted, mineral dust aerosols encounter an increase in 808 the east-to-west trade wind flow over across much of the Saharan Air Layer over West Africa. 800 This increase in advective flow could increase mineral dust transport towards the Americas. 810 The latitude of the WACZ does not change dust emission or transport in winter by 811 precipitation-driven effects. No changes in precipitation over the source regions of the Sahel 812 or Sahara are noted, nor are significant changes observed in the transport zone between West 813 Africa and Barbados. Although not related to transport of mineral dust to Barbados, we 814 note a distinct drying over much of Central Africa, the tropical Atlantic and parts of South 815 America associated with southward movement of the WACZ. 816

No relationship between the intensity of the WACZ and mineral dust was observed. We note that wind strength and direction does not change over key source regions between intense and weak phases of the WACZ. We hypothesize that this lack of change in winds explains why no relationship is observed between intensity of WACZ and mineral dust at Barbados. Although not related to transport of mineral dust to Barbados, weakening of the WACZ over Africa is shown to lead to drying over the equatorial Atlantic and Amazon
Basin.

Studies looking for links between naturally varying systems and the atmosphere often refer to familiar indices such as for example the NAO or ENSO. We show that the location and intensity of the WACZ is a strong alternative to other common indices, with clear links to changes in circulation in the region.

Table 2.1: Spearman rank correlation coefficients between mean DJFM mineral dust at Barbados and climate indices for DJFM season 1965 to 2003. \diamond ,† and * represent significance at <0.001, 1% and 5% respectively.

	W_{ϕ}	W_{λ}	$W_{\rm i}$	NAO	E_{1+2}	E_3	E_4	$E_{3.4}$
Dust at Barbados	-0.69°	0.51^{\dagger}	-0.13	0.18	0.30^{*}	0.18	0.11	0.15

Table 2.2: Cross correlation Spearman rank coefficient between mean winter (DJFM) WACZ COA indices for DJFM season 1965 to 2003. The \diamond represents statistical significance at $\alpha < 0.001$.

	W_{ϕ}	W_{λ}	$W_{\rm i}$
W_{ϕ}	-	-0.73^{\diamond}	-0.08
W_{λ}	-0.73^{\diamond}	-	0.06
$W_{\rm i}$	-0.08	0.06	-

Table 2.3: Years that were used to generate seasonal mean composite images of AI, wind and precipitation based on W_{ϕ} . Each season mean was calculated using the average of four individual months of data; December (Y - 1), January (Y), February (Y) and March (Y)where Y represents the year of the winter season.

	q25			q75		
AI	1983,	1998,	1989,	1984,	1985,	1986,
	1992, 2000, 2001			1988, 1991		
Wind	1977,	1983,	1989,	1966,	1967,	1968,
	1992,	1993,	1995,	1969,	1973,	1974,
	1998,	1999,	2000,	1979,	1986,	1991,
	2001			1996		
Precipitation	1983,	1989,	1992,	1982,	1984,	1986,
	1998, 2000, 2001			1988,1991,1996		

Table 2.4: Years that were used to generate seasonal mean composite images wind and precipitation based on W_i . Each season mean was calculated using the average of four individual months of data; December (Y - 1), January (Y), February (Y) and March (Y) where Y represents the year of the winter season.

	q25			q75			
Wind	1996,	1990,	1995,	1969,	1967,	2003,	
	1968,	1985,	1981,	1966,	1999,	2000,	
	1984,	1989,	1997,	2002,	1979,	1998,	
	1970			1992			
Precipitation	1996,	1990,	1995,	2003,	1999,	2000,	
	1985,1981,1984			2002, 1998, 1992			



Figure 2.1: Mean winter (DJFM) divergence at 925 hPa as computed from NCEP Reanalysis winds. The box over which the WACZ COA is computed is shown in black.



Figure 2.2: The long-term mean (1965-2003) of the WACZ latitude (W_{ϕ}) (blue line, left axis), longitude (W_{λ}) (green line, left axis) and intensity (W_i) (red line, right axis). Error bars represent the standard deviation based on the monthly average for each index.



Figure 2.3: (a) Location of seasonal winter (DJFM) mean of WACZ COA over the period 1965 to 2003. Blue dots represent the northern (75th percentile) phase of WACZ COA, red dots represent southern (25th percentile) phase of WACZ COA. Black dots represent the middle 50 percent seasons of WACZ COA. (b) Difference in divergence of NCEP Reanalysis at 925 hPa during winter (DJFM) over the period 1965 to 2003. Differences are calculated by subtracting the seasonal winter mean (DJFM) of the northern most WACZ seasons (75th percentile) from the seasonal winter mean (DJFM) of southern most WACZ seasons (25th percentile). The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. Black contour represent a significant difference at 10%. The box over which the WACZ COA is computed is shown in black.



Figure 2.4: Comparison of the position of the mean DJFM WACZ latitude (left axis) and mean DJFM dust load at Barbados (right axis, inverted) over the period 1965 to 2003. Seasonal averages are calculated only for seasons that contain concentration data for all months of the season. Both time series are averaged over the winter months of December-March.





Figure 2.5: (a) Composite mean of TOMS AI for the southernmost winter (DJFM) seasons (25th percentile) of the WACZ. (b) As in (a) but for the northernmost WACZ latitude (75th percentile) winter (DJFM) seasons. Seasons with missing AI data are excluded from the composite. (c) The difference (a) minus (b), i.e. conditions of the WACZ latitude index for which dust load is maximized at Barbados. All composites are calculated over the period 1979-93; 1996-2003. Asterisks represent the location of Barbados.



Figure 2.6: Difference in composite mean of NCEP Reanalysis winds at (a) 925 hPa and (b) 850 hPa. Differences are calculated by subtracting the seasonal winter mean (DJFM) of the northern most WACZ seasons (75th percentile) from the seasonal winter mean (DJFM) of southern most WACZ seasons (25th percentile). The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. All composites are calculated over the period 1965-2003; Blue contour represent a significant difference at 10%. Asterisks represent the location of Barbados.



Figure 2.7: (a) Composite mean of GPCP precipitation for the southernmost winter (DJFM) seasons (25th percentile) of the WACZ over the period 1979 to 2003. (b) Difference in composite mean of GPCP precipitation over the period 1979 to 2003. Differences are calculated by subtracting the seasonal winter mean (DJFM) of the northern most WACZ seasons (75th percentile) from the seasonal winter mean (DJFM) of southern most WACZ seasons (25th percentile). The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. Black contour represent a significant difference at 10%. Units are mm day⁻¹ from GPCP. Asterisks represent the location of Barbados.


Figure 2.8: Difference in composite mean of NCEP Reanalysis winds at (a) 925 hPa and (b) 850 hPa. Differences are calculated by subtracting the seasonal winter mean (DJFM) of the weakest WACZ seasons (75th percentile) from the seasonal winter mean (DJFM) of strongest WACZ seasons (25th percentile). Wind composites are calculated over the period 1965-2003; Blue contour lines represent a significant difference at 10%. (c) As (a) and (b) but for GPCP precipitation (mm day⁻¹) over the period 1979 to 2003. Black contour lines represent a significant difference at 10%.

²²³ Chapter 3

Role of the Intertropical Convergence Zone Over West Africa in Controlling Saharan Mineral Dust Transport to the Caribbean in the Dusty Season

3.1 Introduction

The majority of mineral dust emitted into the atmosphere originates from the arid Sahara 834 Desert and the surrounding semi-arid Sahel [Prospero et al., 2002; Washington et al., 2003], 835 with the highest concentrations of mineral dust occurring in the summer season of June 836 through September. Understanding the year-to-year variability of this dust load during 837 summer is critical as mineral dust originating from the Sahara is the dominant aerosol type 838 over much of West Africa and the Tropical North Atlantic [Formenti et al., 2008] and has 839 complex impacts on the radiation budget and cloud properties of the region [Arimoto, 2001]. 840 Mineral dust directly impacts the radiation by absorbing and reflecting incoming shortwave 841 radiation and absorbing outgoing longwave radiation [Sokolik and Toon, 1996; Tegen et al., 842

1996]. Like other aerosols, mineral dust indirectly impacts radiation by increasing droplet 843 concentration [Twomey, 1977], and by altering the precipitation efficiency, albedo and life-844 time of rain clouds [Albrecht, 1989]. Mineral dust has been shown to act as condensation 845 nucleus for ice nucleation [Sassen et al., 2003; DeMott et al., 2003], potentially increasing 846 the abundance of cirrus clouds and thereby warming the climate. On the other hand it has 847 been shown that during particularly dusty summers a cooling of the tropical North Atlantic 848 Ocean can occur *Lau and Kim*, 2007b as a result of the sum of direct and indirect effects. 849 Saharan mineral dust has also been linked to tropical cyclone formation over the tropical 850 North Atlantic Dunion and Velden, 2004; Evan et al., 2006; Lau and Kim, 2007b,a; Braun, 851 2010, which is located downwind of the Sahara Desert. Radiative forcing from mineral dust 852 has been shown to shift the location and strength of the tropical rainband over the Atlantic 853 Ocean [Huang et al., 2009] and may impact the ITCZ-monsoon circulation over Africa [Lau 854 et al., 2009a; Kim et al., 2010; Wilcox et al., 2010]. 855

Saharan mineral dust in summer is transported long distances, often traveling at least 5,000 km and persisting in the atmosphere for periods in excess of a week [*Prospero et al.*, 1970] before reaching the Americas. The quantity of mineral dust transported to the Americas is important for many biogeochemical [*Swap et al.*, 1992; *Jickells*, 1999; *Baker et al.*, 2006; *Koren et al.*, 2006] and geological processes [*Herwitz et al.*, 1996; *Muhs et al.*, 2007].

Dust emission in North Africa is paradoxically maximized in the summer season when 861 the annual northward migration of the Intertropical Convergence Zone (ITCZ) results in 862 the maximum annual rainfall in the semi-arid lands of the Sahel [Moulin et al., 1997; Evan 863 et al., 2006b]. Precipitation reduces dust load in multiple ways [Tegen and Fung, 1994; 864 Zender et al., 2003] both directly and indirectly. Where precipitation occurs concurrently 865 with mineral dust aerosols the dust is removed by wet deposition. Precipitation leads to 866 plant growth [Eklundh and Olsson, 2003; Olson et al., 2005; Herrmann et al., 2005], which 867 in turn increases root activity, reduces soil mobility, improves soil quality and ultimately 868 decreases dust emission [Nicholson et al., 1998]. Hence, increased summer dust load over 869

⁸⁷⁰ West Africa occurs in spite of increases in precipitation, not due to it.

Amongst the atmospheric conditions responsible for maximum dust concentrations during 871 summer are African Easterly Waves, the existence of the Saharan Air Layer [Karyampudi 872 et al., 1999] and increased easterly winds. African Easterly Waves produce strong gusty 873 winds near the surface as they move from east to west over West Africa, resulting in dust 874 emission [Jones et al., 2003]. The Saharan Air Layer (SAL) has long been known to be a key 875 factor in increasing stability and thus allowing mineral dust to travel long distances to reach 876 the Americas [Carlson and Prospero, 1972; Prospero and Carlson, 1972]. In the summer the 877 SAL exhibits its greatest vertical extent and is more prevalent. Lastly, north of the ITCZ, 878 easterly winds increase in the summer months resulting in a higher probability of emitted 879 dust reaching the Americas. 880

While the long-term average quantity of mineral dust emitted in Africa and transported 881 over the tropical North Atlantic Ocean is at its peak in summer, this quantity varies greatly 882 from year to year. Much effort has gone into improving our understanding of the processes 883 that are responsible for this variability. Observations of Saharan mineral dust over the 884 Atlantic Ocean and at Barbados have been related to variability in the North Atlantic Oscil-885 lation (NAO) [Moulin et al., 1997], particularly in the winter season [Chiapello and Moulin, 886 2002]. Similar relationships have been diagnosed from global chemistry and transport model 887 results [Ginoux et al., 2004]. Prospero and Lamb [2003] related mineral dust in Barbados 888 and an El Niño-Southern Oscillation (ENSO) index. Dust transport to the Americas has 889 been linked to the position of the Azores High in winter [*Riemer et al.*, 2006] and in summer 890 to the position of the Azores High and Hawaiian High [Doherty et al., 2008]. Schwanghart 891 and Schütt [2008] found no connection between regional weather systems and dust emission 892 in summer, instead suggesting that dust emission in West Africa is connected to regional 893 convection. More recently the variability of dust concentration at Barbados during winter 894 has been attributed to changes in emissions driven by changes in the latitude of the ITCZ 895 over West Africa [Doherty et al., 2012a]. 896

In this paper we show that the ITCZ is an atmospheric center of action directly related 897 to all key processes affecting mineral dust emission and transport during summer and that 898 the variability of mineral dust concentration in the atmosphere can be attributed to a large 899 extent to the variability in the ITCZ. The ITCZ is a global belt of inclement weather that 900 generally occurs where the trade winds converge and form the ascending branch of the Hadley 901 Circulation. Annually the ITCZ moves north and south following the progression of annual 902 solar maximum [Folland et al., 1991]. Over the ocean the ITCZ circulation is relatively 903 simple whereas over land the structure of the ITCZ becomes more complicated. When the 904 ITCZ occurs over land it can be referred to as a monsoon. For example over Africa parts 905 of the ITCZ are referred to as the West Africa Monsoon (WAM). Much ambiguity exists 906 in terminology regarding the ITCZ [Nicholson, 2009], owing to the increased complexity 907 of the system over land. Over West Africa the annual northward migration of the ITCZ-908 WAM brings critical seasonal rainfall to areas of the Sahel and southern Sahara [Nicholson, 900 2009. As such the location and strength of the ITCZ has implications for seasonal soil and 910 vegetative conditions, and likely dust emission and transport. 911

In this paper we explore how the variability of the ITCZ impacts summer dust emission and transport to the Caribbean. We introduce the data sets used in this study in Section 3.2. In Section 3.3 we describe the quantification of the variability of the ITCZ. In Section 3.4 we demonstrate the long-term relationship between mineral dust in Barbados and the ITCZ during the dusty season, and we compare the physical mechanisms by which the ITCZ impacts the mineral dust concentrations at Barbados. In section 3.5 we conclude our findings.

918 3.2 Datasets

919 3.2.1 Mineral Dust

To quantify the amount of mineral dust in the atmosphere we utilize two datasets; a long term surface based record of dust concentrations at Barbados (1965 - 2003) and a satellite derived measure of optical thickness from the Advance Very High Resolution Radiometer
(AVHRR) instrument over the tropical North Atlantic (1982 - 2010).

The surface record of mineral dust concentrations in Barbados is regarded as the longest 924 continuous observation of aerosols and its record has been extensively studied [Rydell and 925 Prospero, 1972; Prospero and Nees, 1977; Glaccum and Prospero, 1980; Prospero and Carl-926 son, 1980; Prospero and Nees, 1986; Savoie et al., 1987; Moulin et al., 1997; Chiapello et al., 927 1999; Prospero and Lamb, 2003; Ginoux et al., 2004; Chiapello et al., 2005; Prospero et al., 928 2008; Trapp et al., 2010]. Barbados is an ideal location to study mineral dust transport as 929 the island is the easternmost point of the Americas in the Caribbean, and thus the first 930 land encountered by mineral dust that has traversed the North Atlantic Ocean. Barbados is 931 situated approximately 4,800 km west of the western-most point of Africa. Monthly values 932 of mineral dust concentrations in $\mu g m^{-3}$ are available from August 1965 through September 933 2003. In this study, only the years in which all four months (JJAS) of the summer season 934 have data are used in the calculation. 935

To determine how the spatial distribution of mineral dust responds to changes of the 936 location and intensity of the ITCZ we utilize an AVHRR-based dust aerosol optical thickness 937 (AOT) [Evan et al., 2006]. The dataset provides monthly mean AOT with a resolution of 938 $1^{\circ} \times 1^{\circ}$ over the tropical North Atlantic Ocean (0° N to 25° N and 65° W to 10° W). The 939 algorithm used to create the dataset is described fully in Evan et al. [2006] and a general 940 discussion and climatology of the dataset is presented in Evan et al. [2006b]. This dataset 941 was chosen as it has been calibrated for the tropical North Atlantic Ocean to detect optically 942 thick dust over ocean surfaces, was cross-platform calibrating using AERONET, Metosat and 943 TOMS-AI and is of a long record (1982–2010) coincident with much of the Barbados record. 944 The major advantage of this product is that it is able to detect optically thick dust storms 945 (AOT > 2) which many algorithms incorrectly identify as clouds [Evan et al., 2006]. This 946 feature is particularly important in summer (JJAS) when optically thick dust storms are 947 most prevalent. 948

949 3.2.2 Gridded Datasets

In this study we use several gridded reanalysis products to characterize atmospheric and surface conditions. Wind data in this work is taken from NCEP Reanalysis [Kalnay et al., 1996]. NCEP Reanalysis data is available monthly from 1948 to the present, although we constrain our analysis to the period 1965–2010, and has a resolution of $2.5^{\circ} \times 2.5^{\circ}$. Monthly averages of winds at 925 hPa from NCEP Reanalysis are used to calculate divergence, which is needed for the ITCZ Center of Action approach as described in Section 3.3.

To represent precipitation we use Global Precipitation Climatology Project (GPCP) Version 2.1 Combined Precipitation Data Set [Adler et al., 2003]. GPCP precipitation data is available from 1979 to 2010, with a resolution of $2.5^{\circ} \times 2.5^{\circ}$.

The Palmer Drought Severity Index (PDSI) dataset is used to represent soil conditions [*Dai et al.*, 2004]. PDSI data is available from 1870 to 2005, although we constrain our analysis to the period 1965–2003. PDSI has a resolution of $2.5^{\circ} \times 2.5^{\circ}$. A PDSI value of 0 represents the mean state of soil moisture. Negative values represent drought, with values of -3 and -4 representing severe and extreme droughts respectively. Positive values of PDSI represent greater moisture than average soil conditions.

To represent vegetation at the surface the GIMMS Normalized Difference Vegetation 965 Index (NDVI) dataset is used [Tucker et al., 2005]. The NDVI is calculated using irradiance 966 data from the AVHRR instrument carried aboard NOAA satellites. NDVI data is available 967 over the period July 1981–December 2002 at a resolution of $0.25^{\circ} \times 0.25^{\circ}$. Values of NDVI 968 near zero represent surface conditions of barren rock or sand, with increasing values of NDVI 969 representing more lushly vegetated surfaces. For example brush and savannah-like terrains 970 may have an NDVI value of between 0.2 to 0.4, and a tropical rainforest a value between 0.8971 and 1.0. 972

Data was acquired from Physical Sciences Divsion of the Earth System Research Laboratory of NOAA (http://www.esrl.noaa.gov/psd/) with the exception of the AVHRR AOT record which was acquired from the University of Virginia (http://trane.evsc.virginia. edu/Data.html) and GIMMS NDVI which was acquired from the Asia-Pacific Data-Research
Center (http://apdrc.soest.hawaii.edu/) via OPeNDaP.

978 3.3 Methods

⁹⁷⁹ 3.3.1 ITCZ as a Center of Action

Introduced by Rossby [1939] and used by others since [Angell and Korshover, 1974, 1982; Hurrell, 1995] an atmospheric "Center of Action" (COA) refers to semi-permanent atmospheric systems which persist for long periods of time, regularly appear in the same location year after year, and have impacts on regional climate. Some well known COA include the Azores High and Icelandic Low.

Here we treat the ITCZ over West Africa as a COA and seek to quantify its variability. 985 Over West Africa much ambiguity exists regarding the use of the phrases "Intertropical Con-986 vergence Zone" and "monsoon". The system of interest consists of two zones of convergence 987 and uplift, a northern convergence associated with the African Easterly Jet and a southern 988 band of convergence associated with the Tropical Easterly Jet. To highlight the ambiguity of 989 the use of the term ITCZ in the literature we note that Sultan et al. [2003] and Parker et al. 990 [2005] and references therein refer to the southern convergence as the ITCZ, while Nicholson 991 [2009] refer to the northern zone of convergence as the ITCZ. 992

As shown shortly, our approach considers the center of mass of convergence over West Africa and so as to avoid any ambiguity regarding the terms "Intertropical Convergence Zone" or "West African Monsoon" in this work we refer to this band of convergence as the West Africa Convergence Zone (WACZ).

⁹⁹⁷ The Center of Action approach is explained fully in *Doherty et al.* [2012a], but here we will ⁹⁹⁸ briefly review the key components. In the traditional Center of Action approach the center ⁹⁹⁹ of mass of a pressure system is identified and quantified by three indices, its longitudinal ¹⁰⁰⁰ and latitudinal position and its intensity. For each index time series are generated, which characterize the variability of the COA under consideration which then can be used to explain
the variability of a geophysical system of interest. This approach has been successfully
applied globally to explain the variability in many geophysical systems [*Christoforou and Hameed*, 1997; *Croke et al.*, 1999; *Piontkovski and Hameed*, 2002; *Hameed and Piontkovski*,
2004; *Riemer et al.*, 2006; *Doherty et al.*, 2008; *Kolker and Hameed*, 2007; *Bakalian et al.*,
2007; *Jordi and Hameed*, 2009; *Hameed et al.*, 2011].

Here we seek to generate similar time series of location and intensity indices that quantify the variability of the WACZ. In the aforementioned studies pressure has been used as the field to quantify the COA, however in the tropics the gradient of sea level pressure is very small, and so we follow after *Kapala et al.* [1998] and use divergence as the reference field, as described in *Doherty et al.* [2012a].

We define the WACZ by computing convergence at 925 hPa using winds from NCEP Reanalysis [Kalnay et al., 1996] over West Africa (0° N to 25° N and 20° W to 20° E). This domain is outlined in black in Figure 3.1a. Figure 3.1a also shows the climatological mean of JJAS divergence from 1965 to 2003, the center of the WACZ is just north of 18° N across much of West Africa before dipping southward near Lake Chad.

The intensity index of the WACZ W_i is defined as an area-weighted divergence departure from a threshold value over the domain (I, J):

1019
$$W_{i} = \frac{\sum_{i,j=1}^{I,J} (D_{i,j} - D_{t}) \cos \phi_{i,j} (-1)^{M} \delta_{i,j}}{\sum_{i,j=1}^{I,J} \cos \phi_{i,j} \delta_{i,j}},$$
(3.1)

where $D_{i,j}$ is the divergence value at 925 hPa at a grid point (i, j), D_t is the threshold divergence value $(D_t = -5.0 \times 10^{-5} \text{ s}^{-1})$. $\phi_{i,j}$ is the latitude of grid point (i, j). M = 0 for divergent systems and 1 for convergent systems. $\delta = 1$ if $(-1)^M (D_{i,j} - D_t) > 0$ and $\delta = 0$ if $(-1)^M (D_{i,j} - D_t) < 0$. The latitudinal index W_{ϕ} is defined as:

1024

$$W_{\phi} = \frac{\sum_{i,j=1}^{I,J} (D_{i,j} - D_{t}) \phi_{i,j} \cos \phi_{i,j} (-1)^{M} \delta_{i,j}}{\sum_{i,j=1}^{I,J} (D_{i,j} - D_{t}) \cos \phi_{i,j} (-1)^{M} \delta_{i,j}}.$$
(3.2)

52

The longitudinal index W_{λ} is defined analogously. The location indices thus give divergenceweighted mean latitudinal and longitudinal positions of the WACZ. The physical location of the WACZ COA for mean JJAS seasons from 1965 to 2003 is shown in Figure 3.1b.

¹⁰²⁸ 3.3.2 Composite Imaging Analysis

We use composite images to show changes in environmental conditions associated with a 1029 change in phase of W_{ϕ} , W_{λ} or W_{i} . For example to see how changes in the latitudinal position 1030 index W_{ϕ} impact regional wind circulation, the seasonal mean values of W_{ϕ} are ranked in 1031 ascending order. The lowest quartile (q25) of seasonal data is identified as the "south phase", 1032 which corresponds to the seasons when the WACZ was in a southerly position. The highest 1033 quartile (q75) of seasonal data is identified as the "north phase", which corresponds to the 1034 seasons when the WACZ was in a northerly position. All seasons of wind data that have 1035 been selected as southern (q25) or northern (q75) phases are then averaged to form a wind 1036 composite representing wind conditions for the south phase or the north phase, respectively. 1037 This approach can be repeated for other WACZ indices (W_i, W_λ) instead of W_ϕ , and other 1038 environmental variables (e.g. AVHRR AOT, GPCP Precipitation, GIMMS NDVI, PDSI) 1039 instead of wind. Years used to generate the individual composite images are shown in Table 1040 3.3 and Table 3.4. 1041

¹⁰⁴² 3.3.3 Empirical Orthogonal Function Analysis

We use Empirical Orthogonal Function (EOF) analysis to identify the key spatial patterns evident in the variability of environmental data and link the time series of principle components of these key spatial patterns to the time series of the WACZ latitude index. In essence EOF reduces temporally variant gridded data with internal variability to a small number of spatial patterns which represent the maximum possible quantity of the variability of the original data [*Wilks*, 1995]. In this analysis we use singular value decomposition on the covariance matrices of our gridded data to produce eigenvectors. For each pattern identified

by the EOF a spatial map can be generated by multiplying the anomalies of the original data 1050 by the eigenvector, this spatial patterns shows what regions vary in the same or opposing 1051 directions to maximize the variance of the original data. The principle components, or the 1052 time series related to each spatial pattern can be generated by multiplying the anomalies 1053 of the original data by the eigenvalues. Principle component time series can be related to 1054 other physical quantities to determine what outside factors may give rise to observed spatial 1055 patterns in the data. Each mode generated in this way is orthogonal to the others and the 1056 cross-correlations of the principle components are zero. Each mode explains a fraction of the 1057 variance of the original data, the first mode controls the most variance with each subsequent 1058 mode controlling a smaller fraction of the variance of the original data. 1059

1060 3.4 Results

¹⁰⁶¹ 3.4.1 Climatology of WACZ Index During the Dusty Season

Figure 3.2a shows the latitudinal position of the WACZ (W_{ϕ}) during the summer season 1062 and adjacent months. The median value is indicated by a horizontal red bar. Skewness and 1063 spread are shown by the position of the 25th and 75th percentiles indicated by edges of the 1064 central box. Whiskers denote the spread of all non-outlying data with outliers indicated by 1065 red crosses. The WACZ moves steadily northward over West Africa in spring and summer, 1066 reaching its northernmost location on average in August. After September the WACZ shifts 106 rapidly southward. Maximum year-to-year variability in the WACZ latitude index is observed 1068 in autumn. 1069

Figure 3.2b represents mean Sahel precipitation averaged over the domain 15° N to 25° N and 0° to 25° E over the same time period. Precipitation begins as the WACZ moves northward in late spring and summer, and ends as the WACZ begins to move southward in fall. The WACZ latitude index appears to capture the seasonal precipitation cycle well, and our results are consistent with those presented in *Nicholson* [2009].

¹⁰⁷⁵ 3.4.2 Relationship Between Mineral Dust in the Caribbean and ¹⁰⁷⁶ Climatic Indices

1077 Barbados Mineral Dust

Calculating the correlation coefficients between the time series of mineral dust concentration 1078 at Barbados and the three WACZ indices, we find that summer season mineral dust load 1079 is significantly correlated with both the latitudinal and longitudinal location index of the 1080 WACZ at $\alpha = 0.01$ as shown in Table 3.1. Since the WACZ index W_{ϕ} increases with latitude, 1081 the observed negative correlation of W_{ϕ} and mineral dust means that seasonal mineral dust 1082 load increases as the WACZ is displaced southward and seasonal mineral dust load decreases 1083 as the WACZ is displaced northward. Likewise mineral dust increases as the WACZ moves 1084 eastward and decreases as the WACZ moves westward. 1085

The latitude and longitude indices of the WACZ are not independent of one another (Table 3.2). As seen in Figure 3.1b the WACZ is found to be further east when it is in its south phase and further west when it is in its north phase, suggesting that in the summer season the WACZ varies along a NW to SE axis. The NW location correlates to a reduction of mineral dust in Barbados and the SE location correlates to an increase in mineral dust at Barbados.

Figure 3.3 shows the temporal relationship between the latitude index of the WACZ and mineral dust load at Barbados. The scale for mineral dust concentration at Barbados has been inverted for visual ease of comparison. Mineral dust load at Barbados reaches its lowest value during the mid-1960's when the WACZ is found to be at its most northward location. Mineral dust reaches its peak in the mid-1980's when the WACZ reaches its most southward location.

Table 3.1 also includes the correlation coefficients of summer season mineral dust concentration at Barbados and traditional climate indices such as the NAO or ENSO. Only statistically insignificant correlations are observed.

1101 AVHRR AOT

The long-term record at Barbados (1965 to 2003) is of great value as its length allows us to 1102 examine inter-annual variability of dust transport to the Caribbean region. Having estab-1103 lished the relationship between increase in dust concentration at Barbados and a southward 1104 displacement of the WACZ, next we seek to determine if this relationship exists over the 1105 rest of the Caribbean. For greater spatial coverage we utilize the North Atlantic mineral 1106 dust AVHRR aerosol optical thickness product as described in *Evan et al.* [2006], which is 1107 available for the period 1982 to 2010. For winter we utilized TOMS AI data to represent 1108 mineral dust over the Caribbean. Here for the summer season we use the Evan AOT dataset 1109 instead, as instrument drift in later years makes TOMS AI data unreliable [Kiss et al., 2007]. 1110 Seasons in which the WACZ is north in summer are dominated by later years (see Table 1111 3.3), meaning for TOMS the composite difference compares years in which the instrument 1112 drifted (WACZ north, later years) and years in which the instrument did not drift (WACZ 1113 south, early years). In winter TOMS AI is an appropriate choice because north and south 1114 years are evenly distributed, reducing impacts of drift on composite images. Here we use the 1115 Evan AOT instead to ensure difference plots show changes driven by geophysical mechanisms 1116 rather than instrument drift. 1117

Figure 3.4a shows the composite mean in AOT of JJAS seasons in which the WACZ 1118 was in its south phase, as described in Section 3.3.2. A plume originating over Mauritania 1119 is evident, which extends west towards the Americas centered at or just slightly north of 1120 16° N. Figure 3.4b shows the corresponding composite mean of AOT in which the WACZ 1121 was in its north phase. Here we again note a plume forming west of Mauritania, extending 1122 westward towards the Americas centered near to 16° N. Figure 3.4c is the difference in AOT, 1123 subtracting the north phase conditions shown in Figure 3.4b from the south phase conditions 1124 shown in Figure 3.4a representing conditions that lead to dust maximization at Barbados. 1125 Figure 3.4c shows no shift in the location of the dust belt, but rather an increase in AOT over 1126 nearly all of the Tropical North Atlantic. In the center of the dust belt the AOT increases by 1127

0.2 to 0.3 as the WACZ shifts southward. The lack of movement in the dust belt; rather an 1128 increase during seasons in which the WACZ moves southward is confirmed by the first mode 1129 EOF analysis (Figure 3.4d). At all points the relative magnitude of the first EOF mode 1130 (which explains 36% of the total variance in the AOT record) are negative, suggesting that 1131 the AOT responds unidirectionally, either increasing or decreasing over the Tropical North 1132 Atlantic. The principle component of this first EOF is statistically significantly correlated 1133 with the WACZ latitude at $\alpha \leq 0.01$ (r = 0.51), suggesting that southward movement of 1134 the WACZ is indeed coupled with the basin-wide increase in mineral dust. 1135

The observed relationship of a southward movement of the WACZ and increases both in dust concentration at the surface at Barbados and in the wide-spread mineral dust observed in the AVHRR record suggests a robust link between the position of the WACZ and mineral dust reaching the Caribbean. In the remainder of this section we will investigate the physical mechanisms by which the WACZ impacts mineral dust emission on the one hand and transport to the Caribbean on the other hand.

In particular we will analyze how changes in the WACZ impact winds near the surface, precipitation, soil conditions and vegetation over West Africa, which in turn impact mineral dust emission. We will also analyze winds aloft and precipitation over the North Atlantic to see how changes in the WACZ may impact transport of mineral dust across the Atlantic.

1146 3.4.3 Mechanisms Impacting Emission

¹¹⁴⁷ Winds and Precipitation in Source Regions

Figure 3.5 shows the difference in 925 hPa winds between conditions when the WACZ is in the south mode and conditions when the WACZ is in the north mode. These differences represent conditions that lead to dust maximization at Barbados. An increase in the strength of the trade winds across the Sahel is evident, including a maximum strengthening near Lake Chad which is a major source for mineral dust aerosols [*Washington et al.*, 2006]. Winds aloft (not shown) over West Africa show a similar increase in NE to SW flow leading to an ¹¹⁵⁴ increase of export of emitted mineral dust particles.

Figure 3.6a shows the JJAS mean composite of precipitation for the northern phase of 1155 the WACZ. Maximum precipitation is observed in a band stretching from East Africa west 1156 across the Atlantic into South America. However when the WACZ moves southward, so does 1157 the band of precipitation, and a general drying across Africa is evident as seen in Figure 1158 3.6b. This figure shows the difference in precipitation between conditions when the WACZ 1159 is in the south mode (not shown) and conditions when the WACZ is in the north mode 1160 (Figure 3.6a), i.e. conditions which lead to dust maximization at Barbados. Significant 1161 drying occurs over all of Africa between 10° N and 20° N. Just north of the equator a slight 1162 increase in precipitation is noted, related to the southward shift of the WACZ. We note that 1163 the strongest drying occurs over the semi-arid Sahel region. Comparison with Figure 3.5 1164 shows that the area of reduced precipitation is coincident with the area of increased winds, 1165 resulting in an ideal scenario favoring dust emission. 1166

EOF analysis suggests that the north-to-south movement of the precipitation belt rep-1167 resents the majority of variance in the GPCP precipitation product for the summer season 1168 (JJAS). Figure 3.7a shows the first mode (which represents 42% of the total variance) and 1169 Figure 3.7b the second mode (12% of variance). The first mode shows a clear north-south 1170 dipole in precipitation, the second mode features a NE to SW dipole. Both the first and 1171 second modes are significantly correlated with the WACZ latitude index at or below an $\alpha =$ 1172 0.01. Regressing the first and second mode of precipitation together onto the WACZ latitude 1173 index results in a correlation of r = 0.66 (Figure 3.7c). The WACZ latitude index appears 1174 tightly related to the major modes of variability in precipitation over West Africa. 1175

While a widespread reduction in precipitation in the sensitive Sahel region is clearly observed, it is not immediately clear how exactly this reduction in precipitation leads to changes in mineral dust load. Reduced precipitation can reduce wet deposition of mineral dust. In addition reduced precipitation can lead to reductions in soil moisture and vegetation, both of which can allow for increased emission of mineral dust. Hence we next examine ¹¹⁸¹ changes in vegetation and soil moisture tied to latitudinal shifts in the movement of the¹¹⁸² WACZ.

¹¹⁸³ Vegetation and Soil Conditions in the Source Region

GIMMS NDVI is used as a high-resolution proxy for surface vegetation. Reductions in 1184 vegetation are observed in the Sahel in Figure 3.8 which is the composite difference in NDVI 1185 for the south phase of the WACZ minus the north phase of the WACZ which show changes 1186 that lead to dust maximization in Barbados. Vegetation is reduced in a band between 12° N 1187 and 18° N extending from the Atlantic Ocean to the Red Sea, suggesting a robust response 1188 in vegetation due to shifts in the latitudinal position of the WACZ. Figure 3.9a and b show 1189 the first and second modes of the EOF analysis on NDVI for JJAS summer seasons. Mode 1190 1 (39% of the variance) shows a clear north to south dipole, in which vegetation increases 1193 in the Sahel while decreasing to the south, or vice-versa. Mode 2 (12%) of the variance) is 1192 related to variance in the vegetation of the very southern extent of the Sahara Desert. The 1193 principle components of both mode 1 and mode 2 are significantly correlated with the WACZ 1194 latitude index at $\alpha = 0.01$. Regressing the first and second modes of NDVI together onto 1195 the WACZ latitude index results in a correlation of r = 0.68 (Figure 3.9c), i.e. changes in 1196 vegetation are directly related to the latitudinal position of the WACZ. 1197

PDSI is used to represent soil moisture conditions. Figure 3.10a shows the composite 1198 of PDSI during JJAS seasons in which the WACZ is in its south mode, and Figure 3.10b 1190 shows the composite of the north mode. Both north and south phases show drought-like 1200 conditions, reflecting the extreme drought that occurred from the 1970's through the early 1201 1990's in sub-Saharan Africa. However the south phase shows more extreme drying, with 1202 much of the Sahel experiencing values of -3 to -4 PDSI which represent severe to extreme 1203 drought. Subtracting the composite shown in Figure 3.10(b) from the one in Figure 3.10(a)1204 we see over much of the Sahel and southern Sahara desert that drying of this region occurs 1205 concurrently with dust maximization, as the WACZ moves into its south phase. We are not 1206

¹²⁰⁷ able to present an EOF analysis of the PDSI due to many missing data points.

In summary we note that southward movement of the WACZ leads to increases in nearsurface flow in key emission locations. These key emission locations also receive significantly reduced precipitation in seasons in which the WACZ is in its south phase, reducing wet deposition removal of mineral dust in these regions. Reductions in vegetation and soil moisture also follow the southern migration of the WACZ. Thus we hypothesize that southern displacement of the WACZ is critical in increasing mineral dust emission by degradation of the surface soils and increased opportunities for emission via increases in near surface wind.

1215 3.4.4 Mechanisms Impacting Transport

Mineral dust emitted over Africa travels nearly 5,000 km to reach Barbados and the Caribbean. Steering winds over the tropical North Atlantic control where the dust is transported once emitted, and stronger winds result in quicker transport to the Americas reducing gravitational settling and the chance of removal via wet deposition. Next we examine how changes in the latitudinal position of the WACZ impact steering currents aloft and precipitation over the tropical North Atlantic.

Figure 3.11a (850 hPa), 3.11b (700 hPa) and 3.11c (500 hPa) show composite differences 1222 of the wind conditions aloft, which are calculated in the same manner as for Figure 3.5 to 1223 represent conditions in which dust is maximized at Barbados. While just south of the dust 1224 belt (around 16° N) there is significant strengthening of easterly winds between the equator 1225 and 10° N, in the center of the dust belt we see no significant change in transport winds. The 1226 spatial mismatch of the wind augmentation and the location of maximum dust concentration 1227 suggests that the impacts of the movement of the WACZ on circulation are more important 1228 via the emission process. As shown in Section 3.4.3 and Figure 3.5, increases in dust emission 1229 can be linked to increased winds in the important source regions of the Sahel and southern 1230 Saharan desert. 1231

¹²³² Precipitation changes that occur as the WACZ shifts from the south phase to the north

phase, representing conditions in which mineral dust levels are maximized at Barbados, 1233 are shown in Figure 3.11d. Between 15° N and 25° N a drying of the region is noted 1234 with statistical significance over limited areas. Between the equator and 15° N we note an 1235 increase in precipitation, with statistical significance over limited areas. However in contrast 1236 to the extreme drying of the Sahel region (Figure 3.6b) as the WACZ shifts south, no clear 1237 statistically significant pattern emerges over the tropical North Atlantic. We conclude that 1238 precipitation processes in the transport region play a much smaller role in modulating dust 1239 transport to Barbados than does the extreme drying noted over source regions. 1240

¹²⁴¹ 3.4.5 Physical Mechanisms for the Null Case

Table 3.1 shows a near-zero correlation between the intensity of the WACZ (W_i) and mineral dust at Barbados, which means that there is no observed relationship between the WACZ intensity and dust load at Barbados from 1965 to 2003. This may come as a surprising result, and in comparing this "null case" to the WACZ latitude case discussed in Section 3.4.2, we look for differences between the two cases to further elucidate which processes are important for dust emission and transport and which processes are irrelevant.

For the W_i case we again build composite images of wind, precipitation and AVHRR aerosol load for changes in phase of the WACZ intensity to identify reasons which may explain why W_i has apparently no role to play in governing the variability of mineral dust. As the correlation coefficient between W_i and mineral dust is near zero, the choice in direction of difference in composites is trivial.

Figure 3.12a shows the composite difference of 925 hPa winds based on W_i . Here we see a west flow in the Sahel west and south of Lake Chad, which represents a decrease in the near-surface easterly winds. This suggests a reduction in surface dust emission when the WACZ is in a weaker phase. A strong, statistically significant drying occurs over the Sahel and south Sahara (Figure 3.12c), suggesting that a weaker WACZ is associated with decreased precipitation in this region. A similar drying occurs for the W_{ϕ} case (Figure 3.6), however as W_i and W_{ϕ} are not significantly correlated (Table 3.2) these drying patterns are physically independent of each other. In summary over West Africa we see reduced near-surface winds combined with decreased precipitation as the WACZ enters its weaker phase. These changes have opposing effects on dust emission as reduced winds reduce dust emission whereas reduced precipitation increases dust emission. This is consistent with the low correlation coefficient between dust and W_i .

Figure 3.12b shows the composite difference of 850 hPa winds based on $W_{\rm i}$. Wind vectors 1265 point west to east over the mineral dust source regions of the Sahel and Sahara, resulting in 1266 a decrease in easterly winds in these locations. Over the tropical North Atlantic an increase 1267 in east to west flow is observed between the equator and 12° N. Changes in precipitation 1268 over the North Atlantic Ocean are mixed (Figure 3.12c), with decreases in precipitation 1269 near to Africa and increases in precipitation observed over Barbados southward into South 1270 America. This suggests a varied wet depositional change, with reductions near to Africa 1271 and increases near to the Caribbean. The implications for transport to the Americas are 1272 mixed. Reduced precipitation near Africa could increase the lifetime of mineral dust, and 1273 increases in cross-Atlantic flow near the equator could increase transport. However increases 1274 in precipitation over the western tropical North Atlantic could remove mineral dust. The 1275 AVHRR AOT composite difference of the W_i case is shown in Figure 3.12d. An increase in 1276 mineral dust is observed over the North Atlantic, however this increase is constrained only 1277 near to Africa. This response is small compared to the response to changes in W_{ϕ} (Figure 1278 3.4). 1279

From this it becomes clear that changes in intensity of the WACZ do result in changes in wind and precipitation, but for a given phase of intensity one augments, and the other decreases mineral dust emission and transport. This interplay explains the lack of a robust mineral dust response in the Caribbean to changes in WACZ intensity. Table 3.5 summarizes and contrasts the case of W_{ϕ} in the south phase (left column) in which dust is increased at Barbados and the null case of W_{i} in the weak phase (right column). In both cases reduced

precipitation over the source region and steering winds over the tropical North Atlantic act 1286 in a way that would increase dust load in Barbados. In both cases there is no clear signal 1287 from precipitation over the tropical North Atlantic. However in the W_{ϕ} case winds over the 1288 source regions respond in a way that would increase dust emission, whereas the opposite 1289 applies in the W_i case. Our interpretation is that the reduction in surface winds over the 1290 Sahel and Sahara is a key reason that mineral dust load does not increase in the W_i case, 1291 and conversely the increase in surface winds is a key reason that dust load increases in the 1292 W_{ϕ} case. 1293

1294 3.5 Conclusions

While drought conditions in the Sahel, favorable transport circulations, and increased near-1295 surface winds have long been known to increase dust transport to the Americas, no unified 1296 mechanism had hither been identified which could explain changes in key variables. Here we 1297 present the utility of the WACZ as a center of action, which proves capable of explaining the 1298 inter-annual seasonal variability of mineral dust transport to Barbados and to the Caribbean 1299 in summer and of separating out the relative importance of transport and emission. We 1300 found that the latitudinal shift of the WACZ controls much of the variability of mineral dust 1301 over the Caribbean region, and that processes in the source regions over Africa are more 1302 important than processes over transport regions. 1303

The WACZ latitude index is significantly correlated with the mineral dust concentration at Barbados, precipitation in the Sahel, soil moisture and vegetation. Through composite imaging we showed that the southward movement of the WACZ is associated with marked increases in near-surface winds over important dust emitting regions and also with decreases in precipitation over the Sahel, which in turn reduces soil moisture and vegetation. EOF analysis of precipitation and vegetation show that the dominant principle components are statistically significantly correlated to the WACZ latitude. EOF loading patterns resemble ¹³¹¹ composite images, showing the robustness of the WACZ latitude index signal.

In contrast to the WACZ latitude index the intensity index is not significantly correlated with mineral dust at Barbados. The corresponding composite analysis reveals that changes in the WACZ intensity do result in characteristic changes in precipication patterns and wind patterns. However it turns out that increased winds over source regions conincide with increased precipitation and vice versa, hence the two processes counteract each other in their impact on mineral dust in the atmosphere.

In summary, the coupling of changes in near-surface winds with changes in precipitation in emission regions driven by the latitudinal movement of the ITCZ is key in explaining the variability of mineral dust over the tropical North Atlantic, Barbados, and the Caribbean.

Table 3.1: Pearson product moment correlation coefficients between mean JJAS mineral dust at Barbados and climatic indices for JJAS season 1965 to 2003. \diamond represent significance at < 0.1%. W_{ϕ} is the WACZ latitude index, W_{λ} is the WACZ longitude index, W_{i} is the WACZ intensity index, E_{1+2} is ENSO over the ENSO 1+2 box, E_{3} is ENSO over the ENSO 3 box, E_{4} is ENSO over the ENSO 4 box and $E_{3.4}$ is ENSO over the ENSO 3.4 box.

	W_{ϕ}	W_{λ}	$W_{\rm i}$	NAO	E_{1+2}	E_3	E_4	$E_{3.4}$
Dust at Barbados	-0.49°	0.46°	-0.04	-0.16	0.24	0.08	-0.01	-0.02

Table 3.2: Cross correlation Pearson product moment coefficient between mean summer (JJAS) WACZ COA indices for JJAS season 1965 to 2003. The \diamond represents statistical significance at $\alpha < 0.1\%$. W_{ϕ} is the WACZ Latitude, W_{λ} is the WACZ longitude and W_{i} is the WACZ intensity.

	W_{ϕ}	W_{λ}	$W_{\rm i}$
W_{ϕ}	-	-0.67^{\diamond}	0.20
W_{λ}	-0.67^{\diamond}	-	-0.06
$W_{\rm i}$	0.20	-0.06	-

Table 3.3: Years that were used to generate seasonal mean composite images of wind, AOT, precipitation and PDSI based on W_{ϕ} . The lowest quartile (q25) of seasonal data is identified as the "south phase," which corresponds to the seasons when the WACZ was in a southerly position. The highest quartile (q75) of seasonal data is identified as the "north phase," which corresponds to the seasons when the WACZ was in a northerly position. Each season mean was calculated using the average of four individual months of data; June, July, August and September.

	q25			q75		
Wind	1984,	1973,	1990,	1999,	1976,	1979,
	1989,	1982,	1987,	1978,	2001,	1977,
	1972,	1981,	1991,	1967,	1974,	1966,
	1986			1965		
AOT	1984,	1990,	1989,	2005,	2003,	2002,
	1982,	1987,	1991,	1999,	2001,	2008,
	1986			2007		
Precipitation	1984,	1990,	1989,	1995,	2003,	1980,
	1982,	1987,	1981,	2002,	1999,	1979,
	1991			2001		
PDSI	1984,	1973,	1990,	1999,	1976,	1979,
	1989,	1982,	1987,	1978,	2001,	1977,
	1972,	1981,	1991,	1967,	1974,	1966,
	1986			1965		

Table 3.4: Years that were used to generate seasonal mean composite images of wind, AOT and precipitation on based on W_i . Each season mean was calculated using the average of four individual months of data; June, July, August and September.

	q25			q75		
Wind	1984,	1969,	1983,	1997,	1980,	1979,
	1990,	1991,	1981,	1971,	2000,	1998,
	1992,	1994,	1965,	1999,	2001,	1968,
	1976			1974		
AOT	1984,	1983,	1990,	2006,	2000,	2003,
	1991, 1	994, 198	6	2007, 1	998, 200	1
Precipitation	1984,	1983,	1990,	1980,	1979,	2000,
	1991,	1981,	1992,	2003,	1998,	1999,
	1994			2001		

Table 3.5: Summary of factors impacting emission and transport for changes in phase of W_{ϕ} and $W_{\rm i}$.

	W_{ϕ} South Phase	$W_{\rm i}$ Weak Phase		
Wind over Source Regions	Increases Emission	Reduces Emission		
Precipitation over Source Regions	Increases Emission	Increases Emission		
Wind over North Atlantic	Limited Increase in	Limited Increase in		
	Transport	Transport		
Precipitation over North Atlantic	Inconclusive	Varies by Location		



Figure 3.1: (a) Mean summer (JJAS) divergence (s^{-1}) at 925 hPa as computed from NCEP Reanalysis winds. The box over which the WACZ COA is computed is outlined in black. (b) Location of seasonal summer (JJAS) mean of WACZ COA over the period 1965 to 2003. Blue diamonds represent the northern (q75) phase of WACZ COA, red diamonds represent southern (q25) phase of WACZ COA. The lowest quartile (q25) of seasonal data is identified as the "south phase", which corresponds to the seasons when the WACZ was in a southerly position. The highest quartile (q75) of seasonal data is identified as the "north phase", which corresponds to the seasons when the WACZ was in a northerly position. Black diamonds represent the middle 50 percent seasons of WACZ COA.



Figure 3.2: Box-plot of monthly of the WACZ Latitude index $(W_{\phi}, \text{ top})$ and precipitation over the Sahel (15° N to 25° N and 0° to 25° E) from GPCP (bottom) for the summer season of June, July, August, September and adjacent months over the period of 1979 to 2010. Median value indicated by a horizontal red bar, edges of the central box represent the 25th and 75th percentiles. Whiskers denote the spread of all non-outlying data. Outliers are indicated with red crosses.



Figure 3.3: Time-series of JJAS seasonal WACZ Latitude index (W_{ϕ} , blue) and mineral dust concentration at Barbados (green) over the period 1965 to 2003. Seasons in which a month or more of data is missing are ignored.



Figure 3.4: (a) Composite mean of Evan AOT for the southernmost summer (JJAS) seasons (25th percentile) of the WACZ. (b) As in (a) but for the northernmost WACZ latitude (75th percentile) summer (JJAS) seasons. (c) The difference (a) minus (b), i.e. conditions of the WACZ latitude index for which dust load is maximized at Barbados. (d) First EOF mode of Evan AOT summer means, which explains 36% of total variance in the Evan AOT record. All composites and principle components are calculated over the period 1982 to 2010.



Figure 3.5: Difference in composite mean of NCEP Reanalysis winds (ms^{-1}) at 925 hPa. Differences are calculated by subtracting the seasonal summer mean (JJAS) of the northern most WACZ seasons (75th percentile) from the seasonal summer mean (JJAS) of southern most WACZ seasons (25th percentile). The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. All composites are calculated over the period 1965 to 2003; blue contour represent a significant difference at 10%.



Figure 3.6: (a) Composite mean JJAS precipitation during the southernmost latitude years of the WACZ (q25). (b) Difference in composite mean of GPCP precipitation. Differences are calculated by subtracting the seasonal summer mean (JJAS) of the northern most WACZ seasons (75th percentile) from the seasonal summer mean (JJAS) of southern most WACZ seasons (25th percentile). The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. Units are mm day⁻¹ from GPCP. All composites calculated over the period 1979 to 2003, black contour represent a significant difference at 10%.



Figure 3.7: (a) First EOF mode of GPCP seasonal JJAS precipitation over the period 1979-2010, mode one explains 42% of total GPCP variance. (b) As in (a) but for second mode of GPCP precipitation, which explains 12% of total GPCP variance. (c) Linear regression of the principle component of the first two modes (red line) onto the WACZ latitude index $(W_{\phi}, \text{ blue line})$.



Figure 3.8: Difference in composite mean of GIMMS Normalized Difference Vegetation Index (NDVI). Differences are calculated by subtracting the seasonal JJAS mean of the northern most WACZ seasons (75th percentile) from the seasonal JJAS mean southern most WACZ seasons (25th percentile). The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. The NDVI is positive when more vegetation is present than the long-term mean and negative when there is less vegetation than the long-term mean. Composite difference calculated over the period 1982 to 2002. Black contour represent a significant difference at 10%.



Figure 3.9: (a) First EOF mode of NDVI seasonal JJAS mean over the period 1979-2010, mode one explains 39% of total NDVI variance. (b) As in (a) but for second mode of NDVI, which explains 12% of total NDVI variance. (c) Linear regression of the principle component of the first two modes (green line) onto the WACZ latitude index (W_{ϕ} , blue line).



Figure 3.10: (a) Composite mean of Palmer Drought Severity Index for south mode of WACZ (q25). (b) As in (a) but for north mode of WACZ (q75). (c) Composite difference of south mode - north mode. Differences are calculated by subtracting the seasonal summer mean (JJAS) of the northern most WACZ seasons (75th percentile) from the seasonal summer mean (JJAS) of southern most WACZ seasons (25th percentile). The Palmer Drought Severity Index is negative during drought conditions. The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. All composites calculated over the period 1965 to 2003. Black contour represent a significant difference at 10%.



Figure 3.11: Difference in composite mean of NCEP Reanalysis winds at (a) 850 hPa, (b) 700 hPa and (c) 500 hPa. Differences are calculated by subtracting the seasonal summer mean (JJAS) of the northern most WACZ seasons (75th percentile) from the seasonal summer mean (JJAS) of southern most WACZ seasons (25th percentile). The difference represents the conditions of the WACZ latitude index for which dust is maximized at Barbados. Wind composites are calculated over the period 1965 to 2003; blue contour represent a significant difference at 10%. (d) Difference in composite mean of GPCP precipitation. Differences are calculated by subtracting the seasonal summer mean (JJAS) of southern most WACZ seasons (75th percentile) from the seasonal summer mean (JJAS) of southern most WACZ seasons (25th percentile). Units are mm day⁻¹ from GPCP. Precipitation composites calculated over the period 1979 to 2003.



Figure 3.12: Composite mean of difference of JJAS seasonal means from weak intensity (q25) WACZ seasons minus strong intensity (q75) WACZ seasons. Difference in wind (ms⁻¹) (a) 925 hPa and (b) 850 hPa over the period 1965 to 2003. (c) Difference in precipitation (mmday⁻¹) from 1979 to 2003. (d) Difference in AVHRR AOT over the period 1982 to 2003
1321 Chapter 4

Latitudinal Position of the Intertropical Convergence Zone over West Africa: A Comparative Study in Reanalysis and Global Climate Models of the 20th Century

1327 4.1 Introduction

The importance of the position of the Intertropical Convergence Zone (ITCZ) over West Africa in controlling dust emission and transport has been established for both winter and summer in previous chapters. Additionally the ITCZ and West African Monsoon circulation has been shown elsewhere [*Diedhiou et al.*, 1999; *Hsieh and Cook*, 2005; *Jenkins et al.*, 2005; *Nicholson*, 2008; *Sylla et al.*, 2010], to be of critical importance in determining precipitation patterns in drought-sensitive regions in sub-Saharan Africa. Motivated to consider how the position of the ITCZ may change as the Earth's climate warms in the coming century, here we first assess general circulation models (GCM) hindcasts of the 20th century. As the latitude of the ITCZ has been shown to be of critical importance our analysis focuses on this metric.

Accurately predicting year-to-year and decade-to-decade variability of the position and 1338 intensity of the ITCZ over West Africa remains a computational challenge [Sylla et al., 2011]. 1339 The structure of the ITCZ and West African Monsoon is directly impacted by vigorous 1340 convection which varies in size and scale from individual cells (micro-scale) to well organized 1341 convective clusters such as African Easterly Waves (synoptic scale) [Redelsperger et al., 2002]. 1342 Global climate models used in this study vary in resolution as coarse as 4° or 400km to as fine 1343 as 1° or 100km both of which are insufficient to resolve small scale convection. Additionally 1344 global climate models may not have the temporal resolution necessary to initiate and sustain 1345 physically reasonable convection. Variability of the ITCZ over West Africa has been linked to 1346 SST changes in far away ocean basins including the North and South Atlantic, Mediterranean 1347 and Indian Oceans [Giannini et al., 2003]. The ability of climate models to accurately predict 1348 such SST changes is also limited. Furthermore land-surface changes and aerosol loading 1349 (including dust) have been shown to impact the circulation of West Africa [Kim et al., 1350 2010; Wilcox et al., 2010], again providing a significant forecast challenge for climate models. 1351 Evaluation of convectively driven systems over West Africa has been performed in reanalysis 1352 [Diedhiou et al., 1999; Ruti and DellAquila, 2010] and in regional climate models [Sylla 1353 et al., 2010, 2011], but the ability of GCM's to represent these systems has not yet been 1354 demonstrated. 1355

Before we focus our attention on GCM projections for the 21st century climate, we first determine if the models are able to predict the known position and strength of the ITCZ over West Africa in the 20th century. This assessment is the focus of this chapter. To assess model quality we compare model output from the Atmospheric Model Intercomparison Project (AMIP) [*Gates*, 1992] scenario from 11 individual models to reanalysis products from the 20th century. In the AMIP scenario GCMs are constrained by realistic observations of ¹³⁶² sea surface temperature and sea ice coverage. In each model run the atmosphere is able to ¹³⁶³ respond freely to these oceanographic constraints. AMIP scenario data is available for each ¹³⁶⁴ of our eleven models, and is available over the period 1978 to 2002. We compare this model ¹³⁶⁵ output to NCEP, ERA-40 and ERA Interim reanalysis products which are also available ¹³⁶⁶ over the 1978 to 2002 period.

The structure of our comparison is as follows. (1) We qualitatively compare the spatial 1367 fields of divergence in GCM's to the spatial fields of divergence in reanalysis products. First 1368 the difference between GCM divergence output and the median value of three reanalysis 1369 products is analyzed to assess model deviations from observations. Secondly we compute and 1370 compare the EOF pattern for each GCM divergence output and each reanalysis divergence 1371 output. Our qualitative analysis is performed on annual, summer and winter means of 1372 divergence. (2) Next we quantify differences between models and reanalysis by applying 1373 the COA approach, focusing on how well the models are able to reproduce the previously 1374 defined WACZ latitude index. A climatological overview using boxplots of latitude and a 1375 direct comparison of time-series are made for this purpose. Scalar accuracy measures are 1376 then taken, quantifying the mean absolute error of each model and its bias. (3) Last we 1377 perform linear regression to assess trends in the WACZ latitude index. To determine the 1378 significance of observed trends a resampling technique is used, specifically bootstrapping of 1379 residuals. 1380

The utility of looking at multiple GCM models to asses synoptic scale systems is demon-1381 strated in *Sheridan and Lee* [2010]. However analysis using the "Center of Action" approach 1382 with GCM is novel, although some work has been done looking at more traditional climate 1383 teleconnection indicies such as the NAO. As both the ITCZ over West Africa and the NAO 1384 represent spatially variant patterns which are summarized but numerical indicies, we model 1385 our approach after work that has been done on the NAO in GCM. Osborn et al. [1999] 1386 separated analysis into two sections, a more qualtitative analysis of spatial variability and a 1387 quantitative analysis of the derived climate index. Here we follow this approach and divide 1388

this study in a similar manner. Following after Osborn et al. [1999] we use EOF analysis 1389 as the focus of our spatial analysis. Multiple studies of the NAO in GCM use time-series 1390 filtering on the NAO indices to remove noise from the signal [Osborn et al., 1999; Schoof and 1393 Pryor, 2006; McKendry et al., 2006] however seasonal means of the ITCZ already have most 1392 of the noise removed thus filtering is not applied in this work. Stoner et al. [2009] looks at 1393 both spatial and temporal variability of systems like the NAO and finds that while GCMs 1394 are able to recreated the general pattern, their ability to correctly place the system zonally 1395 and their ability to replicate the temporal variability is limited. In this work we are able to 1396 quantify spatial variability through the "Center of Action" approach. In this way our work 1397 demonstrates a simple framework as to how track three-dimensional variability through a 1398 simple index. 1399

A complete discussion of the global climate models, the climate model scenarios and the reanalysis products used in this study is provided in Section 4.2: Datasets. We follow this with an explanation of the statistical techniques performed in Section 4.3: Methods. We document and explain the results in Section 4.4: Results. We place the results in the context of mineral dust transport and using these GCM in 21st century scenarios in Section 4.5: Discussion.

1406 4.2 Data

1407 4.2.1 Reanalysis Data

Reanalysis data is used frequently in atmospheric science applications, as it represents the best current effort to represent historical global atmospheric conditions. However reanalysis products are not pure observations or truth, but rather derived from incomplete observations put through data assimilation and alteration in physically realistic computational models of the atmosphere. They contain biases and errors derived from a lack of observations, erroneous observations, imperfect data assimilation techniques and computational limitations

Dee [2012]. The Sahara Desert and sub-Saharan Africa are among the least sampled global 1414 land regions, featuring low spatial and temporal density of meteorological observations. As 1415 such a larger than average degree of uncertainty exists with respect to atmospheric observa-1416 tions and in turn reanalysis products. To account for potential uncertainty three reanalysis 1417 products are used here; NCEP Reanalysis, ERA 40 and ERA Interim. NCEP Reanalysis 1418 and ERA 40 have a horizontal resolution of $2.5^{\circ} \times 2.5^{\circ}$, and ERA Interim features a slightly 1419 higher resolution of $1.5^{\circ} \times 1.5^{\circ}$. The reanalysis products are available over the entire record 1420 of our comparison (1978 to 2002) with the following exceptions; the ERA Interim record 1421 begins in January 1979, and the ERA 40 record ends in August 2002. In either case any 1422 month with missing data is ignored in any calculations. 1423

Each reanalysis product has unique strengths and weaknesses. NCEP Reanalysis was 1424 one of the first reanalysis products produced and as such has been extensively studied, at 1425 the time of this writing the Kalnay et al. [1996] reference has been cited nearly 14,000 1426 times. It has been shown that NCEP Reanalysis exhibits reduced spatial and temporal 1427 moisture variability particularly over oceans and is less reliable over the Southern Hemisphere 1428 [Simmonds and King, 2004]. Additionally as an older reanalysis product its data assimilation 1429 model is somewhat outdated compared to newer data assimilation products. Two studies 1430 have documented potential errors in NCEP Reanalysis over West Africa. Poccard et al. 1431 [2000] identified a problem in early years of the NCEP Reanalysis in which inexplicable 1432 shifts in precipitation occurred over West Africa, particularly prior to 1967. Later similar 1433 shifts in geopotential height, temperature, humidity and winds were also observed over West 1434 Africa [Camberlin et al., 2001]. These studies concluded that NCEP Reanalysis data over 1435 West Africa was unreliable before 1967. This study begins in 1978, during a period in which 1436 additional information from remote sensing platforms greatly enhanced information about 1437 conditions over data sparse regions such as the Sahel and Sahara. NCEP Reanalysis is used 1438 as our primary reanalysis product as it is the most widely used dataset and also because it 1439 is the best studied reanalysis product and we are confident that major errors have already 1440

1441 been identified.

ERA-40 is the replacement for and extension of the older ERA-15 reanalysis product [*Uppala et al.*, 2005]. ERA-40 was revised to include additional sources of atmospheric data including remote sensing platforms, and has undergone assimilation and model upgrades during the process. ERA-40 has been shown to have a strong positive bias in overproducing tropical precipitation and total column water vapor, which may have direct impacts on the WACZ given that it is a tropical feature impacted by latent heating from precipitation.

ERA Interim is the newest reanalysis product used in this work and as such has the highest spatial resolution. ERA Interim is the next generation of the ERA-40 and has been modified to improve the hydrological cycle problems in ERA-40. However *Dee et al.* [2011] suggest that precipitation, particularly over oceans, has been shifted as a result of problems with satellite assimilation.

Realizing that each reanalysis product is imperfect, we frequently calculate the median monthly values and compare the renalysis products amongst themselves as well as to output from climate models to this median value to assess error. Here we note that the median of the reanalysis product is not perfect, and likely contains errors itself. At present it represents the best available estimate to assess atmospheric conditions, in particular in this case divergence at 925 hPa.

¹⁴⁵⁹ 4.2.2 CMIP3 and AMIP

¹⁴⁶⁰ CMIP3 or Climate Model Intercomparision Project 3 is part of the well known Intergovern-¹⁴⁶¹ mental Panel on Climate Chance (IPCC) and the data from this project was utilized in the ¹⁴⁶² most recent (Fourth Assessment Report (AR4)) IPCC 2007 report [Solomon et al., 2007]. ¹⁴⁶³ CMIP3 existed to form a repository for research groups to contribute their climate model ¹⁴⁶⁴ results and in turn for climate scientists to access said model results [Meehl et al., 2007]. For ¹⁴⁶⁵ ease of inter-comparison each model is forced with the same external conditions, defined as ¹⁴⁶⁶ climate scientists. In this chapter we compare model performance in the AMIP scenario to reanalysis products over the period 1979-2002. In the AMIP scenario ocean surface and sea
ice conditions are set to match observations and the atmosphere is allowed to respond to
this forcing. As such it is the scenario most easily compared to reanalysis products.

Many models from varied international research groups were submitted for included in 1470 the CMIP3 project. In this work models were only included if they were run for the 20th 1471 century comparison scenario AMIP and future scenarios SRESA1B and SRESB1. Models 1472 were excluded if they did not contain projections for the entirety of the 21st century or if 1473 they did not provide u and v component winds at 925 hPa. For this reason the number of 1474 models for inclusion in this work was limited to eleven. Full details of each model included 1475 in this work are contained in Table 4.1. Years included in the AMIP comparison scenario 1476 vary from model to model, and any season or year containing missing data is excluded from 1477 the analysis. Here we note that the version of the UKMO GCM used in the AMIP and 1478 SRESA1B was altered slightly for the SRESB1 scenario and thus omitted. For each model 1479 used and selected u and v wind components were extracted and analyzed in the same way 1480 as reanalysis products, as described in Section 4.3. 1481

$_{1482}$ 4.3 Methods

¹⁴⁸³ 4.3.1 Calculation of Divergence

As in previous chapters divergence is used as a references field for assessing the WACZ. Divergence is calculated locally from the horizontal wind (u and v) components at 925 hPa. Divergence is calculated following *Holton and Staley* [1996].

$$\nabla \cdot \hat{V}_{925} = \frac{1}{r \cos \phi} \left(\frac{\partial u}{\partial \lambda} + \frac{v \cos \phi}{\partial \phi} \right)$$
(4.1)

Here ∇ is the gradient operator $(\frac{\partial}{\partial x} + \frac{\partial}{\partial y})$, \hat{V}_{925} is the wind at 925 hPa height, u is the meridional component of the wind, v is the zonal component of the wind, r is the radius of the Earth, ϕ is the latitude and λ is the longitude.

¹⁴⁹⁰ 4.3.2 Center of Action Approach

To quantify the location and strength of the WACZ the "Center of Action" (COA) approach is applied to GCM and reanalysis divergence fields. The Center of Action is fully described in prior chapters, the same technique is used with only minor changes. For simplicity and ease of comparison a $T_{\rm h}$ of -5×10^{-6} s⁻¹ is used for all seasons, and the domain over which the COA is calculated is set to 0 to 25 °N and -20 °W to 20 °E.

¹⁴⁹⁶ 4.3.3 Statistical Approaches

To quantify how well model predictions match with observations we define two statistical metrics; mean absolute error (MAE) and bias (B) [*Wilks*, 1995]. MAE determines the average absolute deviation from observation, and B notes the direction of the deviation. The specific equations follow:

$$MAE = \frac{1}{n} \sum_{k=1}^{n} \|y_k - o_k\|.$$
(4.2)

$$B = \frac{1}{n} \sum_{k=1}^{n} (y_k - o_k).$$
(4.3)

Here n represents the number of samples, y is the model or reanalysis value and o is the observation value. When calculating mean absolute error and bias we consider the median value of the reanalysis products to be the observation.

¹⁵⁰⁴ 4.3.4 Bootstrapping for Trends

Linear trend analysis is a statistical method for determining rates of change in time-series that is ubiquitously applied in atmospheric science. However a simple linear trend analysis can be misleading. Linear trend analysis is not robust nor resistant, as it requires a timeseries to be linear and is sensitive to outliers, particularly at the start or end of the time-series.
To reduce the potential impacts of outliers here we apply a bootstrap, a common resampling
technique, to assess the uncertainty of observed trends.

Resampling represents an alternative to more traditional frequentist or non-parametric 1511 testing Wilks, 1995]. In a traditional frequentist approach a statistical test, such as linear 1512 regression, is applied and a test statistic is produced. This test statistic is then compared to 1513 an idealized distribution to assess how significant the test result is. For example a correlation 1514 coefficient obtained from a Pearson-product moment test is then compared to a distribution 1515 based on sample size to determine the correlations significance. With a resampling technique 1516 such as the bootstrap analysis used here an observation (or model output) is perturbed a 1517 large number of times (n = 100,000) to reflect uncertainty in the observation and the 1518 statistical test is performed once for each resampling. Instead of comparing the results to 1519 a hypothetical distribution the results of each of the 100,000 tests are used to determine 1520 significance. 1521

In this way resampling can be used to account for uncertainty in observations and accounts for outliers. We note that in this application the requirement that data be linear is not addressed by the resampling method, meaning the linearity assumption for regression still holds. For this analysis we resampled each reanalysis product and model 100,000 times using the data's internal variability. Each statistical test was also performed 100,000 times, and the results summarized in Section 4.4.

1528 4.4 Results

Next we focus on how well the climate models are able to reproduce divergence patterns at
925 hPa over the period 1978 to 2002. We begin with a qualitative assessment of similarity
between model output divergence and reanalysis, examining differences in long-term means

for assessing central tendency and empirical orthogonal function (EOF) loading patterns for 1532 assessment of variance. 1533

Qualitative Analysis of Divergence Spatial Pattern 4.4.11534

Annual 1535

On an annual basis the south (near equator) to north (near 16° to 18° N) seasonal migration 1536 of the WACZ from winter to summer is the dominant feature as shown in Figure 4.1. All three 1537 reanalysis products show a dipole-like feature over West Africa, with a positive mode centered 1538 between 16° and 20° N and a negative mode centered between 6° and 10° N. This dipole 1539 feature suggests that convergence and divergence are coupled in this region, with convergence 1540 associated with the WACZ and divergence of an opposing magnitude in the opposite season. 1543 For example in winter between 6° and 10° N divergence is occurring concurrently with 1542 convergence 16° and 20° N, and vice versa in summer. This annual oscillation, or the south 1543 to north migration of the WACZ is well established in the literature as well as reanalysis 1544 products and the primary source of variability that a GCM must be able to reproduce in 1545 order to be considered successful. 1546

As seen in Figure 4.1 models had varying success replicating the EOF pattern observed 1547 in reanalysis. Each model, most generally speaking, was able to identify a north-south 1548 movement of the WACZ, however the location of the oscillation and its magnitude often 1549 differed from reanalysis. The relative magnitude of the dipole pattern tells us what fraction 1550 of the total variance in divergence over Africa is associated with the annual migration of the 1551 WACZ. For example, if a model produced a lower value of relative magnitude in the dipole 1552 structure than reanalysis this would indicate that less of the total variance of divergence is 1553 controlled by the annual south to north migration of the WACZ and more of the variance is 1554 controlled by shorter term variability (either synoptic or meso- scale disturbances) or locally, 1555 topographically induced divergence. 1556

1557

The three reanalysis products Figure 4.1a–c exhibit nearly identical spatial loading pat-

terns. Some differences are evident in relative magnitude; NCEP and ERA-40 have nearly
the same relative magnitude while ERA Interim produces a weaker EOF pattern. Spatially
NCEP is nearly evenly distributed east to west, while both ERA 40 and ERA Interim show
a slightly stronger response in east Africa.

The models (Figure 4.1d–n) were successful identifying the dipole associated with south 1562 to north migration of the WACZ. CCSM (Figure 4.1d) and GISS (Figure 4.1g) least resem-1563 ble the reanalysis products with both models showing asymmetric and longitudinally offset 1564 dipole patterns. However all models do at a base level identify a positive north phase and a 1565 negative south phase. Most models had relative magnitudes of similar values to reanalysis 1566 products, however both CCSM (Figure 4.1d) and MIROC-h (Figure 4.1j) show weaker rela-1567 tive magnitudes than reanalysis, and GISS (Figure 4.1g) shows a higher relative magnitude 1568 than reanalysis. We hypothesize that this is related to resolution (see Table 4.1 for resolution 1569 details) as both CCSM and MIROC-h feature a higher resolution than ERA-40 and NCEP, 1570 while GISS has a significantly lower resolution than all reanalysis products. A model with 1571 coarser resolution would be unable to resolve local weather and climate features so larger 1572 scale patterns like the annual migration of the WACZ would control a larger fraction of vari-1573 ance, and thus a larger relative magnitude. We note the higher resolution models featured 1574 relative magnitudes closer to the higher resolution reanalysis product, ERA-Interim (Figure 1575 4.1c). 1576

Next we shift from looking at how well the models are able to replicate variability of 1577 divergence to how well they are able to recreate the mean state of divergence. To assess this, 1578 median monthly values of reanalysis products were calculated and then averaged annually. 1579 This quantity was then subtracted from each model's annual mean and the differences av-1580 eraged and plotted in Figure 4.2. The individual reanalysis products (Figures 4.2a-c) show 1581 very small differences from the median of reanalysis products. NCEP shows a northward 1582 movement of the ITCZ relative to the two other reanalyses, as well as more convergence in 1583 West Africa than the median of reanalysis products. ERA-40 most closely resembles the 1584

median of reanalysis products. Small-scale effects, some likely driven by topography are
evident in the ERA Interim which shows the largest difference from the median of reanalysis
products.

Most models differ appreciably from the median of reanalysis products. CCSM, GFDL, 1588 MIROC hres and UKMO (Figure 4.2c,f,j,n) all feature topographic discrepancies similar 1589 to ERA Interim, but generally with greater deviations from the median reanalysis. Two 1590 models, MRI and UKMO (Figure 4.2m–n), displayed a northward bias (divergence north of 1591 the median of reanalysis) while the CCSM (Figure 4.2d) displayed a southward bias. Models 1592 in general underrepresented the quantity of divergence associated with the WACZ including 1593 CNRM, GFDL, IAP, IPSL, MIROCm (Figure 4.2e, f, h, i, j) with only the UKMO (Figure 4.2n) 1594 producing larger divergence than the median of reanalysis products. We note that nearly all 1595 models, even those which are too weak generally show greater divergence over coastal Kenya 1596 than reanalysis products. Overall we find that many models have a geographical bias either 1597 in latitude or longitude in their placement of the WACZ and models in general display a 1598 weak bias with respect to the intensity of divergence. These biases are quantified in Section 1599 4.44.4.2. 1600

1601 Summer (JJAS)

Next we qualitatively examine how well the models are able to reproduce divergence over 1602 West Africa in the summer (JJAS). In contrast to the annual EOF analysis which includes 1603 all months and is indicative of the annual migration of the WACZ, this analysis only includes 1604 the summer months of JJAS. Here a north to south dipole signal would indicate that the 1605 dominant variability in the divergence data is annual differences in which the WACZ is in a 1606 "north phase" versus a "south phase". Figure 4.3 shows the first mode of the EOF analysis. 1607 In reanalysis Figures (4.3a–c) it appears that the majority of variability occurs over central 1608 and east Africa. A north-south pattern emerges around Lake Chad in each of the three 1609 reanalysis products. 1610

Models have widely varying loading patterns, many of which do not resemble reanalysis 1611 products. CNRM, IAP, MRI and UKMO (Figures 4.3e,h,m,n) all have the dominant pattern 1612 settled to the west compared to the reanalysis products. GFDL, GISS, IPSL, MIROC-m, and 1613 MPI (Figures 4.3f,g,i,j,l) all show an eastward positioning of the mode of greatest variability, 1614 similar to reanalysis but often of with varied latitudinal displacement of the patterns. Lastly 1615 CCSM and MIROC-h (Figures 4.3d,k) show an asymmetric pattern that is dissimilar to any 1616 of the reanalysis products. We note that this pattern appears to be strongly influenced by 1617 topographically generated divergence. 1618

Despite geographic shifts in the loading patterns between models, most models have a similar relative magnitude to reanalysis products. Only the GISS model (Figure 4.3g) was stronger than all reanalysis products, wheras the CCSM and MIROC-h (Figures 4.3d,k) are both significantly weaker than reanalysis products.

Figure 4.4 shows the long term mean of divergence in summer. Each reanalysis product 1623 shows an east to west band of divergence between 16° and 18° N, each showing the band 1624 drawn further north over West Africa. ERA Interim is most dissimilar to the other two 1625 reanalysis products with additional local features evident, likely as a function of the local 1626 topography. CCSM and MIROC-h (Figures 4.4d,k) again stand out with neither featuring 1627 the expected east to west band of divergence anticipated from reanalysis, rather in both 1628 local features dominate the signal, to a larger extent that the ERA Interim product. Other 1629 models show the expected band of divergence over West Africa, similar to reanalysis. We 1630 note that CNRM, GFDL and IAP (Figures 4.4c,f,h) are each clearly north of the reanalysis 1631 products. 1632

To quantify the differences from reanalysis we again calculate the median of summer months of divergence, calculate a seasonal mean of reanalysis products and then subtract reanalysis mean divergence from the model mean, results are shown in Figure 4.5. NCEP reanalysis (Figure 4.5a) shows a NW to SE difference, with more convergence over the Niger River basin and more divergence south of Lake Chad relative to median reanalysis. ERA-40 (Figure 4.5b) shows a similar but weaker W-E dipole between the Niger River and south of
Lake Chad, suggesting that again the ERA-Interim is different from the other two reanalysis
products.

¹⁶⁴¹ CCSM and MIROC-hres (Figures 4.5d,k) show significant differences from median reanal-¹⁶⁴² ysis, again due to topographical influence. Here we confirm the northward bias in CNRM, ¹⁶⁴³ GFDL and IAP (Figures 4.5c,f,h) as indicated in Figure 4.4. IPSL exhibit a southward ¹⁶⁴⁴ bias (Figures 4.5i). While the models closely approximate the intensity of the divergence ¹⁶⁴⁵ in reanalysis, GISS, IAP and MPI (Figures 4.5g,h,l) appear to have too little convergence ¹⁶⁴⁶ while UKMO (Figure 4.5n) appear to have too much convergence. We again note models ¹⁶⁴⁷ overestimate divergence near the Kenyan coast.

¹⁶⁴⁸ Winter (DJFM)

For winter in reanalysis products the first mode of the EOF of divergence is a zonally symmetric dipole focused over west Africa, as seen in Figures 4.6a–c. We note the strongest relative magnitudes associated with this pattern are found in the COA box. The magnitude of the loading pattern is largest in NCEP (Figure 4.6a), followed by decreasing magnitudes in ERA-40 (Figure 4.6b) and the lowest magnitude in ERA Interim (Figure 4.6c). We note the same pattern for latitudinal displacement, NCEP is furthest north, ERA-40 slightly offset to the south, and ERA Interim furthest south.

Few models are able to replicate the first EOF geographic positioning of reanalysis products. CNRM, GISS, IAP, MRI (Figures 4.6e,h,m) locate the first mode too far east relative to reanalysis. GFDL, IPSL, MIROC-m, MPI, UKMO (Figures 4.6f,i,j,l,n) hint at the maximum variability shifting to West Africa, but in general are also located to the east of reanalysis. Figures 4.6d,k again suggest that topography induced divergence plays an important role in the CCSM and MIROC-hres model, but we note that relative to summer in winter topographic influences are reduced as the WACZ moves southward.

¹⁶⁶³ Models in general produce similar relative magnitudes to reanalysis products, in partic-

ular CNRM, GFDL, IAP, IPSL (Figures 4.6e,f,h,i). We note again that higher resolution
models like CCSM and MIROC-h, as well as UKMO have lower relative magnitudes (Figures
4.6 d,k,n). Low resolution models, such as the GISS model (Figure 4.6g) appear to have
relative magnitudes greater than reanalysis.

The long term mean of divergence in winter is shown in Figure 4.7. Each of the reanalysis products shows a clear zonal band of divergence between 6° and 12° N. ERA Interim (Figure 4.7c) features a narrower, tighter band closer to the Gulf of Guinea than either NCEP Reanalysis (Figure 4.7a) or ERA-40 (Figure 4.7b). Models appear to do a satisfactory job with the placement of the WACZ, although in general they tend to produce lower values of divergence than reanalysis.

As with summer (in Figure 4.5) for winter we again compute the mean of the difference between median reanalysis and model results in Figure 4.8. NCEP reanalysis (Figure 4.8a) shows a clear northward shift in the WACZ relative to the median of reanalysis products, while ERA Interim (Figure 4.8c) shows a southward shift relative to the median of reanalysis products. ERA-40 (Figure 4.8b) is closest to the median of reanalysis products. We note that differences between individual reanalysis products in winter are larger than those seen in summer (Figure 4.8).

In winter each model has significant differences compared to the median of the reanalysis 1681 products. Many models appear to have features consistent with topographically induced 1682 discrepancies, e.g. CCSM, GFDL, MIROC hres, UKMO (Figure 4.8d,f,j,n). Latitudinal dis-1683 placements relative to reanalysis products were observed with models exhibiting both north-1684 ward (GFDL, MRI and UKMO in Figures 4.8f,m,n) and southward biases (CCSM, CNRM, 1685 MIROC (both) in Figures 4.8d,e,j,k). In general many models under represented convergence 1686 near to the WACZ, e.g CNRM, IAP, IPSL, MIROC (both), MPI (Figures 4.8e,h,i,j,k,l) while 1687 none appear to over represent convergence near to the WACZ. Again as before in summer 1688 and for the annual mean each model generates strong divergence near to the coastline of 1689 Kenya relative to reanalysis. 1690

To summarize a number of results emerged consistently throughout each season and an-1691 nually. Both low resolution and high resolution models are markedly different than reanalysis 1692 products. The lowest resolution model (GISS) underestimated variability across the domain 1693 as a whole and exhibited large differences from reanalysis products in the placement and 1694 strength of the WACZ. High resolution models such as the MIROC-hres and CCSM each 1695 have more noisy divergence patterns, much of which emerging from topographic features 1696 not resolved in reanalysis. All models produce a convergence along the Kenya coastline not 1697 seen in any reanalysis product. While individual models can show large biases from season 1698 to season, these biases are not necessarily consistent across seasons. As such we need to 1690 quantify the bias of each model as a function of season. In the next section we apply the 1700 "Center of Action" approach to each model and quantitatively assess each performance. 1701

1702 4.4.2 Quantitative Analysis of WACZ COA Indices

1703 Annual Mean Position of WACZ Latitude

To assess both the temporal and spatial variability of the WACZ latitude W_{ϕ} here we present 1704 boxplots of monthly values of the WACZ latitude in Figure 4.9. Red lines in the center of 1705 the box represent the median value, and blue lines above and below represent the 75th and 1706 25th percentiles respectively. Blue lines extending out from the center of the box extend 1.5 1707 times the interquartile range from the median. Outliers are denoted by red plus symbols. 1708 Thus boxplots show skewness, spread and central tendency of W_{ϕ} . A companion statistical 1709 summary of Figure 4.9 is shown in Table 4.2. Reanalysis products (Figures 4.9a–c) clearly 1710 shows the annual cycle, with a maximum value of W_{ϕ} in July to August of 16° to 18° N 1711 and a minimum value of W_{ϕ} in December to January of 7° to 9° N. In fall months and to 1712 a lesser extent spring months, year-to-year variability is maximized, leading to the largest 1713 spread. From January through June W_{ϕ} undergoes slow and gradual northward migration. 1714 In contrast from September through November W_{ϕ} experiences a very rapid southward tran-1715 sition. The slow and steady northward transition followed by a rapid southward transition 1716

leads to an asymmetric temporal migration on an annual basis. Models all capture winter to 1717 summer migration observed in reanalysis however subtle, yet important differences emerge. 1718 Most models replicate the mean annual position of the WACZ within range of the reanalysis 1719 products (8 of 11 in Table 4.2). The MIROC-mres model is nearly 2 degrees south of the 1720 reanalysis product, while both GISS and IAP are approximately 1 degree north of the re-1721 analysis products. The majority of models (8 of 11) contain less variance in the latitudinal 1722 position of the WACZ than the reanalysis products. The remaining three models (CNRM, 1723 GISS and IPSL) exceed reanalysis variance, while no model produced variance within the 1724 range of reanalysis variance. 1725

Mean annual conditions are important in assessing the models ability to replicate the 1726 movement of the WACZ but to be considered effective the model must also exhibit similar 1727 temporal variability compared to observations. Absolute error and bias of the model lati-1728 tude are calculated relative to the reanalysis latitude. Individual reanalysis latitudes (NCEP, 1729 ERA-40, ERA Interim) are significantly correlated with the mean and median of the reanal-1730 vsis products as shown in Table 4.3, although NCEP and ERA-Interim are not correlated 1731 with each other on an annual basis. To simplify the analysis we use the median latitude of 1732 the reanalysis to compare to model latitude. To assess the temporal variability of the models 1733 we first correlate the model latitude (W_{ϕ}) to the reanalysis latitude. Although generally the 1734 models are able to capture the mean position of the WACZ successfully, the models fail to 1735 replicate the change in WACZ with time as in the reanalysis. Only the MIROC-mres shows 1736 any skill at replicated in the annual mean position of the WACZ. Here we note that MIROC 1737 both qualitatively and quantitatively does a poor job replicating the mean position of the 1738 WACZ latitude compared to other models, so this result is surprising. MRI and CCSM show 1739 anti-correlation with reanalysis products. 1740

The fact that 10 of 11 models cannot replicate the annual position of the WACZ latitude (Table 4.2) suggests that the models are missing a key component of internal variability needed to accurately predict the mean location on an annual basis, although they do show ¹⁷⁴⁴ some skill in replicating the WACZ latitudes' long term mean position.

¹⁷⁴⁵ Mean Summer (JJAS) Position of WACZ Latitude

Figure 4.10 shows the position of the WACZ as determined by the latitude and longitude 1746 index of the WACZ COA. Table 4.5 presents statistics summarizing W_{ϕ} in summer, which 1747 are supported with bias and error statistics in Table 4.7. Each reanalysis product places 1748 the WACZ north and east of the bend in the Niger River. ERA-40 (Figure 4.10b) appears 1749 to be the furthest south of the three reanalysis products, and ERA Interim (Figure 4.10c) 1750 is furthest north. NCEP Reanalysis (Figure 4.10a) shows extensive longitudinal variability. 1751 Models place the WACZ with varying degrees of success. IAP, MIROC-hres and UKMO 1752 (Figure 4.10h,k,n) are tightly clustered, suggesting less variability than reanalysis. CNRM 1753 and GFDL (Figure 4.10e–f) each appear to have more scatter than reanalysis. 1754

Most models are close to the reanalysis position while only one model was north of all 1755 reanalysis products (GISS) and only two models were found south of all reanalysis products 1756 (MIROC-mres and UKMO). A majority of models in Table 4.5 (9 of 11) display less variance 1757 in the latitudinal position of the WACZ than the median of reanalysis products, while the 1758 CNRM experienced significantly more variance than the median of reanalysis. Only the 1759 GISS produced a variance consistent with reanalysis products. The time-series of all W_{ϕ} 1760 are shown in Figure 4.11, and the correlation between the median of reanalysis and each 1761 model and reanalysis product W_{ϕ} are shown in Table 4.6. Visual inspection of the Figure 1762 4.11 suggests limited temporal coherence between models and reanalysis. No model shows 1763 significant correlation with the median of reanalysis. Again we note models are unable to 1764 replicate the temporal variability observed in reanalysis during the summer. 1765

¹⁷⁶⁶ Mean Winter (DJFM) Position of WACZ Latitude

¹⁷⁶⁷ In winter the WACZ COA in reanalysis tends to be clustered near the intersection of the ¹⁷⁶⁸ Niger and Benue Rivers as seen in Figures 4.12a–c. Differences in position between the

reanalysis products are smaller in winter compared to summer. This is confirmed in Table 1769 4.8 which shows the median and interquartile range of reanalysis products are similar in 1770 winter. In general models place the winter WACZ COA near to reanalysis products, although 1771 again we note some models (CCSM, MIROC-hres and UKMO; Figures 4.12d,k,n) appear 1772 tightly cluster compared to reanalysis and others (MRI and MPI; Figures 4.12l-m) appear 1773 too scattered relative to reanalysis. We note in Table 4.8 that for 5 of 11 models (CCSM, 1774 GFDL, IAP, MIROC-hres, UKMO) the median W_{ϕ} positions were north of all reanalysis 1775 products, and for 3 of 11 models (CNRM, MIROC-mres, MPI) median W_{ϕ} were south of all 1776 reanalysis products. Models tended to exhibit a larger latitudinal spread than reanalysis with 1777 5 of 11 models (CNRM, GISS, IAP, MPI, MRI) having a larger interquartile range (IQR) 1778 than all reanalysis products and only the CCSM model having less spread than reanalysis. 1779 Figure 4.13 shows the timeseries of reanalysis products and models for W_{ϕ} . Reanalysis 1780 products (bold lines) covary tightly in time, as shown by high correlation co-efficients in 1781 Table 4.9. We note more agreement in winter between reanalysis than in summer (Table 1782 4.6), which is reflected in bias and mean absolute error calculations as well (Table 4.10). 1783 Models are largely unable to reproduce the temporal variability seen in the reanalysis with 1784 only IAP and MIROC-hres significantly correlated with reanalysis. 9 of 11 models show no 1785 significant correlation with reanalysis products. 1786

Despite differences in model physics, data assimilation techniques, observations and res-1787 olution the reanalysis products are qualitatively similar in their handling of the divergence 1788 of the WACZ, although minor differences emerge. Models show a mixed ability to replicate 1789 the central tendency and spread of reanalysis products, with models doing better in winter 1790 and showing larger differences in summer. No model is able to replicate the temporal vari-1791 ability of reanalysis. As the climate warms the Hadley cell is expected to drift northward 1792 [Gastineau et al., 2008], and with it perhaps the WACZ. In the next section we assess the 1793 trend in reanalysis and CMIP models. 1794

1795 4.4.3 Trend Analysis of WACZ Latitude

In this section we apply bootstrapping to linear trend analysis of our COA data. To summarize the results we present four figures. The first figure summarizes the mean (x-axis) and standard deviation (y-axis) of the resampled data. The next two figures assess the trend, first by calculating the linear trend (°yr⁻¹) in each resampled set of data and then by calculating the correlation coefficient between the W_{ϕ} and time for each resampled set of data. Last the probability of a significant trend (p-value) is assessed for each resampled set of data.

1802 Trend in Annual Mean

The mean and standard deviations of resampled annual means are shown in Figure 4.14. 1803 Both ERA-40 and ERA Interim show a strong majority of resamplings exhibit a northward 1804 annual trend as evident in Figure 4.15. NCEP reanalysis shows a southward trend, with a 1805 similarly significant quantity of resamples showing this southward trend. While both ERA-40 1806 and ERA Interim show nearly all resamplings have a positive correlation (northward trend) 1807 with time in Figure 4.16, the relationship is not statistically significant for NCEP reanalysis. 1808 The model response is also varied in sign. CNRM, GFDL, GISS, MIROC-hres all suggest a 1809 possible northward trend (Figure 4.15e–g,k and 4.16e–g,k) while MRI and UKMO (4.15m– 1810 n and 4.16m–n) are suggestive of a southward trend. While more reanalysis products and 1811 models suggest a northward trend in the annual mean W_{ϕ} the results are overall inconclusive. 1812 Table 4.11 shows an overview of trend statistics for summer. Interestingly reanalysis 1813 products show contradictory evidence of statistically significant northward and southward 1814 trends. NCEP Reanalysis shows a southward trend that is statistically significant with a 1815 *p*-value of 0.031. ERA-40 and ERA-Interim show a northward trend that are statistically 1816 significant with a p-value of 0.042 and 0.017 respectively (also seen in Figure 4.17). No 1817 clear result is obvious from this analysis or reanalysis. Three models produce a statistically 1818 significant northward trend (CNRM, GFDL, MIROC-hres). 1819

1820 Trend in Summer (JJAS) Mean

The mean of resampled summer means is shown in Figure 4.18. In summer all three re-182 analysis products show evidence of a northward trend in Figures 4.19a–c. ERA Interim has 1822 no resamplings with a non-positive (non-northward) trend, while ERA-40 and ERA Interim 1823 include a few non-positive trends. A similar result is shown in Figures 4.20a–c, with a large 1824 majority of resamplings for each reanalysis product showing a positive (northward) correla-1825 tion with time. Extrapolating each trend over the period 1979 to 2002 suggests a northward 1826 shift of nearly 1°. Models suggested both northward (CNRM, GFDL, MRI; Figures 4.19e– 1827 f,m) and southward trends (IAP, UKMO; Figures 4.19h,n). Reanalysis products conclusively 1828 suggest a robust and significant northward movement in the WACZ from 1979 to 2002, while 1829 some models captured this trend, most did not. 1830

Table 4.12 shows an overview of trend statistics for summer. ERA-40 (r = 0.35, p < 0.03) 1831 and ERA Interim (r = 0.70, p < 0.01) each have a significant trend as indicated by time-series 1832 correlation and *p*-values (significance is seen graphically in Figure 4.21). NCEP Reanalysis 1833 (r = 0.28, p = 0.116) fell just short of significance, but we note it does have an insignificant 1834 northward trend. Two models, CNRM and CCSM, each showed a significant northward 1835 trend as well and the UKMO showed a significant southward trend. Much as the models 1836 could not capture the interannual variability of the WACZ, here they appear to not capture 1837 the temporal trend with any skill either. 1838

1839 Trend in Winter (DJFM) Mean

The mean of resampled winter means is shown in Figure 4.22. In winter all three reanalysis products show evidence of a southward trend in Figures 4.23a–c. NCEP shows the strongest southward trend, with ERA-40 and ERA-Interim each successively showing less of a trend. Four models hint at a southward trend as well (IPSL, MPI and MRI, UKMO) in Figures 4.23i,l–n, while three models suggest the opposite, northward trend (CCSM, GFDL, MIROChres) in Figures 4.23d,f,k. Time-series correlation suggest a similar result in Figure 4.20. Table 4.13 shows an overview of trend statistics for winter. Each of the three reanalysis products shows a negative or southward trend, however based on median correlation and median *p*-values of the resampled tests only the NCEP Reanalysis is statistically significant (r = -0.486, p < 0.01). Significance is shown graphically in Figure 4.25. A negative trend was only found to be statistically significant in two models, MRI and UKMO. Two other models had statistically significant northward or positive trends, GFDL and MIROC-hres.

The summary of the trend analysis is shown in Table 4.15. Overall from reanalysis we 1852 find a statistically significant northward trend of the WACZ in summer in 2 of 3 reanalysis 1853 products, and in winter we find some evidence of a southward trend of the WACZ but this 1854 is only statistically significant in 1 of 3 reanalysis products. On an annual basis results are 1855 mixed with NCEP suggesting a significant southward trend of the WACZ, and ERA-40 and 1856 ERA Interim suggesting a significant northward trend of the WACZ. We conclude from this 1857 that in summer the WACZ is likely moving northward, and in winter the WACZ is possibly 1858 drifting southward, thus potentially increasing the annual range of the WACZ, although this 1859 is not shown to be statistically significant. We note GFDL and MIROC-hres each produces 1860 trends in the opposite direction of reanalysis in both winter and summer. 1861

1862 4.5 Discussion

Each of the eleven models used in this study have been spatially and temporally analyzed 1863 for mean position, variance and trend of W_{ϕ} compared to reanalysis products. Table 4.14 1864 shows model absolute error (MAE) of W_{ϕ} relative to median reanalysis for each season and 1865 annual mean, as well as the total of each season and annual mean. As the latitude of the 1866 WACZ is the focus of this work the sum of MAE is used to rank models from best to worst. 1867 Below we discuss and summarize each models strengths and weaknesses as previously shown 1868 in Section 4.4. Models are divided into three sections, the strongest performing models had 1869 MAE similar to reanalysis products, the middling performing models had MAE over two 1870

times as large as reanalysis products and the poor performing models had MAE three times
as large as reanalysis products. IAP and MIROC-mres are rated as poor performing models.
IPSL, MIROC-hres and MPI are rated as the best performing models.

1874 Strong Performing Models

¹⁸⁷⁵ **IPSL** IPSL was the best performing model, comparable in skill with the ERA Interim model ¹⁸⁷⁶ at determining the position of W_{ϕ} . While overall IPSL did well, the absolute convergence it ¹⁸⁷⁷ generated in the WACZ was less than reanalysis, particularly in winter and the annual mean, ¹⁸⁷⁸ and experienced less variance than the median in summer. IPSL's EOF loading pattern was ¹⁸⁷⁹ slightly east of reanalysis in winter.

MIROC-hres MIROC-hres performed well in spite of including topographically in-1880 duced divergence features not included in reanalysis. We note that the majority of these 1883 topography-induced features were outside of the domain in which the COA is calculated 1882 over West Africa. This increased variance induced by local topography resulted in EOF 1883 patterns that were weaker than reanalysis in all seasons for MIROC-hres. In summer the 1884 EOF pattern in MIROC-hres was asymmetric, and W_{ϕ} experienced less variance than any 1885 reanalysis product. Although MIROC-hres displayed a southward bias in its position of W_{ϕ} 1886 in winter, it was able to recreate annual variability as seen in reanalysis. 1887

¹⁸⁸⁸ **MPI** MPI performed just as well as MIROC-hres overall, performing very well in summer. ¹⁸⁸⁹ MPI displayed a weak bias, that is to say it produced less convergence in the WACZ than ¹⁸⁹⁰ did reanalysis products in both summer and winter. MPI underrepresented variance of W_{ϕ} ¹⁸⁹¹ in both winter and summer, and its EOF loading pattern was shifted east in winter relative ¹⁸⁹² to reanalysis.

CNRM CNRM, as did MIROC-hres and MPI, performed very well in summer, outperforming its winter placement despite summer in general having more variability and less predictability. CNRM displays a weak bias, but under representing convergence in the WACZ relative to reanalysis in winter and annual means. CNRM is biased in both winter and sum¹⁸⁹⁷ mer, with a north bias in winter and a south bias in winter. CNRM's EOF loading pattern ¹⁸⁹⁸ is shifted east of reanalysis in winter. Unlike most other models CNRM has more variance ¹⁸⁹⁹ in W_{ϕ} than reanalysis products in both summer and winter.

¹⁹⁰⁰ Middling Performing Models

¹⁹⁰¹ **MRI** MRI experienced bias in each season, north in annual mean and winter, and a south ¹⁹⁰² bias in summer. EOF loading patterns were longitudinally offset, west in summer and east ¹⁹⁰³ in winter. The W_{ϕ} from MRI experience more variance than reanalysis in winter and less ¹⁹⁰⁴ variance in summer. The annual mean of MRI was anti-correlated with reanalysis products, ¹⁹⁰⁵ suggesting poor skill at representing interannual variability.

GFDL GFDL had the lowest resolution of any model in the study, and visual inspection of its divergence fields suggested poor skill with the WACZ, but in terms of total error from reanalysis the model is in the middle of the pack. GFDL placed the WACZ north of reanalysis products in both winter and summer and underestimated the strength of convergence in the WACZ, particularly in annual means. In summer this model produced less variance than reanalysis. We also noted that GFDL produced an EOF loading pattern east of reanalysis in winter.

UKMO UKMO performed poorly in summer compared to winter, with a nearly four 1913 times larger error in summer, resulting in a moderate overall performance. UKMO showed 1914 marked differences from reanalysis in EOF loading patterns, for example it was west of 1915 reanalysis in summer, east of reanalysis in winter and generally of lower relative magnitudes, 1916 particularly in winter. UKMO showed a northward bias in its position in both winter and 1917 annual means. Topographic induced disturbances were evident, causing differences from 1918 reanalysis products, although mainly north and east of the domain over which W_{ϕ} was 1919 calculated. UKMO was one of the only models to produce convergence in excess of that 1920 observed in reanalysis, particularly in summer and annual means. Variability associated 1921 with W_{ϕ} was lower in summer in UKMO. 1922

¹⁹²³ **CCSM** CCSM struggled to predict W_{ϕ} in summer. The EOF loading pattern in CCSM ¹⁹²⁴ varied from reanalysis in all seasons, we note that the relative magnitude for CCSM was ¹⁹²⁵ less than reanalysis in every season. In summer and on an annual basis the EOF pattern ¹⁹²⁶ is asymmetric. W_{ϕ} showed reduced variance compared to reanalysis in both summer and ¹⁹²⁷ winter. We note that CCSM showed anti-correlation with reanalysis for annual means. ¹⁹²⁸ Topographically induced divergence patterns are evident in CCSM that do not appear in ¹⁹²⁹ reanalysis.

GISS GISS showed a consistent MAE in every season, including the annual mean. GISS underestimated convergence in the WACZ relative to reanalysis, particularly in summer. W_{ϕ} is found to be 1° north of reanalysis in the annual mean. GISS shows marked divergence from reanalysis in its EOF loading pattern in winter as it is too far east, and asymmetric for the annual mode. GISS had relative magnitudes larger than that found in reanalysis in all seasons. The variability of W_{ϕ} in GISS was lower than that of reanalysis in both summer and winter.

¹⁹³⁷ Poor Performing Models

MIROC-mres MIROC-mres had large MAE in each season, but performed particularly 1938 poorly in summer. MIROC-mres produced less convergence in the WAC in winter and for 1939 annual means. The loading pattern of the MIROC-mres EOF was too far east in winter. 1940 MIROC-mres had a significant south bias in winter and was nearly 2° south of the annual 1941 mean of reanalysis. However despite its south bias of annual mean, it was correlated with 1942 reanalysis. In summer the W_{ϕ} of MIROC-mres had less variance than reanalysis in summer. 1943 **IAP** IAP was the worst performing model, faring particularly poorly in summer. IAP 1944 underestimated convergence relative to reanalysis in every season. The loading pattern of 1945 EOF for IAP displayed a west bias in summer and an east displacement in winter. A north 1946 bias was evident in both summer and annual value. W_{ϕ} of IAP showed less variance than 1947 reanalysis in both summer in winter. Despite its overall poor performance IAP was correlated 1948

¹⁹⁴⁹ with the time-series of reanalysis in winter.

1950 4.6 Conclusions

Models were able to recreate the annual south to north migration of the WACZ successfully, 1951 but many models struggled to replicate seasonal mean positions, seasonal variability and all 1952 models were unable to replicate temporal variability. The inability of the models to replicate 1953 temporal variability may limit the utility of model projections for the 21st century. While in 1954 summer clear northward trends were observed in reanalysis products, models as a whole were 1955 unable to simulate this northward migration, although some individual models were able to 1956 simulate the northward migration. Three models were identified as performing similarly to 1957 reanalysis, while two models were identified as performing poorly. 1958

This study began with a spatial analysis of divergence, looking at changes in mean state and variability. Empirical orthoganal function analysis show that models able to capture the north-to-south migration of the WACZ. However models show appreciable differences in the exact positioning of the oscillation, and its strength. With respect to long-term mean individual models displayed north or south biases, but overall underestimated total convergence in the WACZ.

The models exhibit the least coherence in JJAS with the greatest variability in divergence 1965 pattern and largest deviations from reanalysis occurring in summer. Differences between re-1966 analysis products are also maximized in JJAS. The largest fraction of divergence in reanalysis 1967 is shifted east of the box over which the COA is calculated. Based on EOF patterns we note 1968 that the models generally do a poor job in summer replicating reanalysis variance and long 1969 term mean position. We note that higher resolution models and reanalysis products veer 1970 from the traditional zonal band image of the WACZ highlighting potential complexities from 1971 local/small scale sources. 1972

¹⁹⁷³ DJFM highest variance in divergence shifts west over the COA box, and reanalysis prod-

¹⁹⁷⁴ ucts differ slightly in latitudinal position of the WACZ. Models closer to reanalysis in winter,
¹⁹⁷⁵ but in general contain less convergence over WACZ.

The utility of the centers of action approach was shown as an strong quantitative measure 1976 of model performance. In reanalysis products W_{ϕ} clearly demonstrates the annual cycle 1977 maximizing annually in summer between 16° to 18° N and reaching a minimum in winter 1978 between 7° to 9° N. We find an asymmetric migration with a slow steady northward migration 1979 in spring and a rapid southward migration in fall. Eight of eleven models place the mean 1980 annual position within range of reanalysis products, however only one of eleven models 1981 is significantly temporally correlated with reanalysis products suggesting an inability to 1982 replicate interannual variability. 1983

The COA approach confirms many of the biases and mean errors suggested in the qualitative approach for the summer season. With respect to the mean positioning of the COA models do well in summer, with only one model north of all reanalysis products and only two models south of all reanalysis products. Likewise models struggle to recreate the variance of W_{ϕ} with nine of 11 models having less variance than any of the reanalysis products. Similarly to the annual cycle, models show no correlations with any reanalysis product, suggesting in an inability to replicate interannual variability.

In winter, reanalysis products are very similar to each other, with less mean absolute error and bias when compared to summer. Five of eleven models predict latitudinal position of WACZ north of reanalysis, and three of eleven models predict latitudes south of observations in reanalysis products. Again models were unable to recreate the temporal variability, as only 2 of 11 models had any significant correlation with reanalysis products.

The summary of the trend analysis is shown in Table 4.15. Overall from reanalysis we find a statistically significant northward trend of the WACZ in summer in 2 of 3, and in winter we find some evidence of a southward trend of the WACZ but this is only statistically significant in 1 of 3 reanalysis products. On an annual basis results are mixed with NCEP suggesting a significant southward trend of the WACZ, and ERA-40 and ERA Interim suggesting a significant northward trend of the WACZ. We conclude from this that in summer the WACZ
is likely moving northward, and in winter the WACZ is possibly drifting southward, thus
potentially increasing the annual range of the WACZ, although this is not shown to be
statistically significant.



Figure 4.1: First mode of the EOF of 925hPa divergence (all months) from CMIP3-AMIP scenario. Years used in the calculation are shown in in Table 4.1. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.2: Annual model mean divergence from CMIP3-AMIP scenario minus the annual median reanalysis divergence over the period 1978 to 2002. Before differences are calculated all data is regridded to the lowest common resolution between reanalysis products and model. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.3: First mode of the EOF of 925hPa divergence in summer (JJAS) from CMIP3-AMIP scenario. Years used in the calculation are shown in in Table 4.1. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.4: Long term mean of 925hPa divergence in summer (JJAS) from CMIP3-AMIP scenario. Years used in the calculation are shown in in Table 4.1. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.5: Summer (JJAS) model mean divergence from CMIP3-AMIP scenario minus the winter median reanalysis divergence over the period 1978 to 2002. Before differences are calculated all data is regridded to the lowest common resolution between reanalysis products and model. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.6: First mode of the EOF of 925hPa divergence in winter (DJFM) from CMIP3-AMIP scenario. Years used in the calculation are shown in in Table 4.1. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.7: Long term mean of 925hPa divergence in winter (DJFM) from CMIP3-AMIP scenario. Years used in the calculation are shown in in Table 4.1. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.8: Winter (DJFM) model mean divergence from CMIP3-AMIP scenario minus the winter median reanalysis divergence over the period 1978 to 2002. Before differences are calculated all data is regridded to the lowest common resolution between reanalysis products and model. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.


Figure 4.9: Monthly values of WACZ Latitude Index. Red line represent monthly median value, with blue verticle lines representing the 75th and 25th percentiles respectively. Outliers are indicated with an asterisk. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.10: Position of WACZ COA in summer (JJAS) from CMIP3-AMIP scenario over the period 1978 to 2002. Blue diamonds represent the 75th northern percentile of WACZ latitude position. Red diamonds represent the 25th southern percentile of WACZ latitude position. Black diamonds represent the middle 50th percentile of WACZ latitude seasons. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.11: Timeseries of summer (JJAS) WACZ Latitude (W_{ϕ}) for the three reanalysis products and 11 model runs over the period 1978 – 2002.



Figure 4.12: Position of WACZ COA in winter (DJFM) from CMIP3-AMIP scenario over the period 1978 to 2002. Blue diamonds represent the 75th northern percentile of WACZ latitude position. Red diamonds represent the 25th southern percentile of WACZ latitude position. Black diamonds represent the middle 50th percentile of WACZ latitude seasons. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.13: Timeseries of winter (DJFM) WACZ Latitude (W_{ϕ}) for the three reanalysis products and 11 model runs over the period 1978 – 2002.



Figure 4.14: Mean and standard deviation of WACZ latitude of annual means from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.15: Linear trend of WACZ latitude in annual means from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.16: Correlation co-efficients between time and WACZ latitude in annual means from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.17: *P*-values of significance of trend of WACZ latitude in annual means from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.18: Mean and standard deviation of WACZ latitude in summer (JJAS) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.19: Linear trend of WACZ latitude in summer (JJAS) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.20: Correlation co-efficients between time and WACZ latitude in summer (JJAS) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.21: *P*-values of significance of trend of WACZ latitude in summer (JJAS) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.22: Mean and standard deviation of WACZ latitude in winter (DJFM) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.23: Linear trend of WACZ latitude in winter (DJFM) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.24: Correlation co-efficients between time and WACZ latitude in winter (DJFM) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) GISS, (h)IAP, (i)IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.



Figure 4.25: *P*-values of significance of trend of WACZ latitude in winter (DJFM) from CMIP3-AMIP scenario over the period 1978 to 2002. Top row (reanalysis product): (a) NCEP Reanalysis, (b) ERA-40, (c) ERA Interim. Second row: (d) CCSM, (e) CNRM, (f) GFDL. Third row: (g) ISS, (h) IAP, (i) IPSL Fourth row: (j) MIROC-mres, (k) MIROC-hires, (l) MPI. Fifth row: (m) MRI and (n) UKMO-HADGEM.

Model	Host Institution	Location	Resolution	Start	End	Reference	ce
Acronym NCEP Reanal- ysis	NCEP NCAR	Boulder, CO, USA	$2.5^{\circ} \times 2.5^{\circ}$	Jan 1948	present	et [1996]	al.
ERA-40	European Centre For Medium-Range Weather Forecasts	Reading, United Kingdom	$2.5^{\circ} \times 2.5^{\circ}$ Sep 1957	Aug 2002	[Uppale et al., 2005]	a	
ERA- Interim	European Centre For Medium-Range Weather Forecasts	Reading, United Kingdom	$1.5^{\circ} \times 1.5^{\circ}$	Jan 1979	present	5 [<i>Dee et</i> 2011]	al.,
NCAR CCSM	National Center for At- mospheric Research	Boulder, CO, USA	$1.40625^{\circ} \times 1.40625^{\circ}$	Jan 1978	Dec 2000	Collins et [2006]	al.
CNRM- CM3	Centre National de Recherches Meteo- rologques, Meteo-France	Toulouse, France	$2.8125^{\circ} \times 2.8125^{\circ}$	Jan 1979	Dec 2000		
GFDL AM2.1	NOAA Geophysical Fluid Dynamics Labora- tory	Princeton, NJ, USA	$2^{\circ} \times 2.5^{\circ}$	Jan 1980	Nov 1999		
GISS ModelE	NASA Goddard Institute for Space Studies	New York, NY, USA	$4^{\circ} \times 5^{\circ}$	Jan 1979	Dec 2002		
IAP	Institute for Atmo- spheric Physics	Beijing, China	$3^{\circ} \times$ 2.8125°	Jan 1978	Dec 1999	Wang et [2004]	al.
IPSL	Institut Pierre Simon Laplace	Paris, France	$2.5^{\circ} \times$ 3.75°	Jan 1979	Dec 2002		
$\frac{\text{MIROC}}{(\text{mres})}$	Center for Climate System Research	Tokyo, Japan	$2.8125^{\circ} \times 2.8125^{\circ}$	Jan 1979	Dec 2002		
MIROC (hires)	Center for Climate Sys- tem Research	Tokyo, Japan	$\begin{array}{c} 1.125^\circ \times \\ 1.125^\circ \end{array}$	Jan 1979	Dec 2002		
MPI- ECHAM5	Max Planck Institute for 5 Meteorology	Hamburg, Germany	$1.875^{\circ} \times 1.875^{\circ}$	Jan 1978	Dec 1999		
MRI	Meteorological Research Institute	Tsukuba, Japan	$2.8125^{\circ} \times 2.8125^{\circ}$	Jan 1979	Dec 2002	Yukimot et [2001]	to al.
UKMO- HADGEN	Met Office M	Exeter, United Kingdom	$\begin{array}{c} 1.25^{\circ}\times\\ 1.875^{\circ}\end{array}$	Jan 1979	Dec 2000		

Table 4.1: Summary of key information on CMIP GCMs and Reanalysis used, including acronyms, host institution, resolution, start and end times for each scenario and the primary reference for each.

Table 4.2: Exploratory data analysis overview of annual means of WACZ Latitude W_{ϕ} using robust and resistant statistical measures. Median (°) is use to represent central tendency. Interquartile Range (IQR) (°) represents spread. Yule-Kendall Index represents skewness.

Model Acronym	Median	Interquartile Range	Skewness Index
NCEP Reanalysis	12.06	0.32	0.52
ERA-40	11.65	0.29	-0.03
ERA-Interim	12.59	0.36	0.20
CCSM	12.43	0.18	0.68
CNRM	11.98	0.41	-0.07
GFDL	12.54	0.21	-0.48
GISS	13.63	0.40	0.17
IAP	13.38	0.26	0.39
IPSL	12.34	0.38	0.51
MIROC-mres	10.99	0.22	-0.67
MIROC-hres	12.96	0.27	-0.14
MPI-ECHAM	12.08	0.26	-0.48
MRI	12.12	0.28	-0.15
UKMO-HADGEM	12.03	0.24	-0.84

Table 4.3: Pearson product-moment correlation coefficient matrix of annual means of WACZ latitude (W_{ϕ}) COA calculated from reanalysis products and CMIP-AMIP. Column headers abbreviations as follows: NCEP: NCEP Reanalysis, ERA 40: ERA 40, ERA IN: ERA Interim, R_{μ} : mean of three reanalysis products, and $R_{q_{50}}$: median of three reanalysis products. An * indicates statistical significance at 10%, [†] at 5% and ^o at 1%.

Model Acronym			W_{ϕ}		
· ·	NCEP	ERA 40	ERA IN	R_{μ}	$R_{q_{50}}$
NCEP Reanalysis	_	0.52^{\diamond}	-0.01	0.64^{\diamond}	0.85°
ERA-40	0.52^{\diamond}	—	0.63^{\diamond}	0.93^{\diamond}	0.82^{\diamond}
ERA-Interim	-0.01	0.63^{\diamond}	—	0.74^{\diamond}	0.41^{\dagger}
R_{μ}	0.64^{\diamond}	0.93^{\diamond}	0.74^{\diamond}	—	0.90^{\diamond}
$R_{ m q_{50}}$	0.85^{\diamond}	0.82^{\diamond}	0.41^{\dagger}	0.90^{\diamond}	—
CCSM	-0.14	-0.40	-0.17	-0.34*	-0.42
CNRM	-0.12	0.11	0.35^{*}	0.16	0.03
GFDL	-0.04	-0.36*	0.11	-0.11	-0.17
GISS	-0.02	-0.01	-0.21	-0.12	-0.06
IAP	-0.01	0.33^{*}	0.28	0.27	0.16
IPSL	-0.06	0.07	0.09	0.05	-0.07
MIROC-mres	0.21	0.34^{*}	0.32^{*}	0.38^{\dagger}	0.37^{*}
MIROC-hres	0.23	0.43^{\dagger}	0.19	0.37^{*}	0.29
MPI-ECHAM	-0.02	-0.06	-0.18	-0.11	-0.03
MRI	-0.23	-0.20	-0.24	-0.30	-0.32^{*}
UKMO-HADGEM	0.14	-0.24	-0.17	-0.12	-0.01

	W_{ϕ} MAE	W_{ϕ} B
NCEP	0.29	-0.19
ERA 40	030	-0.33
ERA I	0.49	0.49
CCSM	0.52	-0.51
CNRM	0.73	-0.66
GFDL	1.25	1.25
GISS	1.38	1.38
IAP	1.55	1.55
IPSL	0.37	0.05
MIROC-m	1.38	-1.38
MIROC-h	0.59	0.59
MPI	0.65	-0.62
MRI	0.94	-0.93
UKMO	0.70	-0.70

Table 4.4: Mean absolute error and bias of WACZ COA latitude index (W_{ϕ}) in three reanalysis products and eleven global circulation models. Error and bias are calculated with respect to the median WACZ COA latitude index (W_{ϕ}) of three reanalysis products.

Table 4.5: Exploratory data analysis overview of WACZ latitude index for summer season (JJAS) means using robust and resistant statistical measures. Median is use to represent central tendency. Interquartile Range (IQR) represents spread. Yule-Kendall Index represents skewness.

Model Acronym	Median	Interquartile Range	Skewness Index
NCEP Reanalysis	15.03	0.45	-0.10
ERA-40	14.66	0.52	-0.37
ERA-Interim	16.35	0.54	-0.37
CCSM	14.96	0.41	0.18
CNRM	16.09	0.74	-0.28
GFDL	14.87	0.47	0.15
GISS	17.35	0.55	-0.21
IAP	16.24	0.29	1.27
IPSL	14.87	0.36	1.06
MIROC-mres	14.13	0.21	-0.60
MIROC-hres	15.71	0.34	0.39
MPI-ECHAM	16.23	0.37	0.19
MRI	14.97	0.46	-0.98
UKMO-HADGEM	14.47	0.39	0.20

Table 4.6: Pearson product-moment correlation coefficient matrix of summer (JJAS) means of WACZ latitude (W_{ϕ}) COA calculated from reanalysis products and CMIP-AMIP. Column headers abbreviations as follows: NCEP: NCEP Reanalysis, ERA 40: ERA 40, ERA IN: ERA Interim, R_{μ} : mean of three reanalysis products and $R_{q_{50}}$: median of three reanalysis products. An * indicates statistical significance at 10%, [†] at 5% and ^o at 1%.

Model Acronym			W_{ϕ}		
	NCEP	ERA 40	ERA IN	R_{μ}	$R_{\mathbf{q}_{50}}$
NCEP Reanalysis	_	0.56^{\diamond}	0.33 *	0.70^{\diamond}	0.91^{\diamond}
ERA-40	0.56°	—	0.48^{\dagger}	0.83^{\diamond}	0.75^{\diamond}
ERA-Interim	0.33 *	0.48^{\dagger}	—	0.78^{\diamond}	0.41^{\dagger}
R_{μ}	0.70*	0.83^{\diamond}	0.78^{\diamond}	—	0.85^{\diamond}
$R_{ m q_{50}}$	0.91°	0.75^{\diamond}	0.41^{\dagger}	0.85^{\diamond}	—
CCSM	-0.18	-0.25	0.17	-0.09	-0.28
CNRM	0.11	0.12	0.73^{\diamond}	0.42^{\dagger}	0.19
GFDL	0.20	-0.08	0.24	0.15	0.15
GISS	0.00	0.22	0.14	0.17	0.06
IAP	-0.28	-0.24	-0.22	-0.34	-0.38
IPSL	0.10	0.03	-0.01	0.05	0.06
MIROC-mres	0.20	0.01	-0.03	0.06	0.07
MIROC-hres	0.23	-0.01	-0.14	0.03	0.15
MPI-ECHAM	-0.10	0.27	0.13	0.14	-0.01
MRI	-0.32	-0.06	0.06	-0.12	-0.25
UKMO-HADGEM	0.37 *	0.01	0.30	0.28	0.26

	W_{ϕ} MAE	W_{ϕ} Bias
NCEP	0. 21	-0.12
ERA 40	0.78	-0.78
ERA I	0.88	0.88
CCSM	2.07	-2.07
CNRM	0.84	0.53
GFDL	1.04	0.92
GISS	1.35	1.35
IAP	2.22	2.22
IPSL	0.60	-0.27
MIROC-m	2.10	-2.10
MIROC-h	0.57	-0.49
MPI	0.52	0.02
MRI	1.75	-1.75
UKMO	2.14	-2.14

Table 4.7: Mean absolute error and bias of WACZ latitude index (W_{ϕ}) in three reanalysis products and eleven global circulation models for summer (JJAS). Error and bias are calculated with respect to the median WACZ latitude index (W_{ϕ}) of three reanalysis products.

Table 4.8: Exploratory data analysis overview of WACZ COA in winter (DJFM) using robust and resistant statistical measures. Median is use to represent central tendency. Interquartile Range (IQR) represents spread. Yule-Kendall Index represents skewness.

	Median	Interquartile Range	Yule-Kendall Index
NCEP Reanalysis	9.26	0.37	-0.35
ERA-40	8.46	0.30	0.44
ERA-Interim	8.88	0.48	0.07
CCSM	9.74	0.24	-0.76
CNRM	7.73	0.57	-2.19
GFDL	10.37	0.44	0.05
GISS	9.56	0.61	0.74
IAP	10.17	0.68	1.02
IPSL	9.13	0.34	0.58
MIROC-mres	7.69	0.34	0.58
MIROC -hires	9.96	0.37	-1.09
MPI-ECHAM	8.24	0.71	0.40
MRI	9.23	0.72	0.57
UKMO-HADGEM	9.39	0.40	-0.22

Table 4.9: Pearson product-moment correlation coefficient matrix of winter (DJFM) means of WACZ latitude (W_{ϕ}) COA calculated from reanalysis products and CMIP-AMIP. Column headers abbreviations as follows: NCEP: NCEP Reanalysis, ERA 40: ERA 40, ERA IN: ERA Interim, R_{μ} : mean of three reanalysis products and $R_{q_{50}}$: median of three reanalysis products. An * indicates statistical significance at 10%, [†] at 5% and ^o at 1%.

Model Acronym			W_{ϕ}		
	NCEP	ERA 40	ERA IN	R_{μ}	$R_{\mathbf{q}_{50}}$
NCEP Reanalysis	_	0.71°	0.77^{\diamond}	0.88^{\diamond}	0.84°
ERA-40	0.71*	—	0.86^{\diamond}	0.94^{\diamond}	0.88^{\diamond}
ERA-Interim	0.77*	0.86^{\diamond}	_	0.95^{\diamond}	0.97^{\diamond}
R_{μ}	0.88*	0.94^{\diamond}	0.95^{\diamond}	—	0.98^{\diamond}
$R_{ m q_{50}}$	0.84*	0.88^{\diamond}	0.97^{\diamond}	0.98^{\diamond}	—
CCSM	-0.24	-0.18	-0.33*	-0.23	-0.24
CNRM	0.19	0.25	0.30	0.29	0.30
GFDL	-0.31	-0.11	-0.11	-0.18	-0.15
GISS	0.01	-0.19	-0.06	-0.11	-0.10
IAP	0.45^{\dagger}	0.44^{\dagger}	0.56^{\diamond}	0.52^{\diamond}	0.57^{\diamond}
IPSL	0.06	0.09	-0.16	0.01	-0.07
MIROC-mres	-0.04	-0.08	-0.08	-0.08	-0.09
MIROC-hres	0.41^{\dagger}	0.24	0.31	0.33^{*}	0.36^{*}
MPI-ECHAM	0.19	-0.08	-0.05	0.01	-0.02
MRI	0.12	-0.07	0.01	-0.01	-0.05
UKMO-HADGEM	0.26	-0.06	0.04	0.08	0.08

Table 4.10:	Mean	absolute err	or and bi	ias of II	ГCZ	COA	indi	$\cos in$	three	rean	alysis p	rodı	acts
and eleven	global	circulation	models.	Error	and	bias	are	calcula	ated	with	respect	to	${\rm the}$
median ITC	CZ CO.	A indices of	three rea	analysis	prod	lucts.							

	W_{ϕ} MAE	W_{ϕ} Bias
NCEP	0.18	-0.05
ERA 40	0.09	-0.04
ERA I	0.15	0.01
CCSM	0.95	0.95
CNRM	1.48	-1.45
GFDL	0.90	0.88
GISS	1.05	1.00
IAP	0.90	0.90
IPSL	0.51	0.17
MIROC-m	1.16	-1.10
MIROC-h	1.19	1.17
MPI	1.18	-1.18
MRI	0.41	0.02
UKMO	0.59	0.46

Table 4.11: Summary of statistics associated with the bootstrap analysis of linear trend of annual means over the period 1979 to 2002. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . $r_{0.5}$ is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a t-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
NCEP	-0.0152	-0.425	0.424	-0.0532	0.031
ERA-40	0.0154	0.00497	0.65	0.355	0.042
ERA Interim	0.0171	-0.0487	0.593	0.287	0.0167
CNRM	0.0203	-0.193	0.637	0.253	0.0716
CCSM	0.00368	-0.367	0.51	0.0963	0.128
GFDL	0.0251	-0.169	0.723	0.342	0.0494
GISS	0.0118	-0.196	0.628	0.236	0.141
IAP	-0.0032	-0.429	0.375	-0.0284	0.212
IPSL	-0.0051	-0.492	0.213	-0.138	0.185
MIROC-hres	0.0116	-0.133	0.595	0.261	0.0395
MIROC-mres	-0.0079	-0.433	0.471	-0.0255	0.178
MPI	0.0054	-0.415	0.471	0.029	0.196
MRI	-0.00598	-0.556	0.244	-0.149	0.171
UKMO	-0.00546	-0.646	0.0775	-0.329	0.113

Table 4.12: Summary of statistics associated with the bootstrap analysis of linear trend in summer (JJAS) over the period 1979 to 2002. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . $r_{0.5}$ is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a *t*-test.

	Trend _{0.5}	$r_{ m min}$	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
NCEP	0.0134	-0.172	0.744	0.275	0.116
ERA-40	0.0261	0.00833	0.646	0.351	0.0303
ERA Interim	0.0421	0.504	0.833	0.703	0.0000101
CNRM	0.0524	-0.0128	0.684	0.37	0.0174
CCSM	0.0136	-0.141	0.619	0.297	0.0287
GFDL	0.0225	-0.282	0.603	0.185	0.164
GISS	-0.00404	-0.624	0.257	-0.192	0.205
IAP	-0.0227	-0.599	0.15	-0.261	0.137
IPSL	0.00155	-0.289	0.467	0.111	0.217
MIROC-hres	-0.00301	-0.422	0.408	-0.0223	0.213
MIROC-mres	0.00126	-0.349	0.472	0.07	0.217
MPI	0.00595	-0.423	0.477	0.00226	0.209
MRI	0.0243	-0.225	0.559	0.2	0.103
UKMO	-0.0152	-0.78	-0.129	-0.493	0.0694

Table 4.13: Summary of statistics associated with the bootstrap analysis of linear trend of winter (DJFM) means over the period 1979 to 2002. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . $r_{0.5}$ is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a t-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
NCEP	-0.0294	-0.789	-0.102	-0.486	0.00967
ERA-40	-0.0109	-0.491	0.2542	-0.148	0.161
ERA Interim	-0.00711	-0.534	0.266	-0.158	0.182
CNRM	-0.0191	-0.487	0.46	-0.0158	0.163
CCSM	0.00657	-0.308	0.546	0.163	0.128
GFDL	0.0475	0.00421	0.813	0.442	0.0145
GISS	0.0111	-0.368	0.509	0.0881	0.178
IAP	-0.0116	-0.527	0.367	-0.106	0.183
IPSL	-0.0142	-0.58	0.103	-0.258	0.12
MIROC-hres	0.0176	-0.0372	0.619	0.331	0.0551
MIROC-mres	-0.0136	-0.522	0.237	-0.179	0.185
MPI	-0.0223	-0.633	0.293	-0.211	0.129
MRI	-0.0332	-0.739	-0.226	-0.526	0.00269
UKMO	-0.00951	-0.672	0.156	-0.312	0.0813

Table 4.14: Summary of total absolute mean error (MAE) in models and reanalysis relative to median reanalysis values. The models are sorted in ascending order of MAE.

	Annual	DJFM	JJAS	Total
NCEP	0.29	0.18	0.21	0.68
ERA 40	0.30	0.09	0.78	1.17
ERA I	0.49	0.15	0.88	1.52
IPSL	0.37	0.51	0.60	1.48
MIROC-hres	0.59	1.19	0.57	2.35
MPI	0.65	1.18	0.52	2.35
CNRM	0.73	1.48	0.84	3.05
MRI	0.94	0.41	1.75	3.10
GFDL	1.25	0.90	1.04	3.19
UKMO	0.70-	0.59	2.14	3.43
CCSM	0.52	0.95	2.07	3.54
GISS	1.38	1.05	1.35	3.78
MIROC-mres	1.38	1.16	2.1	4.64
IAP	1.55	0.90	2.22	4.67

Table 4.15: Summary of p-value statistics associated with the bootstrap analysis of linear trend of winter (DJFM) means over the period 1979 to 2002. Table counts instances in which a reanalysis product (left) or model (right) expresses a significant trend, as determined by a p-value from a two-tailed asymmetric t-test statistic. P-values of greater than 0.1 are described as an insignificant trend or no trend, p-values below 0.1 but in excess of 0.02 are described as significant trend, and p-values below 0.02 are described as extremely significant.

	Reanalysis					CMIP AMIP Models				
	Ext.	Sig.	No	Sig.	Ext.	Ext.	Sig.	No	Sign.	Ext.
	Neg.	Neg.	Trend	Pos.	Pos.	Neg.	Neg.	Trend	Pos.	Pos.
	Trend	Trend		Trend	Trend	Trend	Trend		Trend	Trend
	p <	p <	$p \geq$	p <	p <	p <	p <	$p \geq$	p <	p <
	0.02	0.1	0.1	0.1	0.02	0.02	0.1	0.1	0.1	0.02
Annual	0	1	0	1	1	0	0	8	3	0
JJAS	0	0	1	1	1	0	1	8	2	0
DJFM	1	0	2	0	0	1	0	9	1	0

²⁰⁰⁵ Chapter 5

Predicted Changes in Position of Intertropical Convergence Zone Over West Africa in 21st Century in CMIP3

2010 5.1 Introduction

Precipitation in the tropics, the narrow band between 15° N and 15° S accounts for more than one third of global precipitation [*Simpson et al.*, 1988]. The latent heating resulting from this extreme rate of precipitation is of great importance to global climate; impacting both regional weather and weather in mid-latitude regions. Moreover the radiation of the tropical atmosphere is of great interest and has been studied extensively on decadal timescales [*Chen et al.*, 2002; *Wielicki et al.*, 2002].

A large fraction of the precipitation in the tropics is associated with upward motion in the ascending branch of the Hadley Cell, the dominate feature in the tropical atmosphere [*Oort and Yienger*, 1996]. As such changes in circulation associated with the Hadley Cell

have been extensively studied. Multiple studies using observations have suggested that the 2020 Hadley Cell has increased in strength towards the end of the 20th century [Chen et al., 2002; 2021 Tanaka et al., 2004; Mitas and Clement, 2005]. However in contrast most GCMs predict a 2022 weakening of the Hadley Cell in recreations of the 20th century [Mitas and Clement, 2006]. 2023 Both observational and modeling studies have focused on the "strength" of the Hadley Cell 2024 to assess variability. For example in both Mitas and Clement [2005] and Mitas and Clement 2025 [2006] streamfunction and velocity potential are used to quantify the strength of the Hadley 2026 Cell. While this approach is effective in assessing changes in the mean overturning of the 2027 atmosphere, this approach does not provide information about potential changes in the 2028 position of the Hadley Cell. 2020

More recently research suggests that the Hadley Cell expanded towards the end of the 2030 20th century and is likely to expand further in the 21st century as the climate warms. Hu and 203 Fu [2007] find using outgoing longwave radiation (OLR) data derived from multiple satellite 2032 measurements a poleward expansion of the Hadley Cell of 2.4° in winter (DJF) and 2.2° in 2033 summer (JJA) over both hemispheres over the period 1979 to 2005. Some of the widening 2034 of the Hadley Cell has been attributed to depletion of ozone [Polvani and Kushner, 2002] 2035 while the pole to equator temperature gradient and mid-latitude storm track activity are 2036 also related to the poleward position of the Hadley Cell. 2037

Over West Africa the WACZ represents the ascending branch of the Hadley Cell [Folland 2038 et al., 1986]. The position of the WACZ is of great importance in determining the spatial 2039 extent of precipitation in sub-Sahran Africa, where northward movement of the WACZ could 2040 lead to increases in precipitation in the Sahel and decreases in precipitation in traditionally 2041 wetter regions nearer to the equator. Interannual variability of the latitude of the WACZ has 2042 been shown previously to be important in determining the annual emission and transport of 2043 mineral dust from West Africa to the Americas [Doherty et al., 2012a]. The natural question 2044 arises of what are the implications of changes in strength and poleward expansion of the 2045 Hadley Cell on the position of the WACZ? While as previously mentioned much attention 2046

has been paid to the Hadley Cell in general, less work has been paid to the ascending branch
of it in detail. *Gastineau et al.* [2008] looking at multiple GCMs showed that the ascending
branch of the Hadley Cell, located near to the equator, may shift in the 21st century, although
the direction of the shift was unclear. This work was done examining the streamfunction at
500 hPa.

In this work we apply the COA framework to 21st century projections in CMIP GCMs 2052 to assess changes in the latitudinal position of the WACZ focusing on answering three ques-2053 tions. (1) What interannual and decadal variability in the latitude of the WACZ do GCMs 2054 predict for the 21st century and how does this compare to 20th century observations and in 2055 GCMs from AMIP? (2) Is the northward trend observed in the 20th century (see chapter 2056 4) continued in GCMs in the 21st century? (3) Do models that performed poorly in AMIP 2057 perform differently than those models identified as strong performing models in the 21st 2058 century? 2059

The structure of this chapter is as follows. In Section 5.2 the models and climate scenarios used in this project are introduced. In Section 5.3 we present the results, divided by scenario. For each scenario annual, summer and winter means are discussed focusing on changes in mean state compared to 20th century, changes in variability of W_{ϕ} and trends over the 21st century. In Section 5.4 we discuss our results in context of the literature and the implications for mineral dust emission and transport in the 21st century. In Section 5.5 we conclude our findings.

²⁰⁶⁷ 5.2 Data and Methods

²⁰⁶⁸ 5.2.1 CMIP and the SRES Scenarios

For the sake of consistency models used in this analysis are identical to those used in the prior chapter. All models were again taken from the CMIP-3 modeling inter-comparison project. The exact years included in the analysis are contained in Table 5.1. Here we note that the ²⁰⁷² UKMO model employed in the AMIP analysis in Chapter 4 is only available for one of two ²⁰⁷³ climate scenarios analyzed here, it is omitted from the scenario in which it is missing. Table ²⁰⁷⁴ 5.2 provides an overview of the socioeconomic assumptions of the two climate scenarios used ²⁰⁷⁵ in this work (reproduced from *Solomon et al.* [2007]). Each scenario is described in more ²⁰⁷⁶ detail below.

The A1 scenarios are characterized by a world in which rapid economic growth is pri-2077 oritized within an "integrated world" or one that exchanges new and efficient technological 2078 advances freely. In the A1 scenarios global human population continues its exponential 2079 growth until reaching a maximum of 9 billion in the year 2050 and then following the demo-2080 graphic transition slowly decreases with time. In this scenario it is assumed that developing 2081 countries develop quickly and in time global income and lifestyles become more homoge-2082 neous. The specific scenario used in this study (A1B) assumes that energy is derived from 2083 a mix of fossil fuel, nuclear and alternative energies. For comparison other A1 scenarios 2084 (not used in this study) utilize an either an all fossil fuel energy source (A1FI) or an energy 2085 supply focused on alternatives (A1T). As such A1B represents a middle ground between the 2086 extremes of energy source scenarios. 2087

In contrast the B1 scenarios reflect a future that is considered to be ecologically friendly in which global attention is paid to solving environmental problems as they emerge. In these scenarios rapid economic growth indeed occurs however the focus of growth is a service and information based economy, demanding less energy use. These scenarios assume reductions in material consumption and development and distribution of clean, resource efficient technologies. Population growth is identical to scenario A1.

Figure 5.1 shows the total emissions in the 21st century associated with both scenarios and additional SRES scenarios. Scenario A1B shows enhanced emissions relative to Scenario B1, but less than other scenarios in which fossil fuels use is encouraged. Likewise Scenario B1 shows a reduction relative to Scenario A1B, but is not the scenario with minimal emissions. As such Scenario A1B and Scenario B1 represent moderate scenarios within the manifold of all expected scenarios. Indeed as the "best estimate" of 21st century warming is given a range of between 0.6°C and 4.0°C in the varied scenarios, the warming expected with A1B and B1 of 2.8°C and 1.8°C, respectively, again places the scenarios toward the middle of possible outcomes.

Otherwise all methods employed in this chapter are identical to those used in Chapter 4, Section 4.3.

2105 5.3 Results

Next we present an overview of the mean state, variability and trends of divergence at 2106 925 hPa and W_{ϕ} as calculated from divergence in CMIP3 GCM. We compare the output 2107 from two scenarios SRES-A1B and SRES-B1 to each other and to 20th century reanalysis 2108 and the CMIP3-AMIP scenario as discussed in Chapter 4. The structure of this section is as 2109 follows. For each scenario we present a brief overview of how the model handles the annual 2110 migration of the WACZ and its interannual variability. Next for both summer (JJAS) and 2111 winter (DJFM) we present a detailed overview of how models handle the spatial distribution 2112 of divergence, discuss both inter-annual and decedal variability in W_{ϕ} and conclude with a 2113 discussion of trends in each season. 2114

2115 5.3.1 Scenario: SRES-A1B

2116 Annual Means

The primary mode of variability for the ITCZ over West Africa is its annual south to north migration occurring each boreal winter into summer. Figure 5.2 presents the first loading pattern of the EOF of 925 hPa divergence for the eleven CMIP models in scenario SRES-A1B for all months over the period 2000 to 2100. Each model replicates the south-north dipole, with a pattern flipping sign in the latitude belt 8° N to 16° N. We note differences in relative magnitude are proportional to model resolution, with higher resolution models having lower relative magnitudes suggesting that a larger degree of variability in these models is containedin other features.

The seasonal cycle of W_{ϕ} is shown in Figure 5.3. Again each model captures a south to 2125 north migration of the WACZ latitude index however the amplitude (annual range) and phase 2126 (timing) of the migration vary from model to model estimation of the oscillation. In the 20th 2127 century reanalysis we noted a asymmetric seasonal cycle, with a slow and steady northward 2128 migration in boreal spring into early summer, and then a rapid irregular southward migration 2129 in autumn. We note that CCSM, IPSL, MIROC-h, MRI and UKMO (Figures 5.3a,f,h,j-k) 2130 show a rather steady and smooth southward migration in fall, in contrast to 20th century 2131 reanalysis which featured a less symmetric north to south migration. GFDL, GISS and 2132 UKMO (Figures 5.3c-d,k) show two annual peaks, an early peak in May and a secondary 2133 peak in July–August, this feature was not evident in 20th century reanalysis or in AMIP 2134 model scenarios. 2135

The time-series of 21st century W_{ϕ} in scenario SRES-A1B is shown in Figure 5.4, for 2136 comparison 20th century reanalysis products are shown in Figure 5.5. Visual inspection of 2137 the time-series of W_{ϕ} suggest a northward trend with time, later in this section we confirm 2138 this trend in a full trend analysis. Accompanying Figure 5.4 is Table 5.3, which shows the 2139 median, interquartile range and skewness of W_{ϕ} annual means. Here we note that the models 2140 show reduced interannual variability compared to 20th century reanalysis (Figure 5.5) and 2141 do not show any evidence of multi-decadal variability. Figure 5.5 shows evidence of the 2142 drought-like conditions in the 1970's and 1980's in which the WACZ was shifted southward, 2143 and precipitation was correspondingly reduced in the Sahel. During this period in NCEP 2144 Reanalysis the WACZ is on average 2 degrees south of its position in the 1950's and 1960's, 2145 and a degree south of its position in the 2000's. Visual analysis of model WACZ in Figure 2146 5.4 shows no similar multi-decadal shifts in the position of the WACZ. Likewise inter-annual 2147 variability is reduced in models in the 21st century in Figure 5.4 compared to reanalysis 2148 products in the 20th century in Figure 5.5. 2149

In summary, models are able to capture the annual migration of the ITCZ over West Africa, although some irregularities in monthly position are noted as compared to the 20th century. A northward trend is visibly evident in the time-series of W_{ϕ} . Less variability is produced by models relative to the 20th century observations on both inter-annual and decadal scales. It is not clear if such reductions in variability are related to model error, or rather are prediction for reductions in the 21st century. In the next section we analyze models prediction of the summer season.

2157 Summer Means

We expect, based on 20th century reanalysis, that the first mode of summer (JJAS) means 2158 should reflect changes in phase associated with seasons in which the WACZ is north and 2159 seasons in which the WACZ is south. Figure 5.6 shows the first mode of annual summer 2160 means over the period 2000–2100 in scenario SRES-A1B. In general we note the first loading 2161 pattern of the EOF is associated with changes in phase of the WACZ, with a dipole evident 2162 about the mean summer position of the WACZ (Figure 5.7, generally 16° N to 18° N). 2163 In models with high resolution such as CCSM, MIROC-h (Figures 5.6a,k) we note lower 2164 relative magnitudes, suggesting less of total variability is associated with changes in north-2165 south phase. 2166

The long term mean 925 hPa divergence is shown in Figure 5.7. The majority of models 2167 show a East-West band of convergence just north of 16° N. Over westernmost Africa, this 2168 East-West band experiences a slight bend and slight northward jog relative to central and 2169 east Africa (this was also seen in the 20th century). Figure 5.8 shows the difference of 2170 individual models in 21st century, scenario SRES-A1B minus the median of 20th century 2171 reanalysis products. Under this convention negative values represent increased convergence 2172 in the 21st century relative to the 20th, and positive values represent reduced convergence in 2173 the 21st century than the 20th century. IAP (Figure 5.8e) is a good example of mean model 2174 response, with a reduction in convergence century north of 16° N concurrent with increased 2175

convergence south of 16° N, overall suggesting a mean southward shift of the WACZ in SRES-2176 A1B in the 21st century. MIROC-h and UKMO (Figures 5.8h,k) are examples of models 2177 that show the opposite, both exhibiting a northward shift of the WACZ in SRES-A1B. The 2178 southward shift in position of the zone of convergence associated with the WACZ seen in 2179 most models from 20th century to the 21st century could be a result of one of two factors; a 2180 net bias in position between scenarios or a shift with time and changing atmospheric forcing. 2181 In the case of the former, a large shift in position between the 20th century and 21st century 2182 without a trend would be indicative of model induced bias. We examine the latter possibility 2183 by looking at trends in the record later in this section. 2184

Figure 5.9 couples W_{ϕ} with W_{λ} to show a graphical representation of the position of the 2185 WACZ over West Africa. A summary of median of W_{ϕ} location and spread (interquartile 2186 range) are presented in Table 5.5. Comparison with similar plots from the AMIP scenario of 2187 the 20th century as shown in Chapter 4 confirms the southward shift as suggested in previous 2188 figures. Figure 5.10 shows the boxplot of summer means of W_{ϕ} in all models, presented 2189 next to each other for comparison purposes. Horizontal lines across Figure 5.10 show the 2190 maximum and minimum values of median annual values. IAP and IPSL have medians above 2191 (north of) the range of the model median, and MIROC-m and UKMO have medians below 2192 (south of) the range of model medians. IPSL, MIROC-h and MRI all have reduced ranges 2193 compared to other models, suggesting lower seasonal variance. The time-series of W_{ϕ} in 2194 the 21st century for SRES-A1B is presented in Figure 5.11 and for comparison the time-2195 series of W_{ϕ} in reanalysis products during the 20th century is shown in Figure 5.12. The 2196 models produce less inter-annual variability than do 20th century reanalysis products. Again 2197 evidence of a southern phase of W_{ϕ} in the 1970's and 1980's is seen in reanalysis while models 2198 do not predict any decadal shifts of similar magnitude in the 21st century. A northward trend 2199 over the 21st century is evident in most models. 2200

To assess the significance of the northward trend we employ a bootstrapping technique on linear regression. Our time-series of summer means is re-sampled 10,000 times and the
linear trend, correlation with time, a p-value associated with the slope of the linear trend are 2203 calculated once for each of our 10,000 time-series. An overview of the median and standard 2204 deviation of each re-sampling is shown in Figure 5.13. A summary of key statistics is shown 2205 in Table 5.6. Figure 5.14 shows the distribution of linear trends for each of the 10,000 2206 model re-samplings, a clear majority of models exhibit a positive (northward) trend on the 2207 order of 0.01 $^{\circ}$ yr⁻¹. GISS (Figure 5.14d) is the outlier, indicating a negative (southward) 2208 trend. Figure 5.15 shows a positive correlation between W_{ϕ} and time suggesting a northward, 2209 linear trend. Figure 5.16 shows the significance of the trend as assessed by a student's t-2210 test on linear regression co-efficients. Empty histograms show that all tests were near zero, 2211 indicating significance of the trend. CCSM, GISS and MRI all contained some samplings 2212 that were insignificant, however the majority of cases for each (at least 95%) were significant 2213 at $\alpha = 0.1$. 2214

In summary we note in summer that a north-south dipole is the main mode of variability 2215 suggesting that north-south changes in phase of W_{ϕ} is the dominant climate factor associated 2216 with 925 hPa divergence. As predicted W_{ϕ} has little inter-annual and multi-decadal vari-2217 ability, especially when compared to 20th century reanalysis variability, which showed both 2218 large interannual and decadal variability. Compared to 20th century reanalysis the majority 2219 of models experience a southward shift in mean state, but experience a northward trend in 2220 time, suggesting a major southward bias introduced in 21st century model runs which is 2221 manifested at year 2000 and reduces over time accounting to the superimposed northward 2222 temporal trend. 2223

2224 Winter Means

Most models identify a north-south dipole associated with inter-annual changes in phase of the WACZ during winter? as shown in Figure 5.17. We note the dipole shifts towards West Africa, consistent with 20th century reanalysis. All models feature this dipole pattern, each centered about 8° to 10° N. The climatological winter mean of 925 hPa divergence as seen in scenario SRES-A1B is shown in Figure 5.18. Clear, consistent east to west bands of convergence are visible between 8° to 12° N in each model, although clear differences in mean state are visible between models. Model-to-model variability appears to be reduced from summer (as it was in the 20th century), a strong example of which is shown in CCSM (Figure 5.18a).

Spatial plots showing the 20th century divergence from median reanalysis minus the 2234 21st century divergence from the models in winter are shown in Figure 5.19. The negative 2235 values (yellow) represent increased convergence from the 20th century, and positive values 2236 (green) reduced convergence from the 20th century. Thus a green strip north of a yellow 2237 strip implies a southward shift and vice-versa. The color shading shows the magnitude of 2238 difference, with darker colors representing large difference and white shading representing no 2239 difference. Four models suggest a southward shift in the WACZ (CCSM, CNRM, MPI and 2240 UKMO; Figures 5.19a,b,i,k) while three models suggest a northward shift (IPSL, IAP, GFDL; 2241 Figures 5.19c,e–f). Two models show very little difference between 21st and 20th century 2242 runs (GISS, MRI; Figures 5.19d,j). Both MIROC simulations (MIROC-h and MIROC-m; 2243 Figures 5.19g-h) show less divergence in 20th century than in simulation over most of the 2244 domain. Shifts in mean position could indicate a trend, or a discontinuity or shift in model 2245 state between simulations. We further evaluate observe trends later in this section. 2246

Next we examine the geographical placement of the WACZ COA on a year to year basis 2247 by coupling W_{ϕ} and W_{λ} in Figure 5.20. Information about the mean state, spread and 2248 skewness of W_{ϕ} is shown in Table 5.7. Models place the COA generally in the Niger River 2249 basin, on average just to the west of the meeting of the Niger and the Benue River. There are 2250 differences between models in placement of the WACZ with the MIROC-mres model placing 2251 it near to the Atlantic Ocean (Figure 5.20g) and the IPSL model placing the WACZ just 2252 south of the Sahel (Figure 5.20f). This range in latitudinal placement is also seen in Figure 2253 5.21. The horizontal lines represent the maximum and minimum values of the median of 2254 winter means of W_{ϕ} in all model runs over the period 2000 to 2100. IPSL and MIROC-mres 2255

²²⁵⁶ are shown to be north and south outliers respectively.

The time-series of W_{ϕ} in each model run is shown in Figure 5.22, for comparison the 2257 time-series of W_{ϕ} from reanalysis is shown in Figure 5.23. In 20th century reanalysis we note 2258 more cohesion between reanalysis products in time and less inter-annual variability than in 2259 summer. In winter there is less evidence for decadal variability than in summer, with values 2260 in general declining from the 1960's into the drought period of the late 1970's and 1980's, but 2261 no northward recovery into the 1990's and 2000's or other oscillations evident. 21st century 2262 model runs consistently produce less inter-annual variability than the 20th century and also 2263 produce less variability than 20th century reanalysis. We note that while NCEP reanalysis 2264 had a range of 2 degrees from 1980 to 2000, few 21st century models replicate this degree 2265 of variability. It is unclear if this reduction in variability is a result of model errors or are 2266 driven by changes in climate. 2267

A northward progression in the position of W_{ϕ} is visible in Figure 5.22. To assess the 2268 significance of this trend we perform a bootstrap analysis on the linear regression of W_{ϕ} , 2269 the means and standard deviations of which are shown in Figure 5.24. Ten of eleven models 2270 indicate a northward trend in time as shown in Figures 5.25 and 5.26 and Table 5.27. Of 2271 the ten models with a northward trend all ten are significant at an alpha of 10% and nine 2272 of ten are significance at an alpha of 2%. The significance of the trend is summarized in 2273 Figure 5.27. Again, similar to the summer means, the outlying model that does not indicate 2274 a northward trend is the lower-resolution GISS model. 2275

To summarize, in winter, models identify a north-south dipole as the first EOF mode suggesting that north south variability in the position of the WACZ is the dominant feature of 925 hPa divergence. Models produce less variability of W_{ϕ} in the 21st century than the 20th century on an annual timescale, but are consistent with 20th century reanalysis for decadal variability. Over the course of the 21st century W_{ϕ} is shown to significantly increase in time in ten of eleven models.

2282 5.3.2 Scenario: SRES-B1

We repeat the analysis of 925 hPa divergence and W_{ϕ} in scenario SRES-B1 over the period 2283 2000–2100. As discussed in Section 5.2 the SRES-B1 is a less extreme climate scenario, 2284 assuming that some actions are taken to mitigate the release of fossil fuels and in term 2285 greenhouse gas forcing on the climate system. Overall results for Scenario SRES-B1 are 2286 similar to SRES-A1B (as previously discussed) with only slight changes in strength, position 2287 and trends of divergence and W_{ϕ} observed. In this section we will present a shortened 2288 overview of the analysis and a comparison of SRES-B1 to SRES-A1B which has already 2289 been discussed. 2290

2291 Annual Means

Figure 5.28 shows the first loading pattern of the EOF of all months over the period 2000 to 2100. Similar to both 20th century reanalysis, AMIP model runs and SRESA1-B scenario runs the dominant feature is a north-south dipole that reflects the annual south to north migration of the WACZ. The relative magnitude of the pattern is inversely proportional to model resolution, that is to say high resolution models have lower relative magnitudes associated with the loading pattern.

The annual latitudinal migration of the WACZ is shown in Figure 5.29, which is a boxplot of W_{ϕ} . A smother annual cycle is observed compared to SRES-A1B in which a few models had two annual peaks, this double peak phenomena is not present in SRES-B1 (save GFDL model). SRES-B1 models show similarities to 20th century AMIP and reanalysis in that the fall southward migration appears to be more rapid than the spring northward migration.

The time-series of annual means of W_{ϕ} is shown in Figure 5.30. Similar to SRES-A1B the lack of inter-annual and decadal variability is clearly evident. Information of model median, interquartile range and skewness is shown in Table 5.9. Eight of ten models are south in their median position compared to their position in the 20th century, suggesting a southward shift in WACZ in this scenario relative to AMIP. Some weak trends appear, although they appear to be lower in magnitude than SRES-A1B. Table 5.10 shows a statistical overview of trends in annual mean via a bootstrap analysis of linear regression. Nine of ten models show a positive linear trend and positive correlation with time. Each of these nine models are significant at an alpha of 10% and seven of nine models are significant at an alpha of 2312 2%. We discuss trends in W_{ϕ} in the following sections.

2313 Summer Means

Figure 5.31 shows the first loading pattern of the EOF associated with mean summer 925 hPa divergence. The dominant pattern is a dipole associated with changes in phase of the WACZ from north to south years. We note the relative magnitudes are lower compared to SRES-A1B in CCSM and MIROCH-h (Figure 5.31a,h).

The long term mean of 925 hPa divergence is shown in Figure 5.32, the climatological 2318 statistics on W_{ϕ} are shown in Table 5.11. As expected an east to west band of divergence is 2319 evident across North Africa, centered about 18° N. As in both the 20th century and SRES-2320 A1b the strongest convergence is shifted toward central and eastern Africa. The median 232 summer position of W_{ϕ} is shown to be south of its position in AMIP, with a southward shift 2322 in position on average of -1.52° observed. In Figure 5.33 we present the difference of 20th 2323 century models from the AMIP scenario from the long term mean 925 hPa divergence in 2324 SRES-B1. Here green (positive values) north of yellow (negative values) suggests a southward 2325 shift in the WACZ from the 20th century to the 21st century, which is observed in most 2326 models. 2327

The position of the WACZ COA is shown in Figure 5.34, and boxplots of W_{ϕ} are shown in Figure 5.35. We note the COA varies in both latitudinal and longitudinal placement from model to model, similarly to both the 20th century AMIP scenario and the SRES-A1B scenario. The interquartile range of W_{ϕ} in Table 5.11 shows a reduction in comparison to SRES-A1B suggesting a reduction in near median inter-annual variability in SRES-B1. This reduction in inter-annual variance in the top and bottom quartiles is seen also in Figure

5.36 which shows the time-series of W_{ϕ} . A lack of decadal variability is evident, similar to 2334 SRES-A1B. A trend is also visible in some models, the significance of which we assess next. 2335 Bootstrapping on linear regression of W_{ϕ} is performed, the mean and standard devia-2336 tion of the boostrapped time-series are shown in Figure 5.37. Overall a northward trend 2337 is observed in the models, albeit reduced in magnitude and significance from SRES-A1B. 2338 Figure 5.38 shows the distribution of linear trends and Figure 5.39 shows the distribution 2330 of correlation with time and significance of trend shown in Figure 5.39p. Here in addition 2340 to the reduction in magnitude of the trend we note that the GISS model no longer shows 2341 a southward trend in this scenario, rather it shows a weak northward trend. The statistics 2342 associated with the trend analysis are seen in Table 5.12. Nine of ten models show a north-2343 ward trend in W_{ϕ} , although only eight of the nine models are significant at alpha equal to 2344 10% and only four of the eight are significant at the more stringent alpha equal to 2%. The 2345 significance of the linear regressions are summarized in Table 5.12. 2346

2347 Winter Means

The majority of models identify north to south changes in phase of the WACZ as the primary mode of variability in an EOF analysis of 925 hPa divergence as seen in Figure 5.41. As with SRES-A1B and AMIP there is an increase in relative magnitude over West Africa, relative to summer.

The climatology of 925 hPa divergence for winter means is shown in Figure 5.42. An 2352 east to west band of convergence is visible across North Africa, generally near to 8° N. Table 2353 5.13 shows the median, interquartile range and skewness of W_{ϕ} . Half of models (five of ten) 2354 show either no shift or a small northward shift in the position of the W_{ϕ} relative to the 2355 AMIP scenario. Only one model has a large southward shift in position (IAP), and only 2356 four models have a large northward shift in position (CNRM, IPSL, MIROC-h, MPI). These 2357 results are verified in Figure 5.43 which shows the varied shifts in mean divergence relative 2358 to the 20th century. 2359

The winter mean position of W_{ϕ} and W_{λ} are shown in Figure 5.44. North to south spread between models is evident, and summarized in Table 5.13, but is consistent with position in SRES-A1B. A graphical comparison between models is shown in Figure 5.45, with maximum and minimum of the median of the models shown in horizontal lines. MIROC-m is nearly always beneath the minimum median W_{ϕ} and IPSL is entirely outside of the maximum median W_{ϕ} . The median position and variability of W_{ϕ} is comparable to that seen in SRES-A1B.

The time-series of W_{ϕ} is shown in Figure 5.46, similar to SRES-A1B we note reduced 2367 inter-annual and decadal variability compared to AMIP. A slight trend is noticeable in some 2368 models, but less evident than in SRES-A1B. To assess the significance of the trend bootstrap-2369 ping was applied to linear regression analysis of W_{ϕ} . The means and standard deviations 2370 associated with the bootstrap are shown in Figure 5.47. While models suggest there could 237 be a weak northern trend in the data (Figure 5.48), the magnitude of the trend is reduced 2372 as is the correlation coefficient between W_{ϕ} and time (Figure 5.49). Significance of the trend 2373 is shown in Figure 5.50 and Table 5.14 relative to SRES-A1B. Seven of ten models show a 2374 positive phase that is significant at alpha of 2%, one of ten models (GISS) shows a south-2375 ward trend that is significant at an alpha of 10%. Overall less models indicate significant 2376 northward trends in SRES-B1 compared to SRES-A1B. 2377

2378 5.4 Discussion

2379 5.4.1 Model Specific Results

²³⁸⁰ By quantifying their bias in W_{ϕ} relative to reanalysis in the 20th century, in Chapter 4 ²³⁸¹ we ranked model performance in the AMIP scenario identifying models as performing well ²³⁸² (IPSL, MIROC-hres, MPI, CNRM), middling (MRI, GFDL, UKMO, CCSM, GISS) or ²³⁸³ poorly (MIROC-mres, IAP). Based on these findings the question arises if models ranked ²³⁸⁴ poor or strong project similar changes in the position of the WACZ in 21st century climate 2385 runs.

Median values of W_{ϕ} in each scenario and model run for the 21st century are shown 2386 in Table 5.15 for both summer and winter. Model biases appear to be consistent from 2387 AMIP to SRES-A1B and SRES-B1. For example, the poorly performing model MIROC-2388 mres represented a southern outlier in winter consistently in AMIP, SRES-A1B and SRES-2389 B1. Likewise, the poorly performing IAP represents the northern outlier in summer in all 2390 of AMIP, SRES-A1B and SRES-B1. Model biases relative to reanalysis appear to extend 2391 from AMIP into both SRES-A1B and SRES-B1 scenarios. With no observations of the 21st 2392 century available to compare to, it is more difficult to identify models that perform well. 2393 Therefore we can only identify models that behave similarly to other models and those that 2394 are outliers (as in the above case). As such we can clearly define IAP and MIROC-mres as 2395 outliers as they are clearly different than their companion models, as they were in the AMIP 2396 case as well. 2397

The IQR (inter-quartile range) represents the range in value of W_{ϕ} between the 75th and 2398 25th percentiles, and as such represents the spread of W_{ϕ} , although constrained to the middle 2399 50% of data. The IQR does not include any information about the upper and lower 25%2400 of data, data in which comprises the northernmost and southernmost years of W_{ϕ} . As such 2401 the IQR is not suitable to examine true inter-annual variability, as much of the geophysical 2402 impacts of W_{ϕ} are related to values outside of the middle 50%. As shown previously the 2403 variability of the extreme 25% of data is less than that observed in 20th century reanalysis. 2404 However, as seen in Table 5.16, variability of the middle 50% of data is generally larger in 2405 SRES-A1B and SRES-B1, than in AMIP. For our poorly performing model both IAP and 2406 MIROC-hres see at least doubling of the IQR from AMIP to both SRES scenario in summer, 2407 and doubling of the IQR for MIROC-hres in winter (IAP does not change greatly in winter). 2408 However this large increase (doubling or more) in IQR from AMIP to the SRES scenarios 2409 is not seen in any models identified as strong performers, suggesting that this phenomenon 2410 is limited to models that performed poorly in the 20th century and may be indicative of 2411

²⁴¹² weaker models.

The key finding of this work is the clear northward trend in W_{ϕ} in both winter and 2413 summer, which was strongest in Scenario SRES-A1B suggesting a strengthening of the trend 2414 with increased climate forcing. Table 5.17 shows a side by side comparison of linear trends 2415 in each of the GCMs for all scenarios. No relationship between model bias in position of W_{ϕ} 2416 with respect to reanalysis and linear trend in future scenarios is evident. The GISS model is 2417 the outlier in terms of linear trend suggesting a southward migration of the WACZ in three 2418 of four future scenarios; the GISS model was defined as a middle performing model in the 2419 AMIP scenario. The two poor-performance models indicate a northward trend (significant 2420 in three of four scenarios). Models that did poorly, in the middle or well placing the WACZ 2421 in the 20th century all show a significant northward movement of W_{ϕ} in the 21st century. 2422 In summary we examine the two models that did worst at placing the WACZ in the 20th 2423 century. These models exhibit large deviations from other models with placement of W_{ϕ} in 2424 the 21st century. The IQR of both the poorly performing models at least doubles, suggesting 2425 quite a bit of variability about the mean value, although we note a lack of variability in 2426 extreme values (few far-north and far-south years). However despite poor performance in 2427 mean and variability poorly performing models support the premise of northward migration 2428

²⁴²⁹ of W_{ϕ} in the 21st century.

²⁴³⁰ 5.4.2 Model Shift in Divergence

Tables 5.3, 5.5, 5.9, and 5.11 all highlight the large southern shift in model divergence from AMIP to SRES-A1B and SRES-B1 in both annual and summer means compared to the 2432 20th century. The observed shift was also seen in DJFM of SRES-A1B and to a lesser degree 2434 in SRES-B1. Significant northward trends were observed in both scenarios in summer and 2435 annual W_{ϕ} during the 21st century. The superposition of this trend suggests W_{ϕ} was at its 2436 most southern point at the start of the 21st century, and that its difference from AMIP (20th 2437 century) was largest at this point as well. Visual comparison between the position of W_{ϕ} in ²⁴³⁸ 2000 in reanalysis (e.g. Figure 5.12) to the position of W_{ϕ} in 2000 in any GCM simulations ²⁴³⁹ of the 21st century (e.g. Figure 5.11) confirms this abrupt southward shift, followed by a ²⁴⁴⁰ northward trend.

The observed shift is likely due to differences between coupled and non-coupled ocean-2441 atmosphere in CMIP-SRES and AMIP respectively. In the AMIP scenarios the sea surface 2442 temperature and sea ice coverage are prescribed. Models then are free to produce an atmo-2443 spheric response to the oceanic conditions. In both the SRES-A1B and SRES-B1 the models 2444 are based on the same physics and parameterizations but predict their own SST and sea 2445 ice coverage. In both scenarios, in the year 2000 greenhouse gas forcing must be similar to 244F greenhouse gas forcing at the end of the 20th century AMIP scenario. The discontinuity in 2447 the placement of the WACZ from the end of the 20th century to the start of the 21st century, 2448 which is of the order of 2.5° in extreme cases, is due to model differences in the treatment 2449 of SST and ocean processes. 2450

²⁴⁵¹ 5.4.3 Implications for Mineral Dust Emission and Transport

In Chapters 2 and 3 the critical relationship between W_{ϕ} and mineral dust transport to Barbados is shown. W_{ϕ} drives dust transport by controlling the strength and direction of wind over critical dust hot spot emission regions in the Sahel and Sahara in both winter and summer seasons. We also found that in summer W_{ϕ} impacts dust transport via precipitation and in turn soil moisture and vegetative cover. The relationship between W_{ϕ} and mineral dust at Barbados holds on inter-annual timescales by examining seasonal means of dust load and seasonal means of W_{ϕ} .

Model predictions for the 21st century show reduced inter-annual variability in W_{ϕ} compared to 20th century reanalysis products. This reduction in variability is shown in both 2461 21st century scenario SRES-A1B and SRES-B1 and holds for annual, winter and summer 2462 means. Model runs in the AMIP scenario also showed reduced inter-annual variability, and 2463 showed no skill in reproducing the time-series of W_{ϕ} from reanalysis. As such we conclude that the models likely are unable to predict inter-annual or even decadal changes in mineral dust emission and transport. Additionally the models' inability to replicate temporal variability in the 20th century suggests that current models may lack some key physical processes necessary to accurately predict the position of the WACZ, and also climate-dust feedbacks are excluded in CMIP3 models.

Prior work has suggested an expansion of the Hadley Circulation as the Earth's climate 2469 warms which could have implications for the positioning of the WACZ with time and in 2470 turn mineral dust emission and transport. A northward trend is predicted in the annual 2471 and summer position of the WACZ in the 21st century in this study. Northward migration 2472 of the WACZ could lead to reduced transport of mineral dust to Barbados, as a result of 2473 shifting of surface winds away from key source regions and increased precipitation in general 2474 in the Sahel. Such a projection would be of low confidence as source regions could experience 2475 significant change over the coming century due to climatic shifts and anthropogenic land uses 2476 changes with time. To estimate how northward migration of the WACZ could impact mineral 2477 dust transport we first establish the relationship between the WACZ and mineral dust in 2478 Barbados in the 20th century via linear regression. For a one degree northward shift mineral 2479 dust at Barbados is reduced by by $-6.52 \ \mu g m^{-3} - 3.75 \ \mu g m^{-3}$ and for summer and winter 2480 respectively. Next we estimate the change in the position of the WACZ over the 21st century 2481 in SRES-A1B and SRES-B1. To do so we calculate the trend of the median seasonal value of 2482 every model output (shown in Tables 5.6, 5.8, 5.12, 5.14). To assess uncertainty of the trend 2483 we repeat this calculation for the 2.5 and 97.5 percentiles to create a 95% confidence interval 2484 about our projection. To calculate the change in dust load the trends are multiplied by 100 2485 years to calculate the 100 year change in WACZ position, and then multiplied through by 2486 the dust per degree of WACZ shift as calculated in the 20th century. Results are shown in 2487 Table 5.18. For SRES-A1b we expect a change of $-3.8 \ \mu g m^{-3}$ in summer $(-3.0 \ \mu g m^{-3}$ to 2488 $-4.7 \ \mu g m^{-3}$ confidence interval) and a change of $-2.2 \ \mu g m^{-3}$ in winter ($-1.3 \ \mu g m^{-3}$ to 2489 $-2.4 \ \mu g m^{-3}$ confidence interval). The predicted change in mean dust load from the end of 2490

the 20th century to the 21st century is of the same order of magnitude as the inter-annual 2493 variance in the 20th century, or put another way, the predicted reduction is expected to 2492 be nearly as large as the current year-to-year changes in dust load. As previously noted 2493 SRES-B1 produced smaller northward trends than SRES-A1b, so reductions in dustload 2494 are less; $-1.9 \ \mu g m^{-3}$ in summer (-1.1 $\mu g m^{-3}$ to -2.7 $\mu g m^{-3}$ confidence interval) and 2495 $-1.5 \ \mu g m^{-3}$ in winter (-1.0 $\mu g m^{-3}$ to $-2.0 \ \mu g m^{-3}$ confidence interval). A fully coupled 2496 vegetation-land surface and climate model would be necessary to determine the magnitude of 2497 change of mineral dust transport in the 21st century, but here we show based on shifts in the 2498 WACZ expected changes in mineral dust emission and transport on the order of inter-annual 2490 variability of mineral dust in the 20th century. 2500

2501 5.5 Conclusions

This work examined changes in divergence over West Africa at 925 hPa in the 21st century 2502 in GCMs from the CMIP3 experiment for the SRES-A1B and SRES-B1 scenarios. Climate 2503 models captured the annual migration of the WACZ and identified north-south shifts in 2504 the annual position of the WACZ as the dominant mode of variability. However models 2505 showed reduced inter-annual and decadal variability of W_{ϕ} particularly in the north and south 2506 quartiles, quartiles previously identified as playing key roles in determining precipitation, 2507 wind and in turn mineral dust transport. A southward shift of up to 2.5° in W_{ϕ} from the 2508 AMIP/20th century to the 21st century GCM runs was identified. Northward migration of 2509 W_{ϕ} over the 21st century is seen in annual, winter and summer means. 2510

Overall GCM output for the 21st century showed reduced variability compared to 20th century reanalysis. Reductions in inter-annual variability in W_{ϕ} we shown for summer and winter means. Additionally no decadal variability in W_{ϕ} was seen, in contrast to 20th century in which decadal shifts in W_{ϕ} were observed.

²⁵¹⁵ Compared with the 20th century, a southward shift in the WACZ is evident in both 21st

century scenarios. Concurrently over the 21st century a clear northward trend is evident. 2516 A northward trend in time combined with an overall southward bias suggests the largest 2517 southward displacement occurred nearest to the year 2000 and decreased over the 21st cen-2518 tury. Such a large discontinuity cannot be the result of changes in greenhouse gas forcing (as 2519 forcing near to 2000 are identical). The apparent conflict must be related to oceanic model 2520 variability. In the AMIP 20th century scenario the models were forced with prescribed SST 2521 and sea ice conditions based on observation, in contrast in the 21st century models were 2522 free to develop their own SST conditions. As weak northward trends were observed in the 2523 20th century summer and annual means it appears the northward trend is robust over varied 2524 scenarios, and the southward discontinuity near the year 2000 a model artifact. 2525

Northward trends in W_{ϕ} were seen in annual, winter and summer means and significance of the trends was assessed by bootstrapping. Northward trends were larger in SRES-A1B than SRES-B1; the median linear trend of W_{ϕ} was $5.88 \times 10^{-3} \,^{\circ}\text{yr}^{-1}$ and $5.48 \times 10^{-3} \,^{\circ}\text{yr}^{-1}$ for summer and winter respectively in SRES-A1B and $2.92 \times 10^{-3} \,^{\circ}\text{yr}^{-1}$ and $3.83 \times 10^{-3} \,^{\circ}\text{yr}^{-1}$ for summer and winter respectively in SRES-B1.

²⁵³¹ While reductions in inter-annual and decadal variability render predictions of dust load ²⁵³² moot for the 21st century, estimates for changes in dust load at Barbados over the 21st ²⁵³³ century were performed. In SRES-A1b mean dust load is expected by $-3.8 \ \mu g m^{-3}$ and ²⁵³⁴ $-2.2 \ \mu g m^{-3}$ in summer and winter respectively. For SRES-B1 expected reductions in dust ²⁵³⁵ load were $-1.9 \ \mu g m^{-3}$ and $-1.5 \ \mu g m^{-3}$ in summer and winter respectively. This is consistent ²⁵³⁶ with northward migration of the WACZ leading to reduced dust emission and transport to ²⁵³⁷ Barbados.

Prior work has suggested a poleward expansion of the Hadley Cell as the climate warms [Fu et al., 2006; Hu and Fu, 2007; Frierson et al., 2007; Gastineau et al., 2008; Johanson and Fu, 2009]. Our work focuses on the equatorial ascending branch of the Hadley Cell over West Africa, the WACZ. Our work in general supports the hypothesis of poleward expansion of the Hadley cell on annual, summer and winter means as we identify clear northward trends in the latitudinal position over the 21st century. We note that prior work in general focuses on the
northern extent of the Hadley Cell, which is subsidence generally well north of the WACZ.
Our work contrasts the findings of *Gastineau et al.* [2008] who found a southward migration
in the equatorial-ascending branch of the Hadley Cell by examining 500 hPa streamfunction
in GCM simulations, as here we find a northward migration of the ascending branch in the
21st century in divergence nearer to the surface.

The results of this project raise a number of questions that could be addressed as future 2549 work. We hypothesize that changes in the WACZ would reduce transport of dust to Barbados 2550 in the 21st century. A detailed land-surface model could be coupled to a GCM to assess how 2551 long-term shifts in the position of the WACZ would affect surface conditions, which could 2552 lead to marked changes in mineral dust emission. We suggest that the large shift in position 2553 of the WACZ from the end of AMIP to the start of the SRES scenarios is attributable 2554 to changes in ocean conditions. A study should be performed to analyze differences in 2555 SST in GCMs and note how the WACZ responds to such changes. Lastly, here we note a 2556 northward trend in the position of the ascending branch of the Hadley Cell. The center of 2557 action approach could be inverted to examine movement of areas of near surface divergence 2558 associated with the poleward descending branch of the Hadley Cell. It would be of interest 2559 to note if changes in position of the divergence zone are related to changes in the position 2560 of the convergence zone. 2561



Figure 5.1: Time-series of emissions of greenhouse gases in multiple SRES Scenarios. Image from *Solomon et al.* [2007].



Figure 5.2: First mode of the EOF of 925 hPa divergence of all months in the period 2000–2100 for SRESA1-B. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.3: Annual south to north migration of WACZ as illustrated by boxplot of monthly means of W_{ϕ} over the period 2000–2100 for SRES-A1b. Red center line represents median, blue lines above and below median represent the 75th and 25th percentiles respectively. Red crosses represents outliers. Black lines extend 1.5 times the interquartile range out from the median. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.4: Time-series of annual means of W_{ϕ} over the period 2000–2100 for SRES-A1b.



Figure 5.5: Time-series of annual means of W_{ϕ} over the period 1948 to 2011 in reanalysis.



Figure 5.6: First mode of the EOF of 925 hPa divergence during summer (JJAS) in the period 2000–2100 for SRESA1-B. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.7: Long-term mean of 925 hPa divergence during summer (JJAS) in the period 2000–2100 for SRESA1-B. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.8: Long-term mean divergence over the period 2000–2100 for SRESA1-B minus long term mean divergence over the period 1978–2002 for AMIP. Divergence calculated at 925 hPa for summer months. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.9: Position of WACZ as determined by W_{λ} and W_{ϕ} . Each summer mean position is plotted. The quarter of years that are furthest north are blue diamonds, the quarter of years that are furthest south are red diamonds and the interquartile range are black diamonds. COA plotted over the period 2000–2100 for SRESA1-B. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.10: Boxplot of summer means of W_{ϕ} over the period 2000–2100 for SRES-A1b. Red center line represents median, blue lines above and below median represent the 75th and 25th percentiles respectively. Red crosses represents outliers. Black lines extend 1.5 times the interquartile range out from the median.



Figure 5.11: Time-series of summer means of W_{ϕ} over the period 2000–2100 for SRES-A1b.



Figure 5.12: Time-series of summer means of W_{ϕ} over the period 1948 to 2011 in reanalysis.



Figure 5.13: Mean (x-axis) and standard deviation (y-axis) of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d) GISS, (e) IAP, (f) IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.14: Linear trend of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.15: Pearson product moment correlation coefficients between W_{ϕ} and time in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.16: Probability to reject the hypothesis that there is a significant linear trend of W_{λ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.17: First mode of the EOF of 925 hPa divergence during winter (DJFM) in the period 2000–2100 for SRESA1-B. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.18: Long term mean of 925 hPa divergence during winter (DJFM) in the period 2000–2100 for SRESA1-B. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d) GISS, (e) IAP, (f) IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.19: Long term mean divergence over the period 2000–2100 in winter (DJFM) for SRESA1-B minus long term mean divergence over the period 1978–2002 for AMIP. Divergence calculated at 925 hPa for winter months. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d) GISS, (e) IAP, (f) IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.20: Position of WACZ as determined by W_{λ} and W_{ϕ} . Each winter mean position is plotted. The quarter of years that are furthest north are blue diamonds, the quarter of years that are furthest south are red diamonds and the interquartile range are black diamonds. COA plotted over the period 2000–2100 for SRESA1-B. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.21: Boxplot of winter means of W_{ϕ} over the period 2000–2100 for SRES-A1b. Red center line represents median, blue lines above and below median represent the 75th and 25th percentiles respectively. Red crosses represents outliers. Black lines extend 1.5 times the interquartile range out from the median.



Figure 5.22: Time-series of winter means of W_{ϕ} over the period 2000–2100 for SRES-A1b.



Figure 5.23: Time-series of winter means of W_{ϕ} over the period 1948 to 2011 in reanalysis.



Figure 5.24: Mean (x-axis) and standard deviation (y-axis) of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario for winter (DJFM). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.


Figure 5.25: Linear trend of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario in winter (DJFM). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.26: Pearson product moment correlation coefficients between W_{ϕ} and time in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario in winter (DJFM). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.27: Probability to reject the hypothesis that there is a significant linear trend of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRES-A1B scenario in winter (DJFM). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.28: First mode of the EOF of 925 hPa divergence of all months in the period 2000–2100 for SRESB1. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI



Figure 5.29: Annual south to north migration of WACZ as illustrated by boxplot of monthly means of W_{ϕ} over the period 2000–2100 for SRES-A1b. Red center line represents median, blue lines above and below median represent the 75th and 25th percentiles respectively. Red crosses represents outliers. Black lines extend 1.5 times the interquartile range out from the median. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI



Figure 5.30: Time-series of annual means of W_{ϕ} over the period 2000–2100 for SRES-A1b.



Figure 5.31: First mode of the EOF of 925 hPa divergence during summer (JJAS) in the period 2000–2100 for SRESB1. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI and (k) UKMO-HADGEM.



Figure 5.32: Long term mean of 925 hPa divergence during summer (JJAS) in the period 2000–2100 for SRESB1. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.33: Long term mean divergence over the period 2000–2100 for SRESB1 minus long term mean divergence over the period 1978–2002 for AMIP. Divergence calculated at 925 hPa for summer months. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.34: Position of WACZ as determined by W_{λ} and W_{ϕ} . Each summer mean position is plotted. The quarter of years that are furthest north are blue diamonds, the quarter of years that are furthest south are red diamonds and the interquartile range are black diamonds. COA plotted over the period 2000–2100 for SRESb1. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.35: Boxplot of summer means of W_{ϕ} over the period 2000–2100 for SRES-A1b. Red center line represents median, blue lines above and below median represent the 75th and 25th percentiles respectively. Red crosses represents outliers. Black lines extend 1.5 times the interquartile range out from the median.



Figure 5.36: Time-series of summer means of W_{ϕ} over the period 2000–2100 for SRES-A1b.



Figure 5.37: Mean (x-axis) and standard deviation (y-axis) of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.38: Linear trend of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.39: Pearson product moment correlation coefficients between W_{ϕ} and time in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.40: Probability to reject the hypothesis that there is a significant linear trend of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario in summer (JJAS). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.41: First mode of the EOF of 925 hPa divergence during winter (DJFM) in the period 2000–2100 for SRESB1. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.42: Long term mean of 925 hPa divergence during winter (DJFM) in the period 2000–2100 for SRESB1. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.43: Long term mean divergence over the period 2000–2100 for SRESB1 minus long term mean divergence over the period 1978–2002 for AMIP. Divergence calculated at 925 hPa for winter months. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.44: Position of WACZ as determined by W_{λ} and W_{ϕ} . Each winter mean position is plotted. The quarter of years that are furthest north are blue diamonds, the quarter of years that are furthest south are red diamonds and the interquartile range are black diamonds. COA plotted over the period 2000–2100 for SRESb1. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROChires, (i) MPI. Fourth row: (j) MRI.



Figure 5.45: Boxplot of winter means of W_{ϕ} over the period 2000–2100 for SRES-A1b. Red center line represents median, blue lines above and below median represent the 75th and 25th percentiles respectively. Red crosses represents outliers. Black lines extend 1.5 times the interquartile range out from the median.



Figure 5.46: Time-series of winter means of W_{ϕ} over the period 2000–2100 for SRES-A1b.



Figure 5.47: Mean (x-axis) and standard deviation (y-axis) of W_{λ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario. Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.48: Linear trend of W_{ϕ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario in winter (DJFM). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.49: Pearson product moment correlation coefficients between W_{ϕ} and time in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario in winter (DJFM). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.



Figure 5.50: Probability to reject the hypothesis that there is a significant linear trend of W_{λ} in 10,000 resamplings via bootstrapping over period 2001–2099 in SRESB1 scenario in winter (DJFM). Top row: (a) CCSM, (b) CNRM, (c) GFDL. Second row: (d)GISS, (e)IAP, (f)IPSL Third row: (g) MIROC-mres, (h) MIROC-hires, (i) MPI. Fourth row: (j) MRI.

Table 5.1: Summary of key information on CMIP GCMs used, including acronyms, resolution, start and end times for SRES-A1b and SRES-B1 scenarios, and number of complete years simulated. Additional information on models available in prior chapter. UKMO model only available for SRES-A1b.

Model Acronym	Resolution	Start	End	Total Years
CCSM	$1.40625^{\circ} \times 1.40625^{\circ}$	Jan 2000	Dec 2099	100
CNRM	$2.8125^{\circ} \times \ 2.8125^{\circ}$	Jan 2000	Dec 2099	100
GFDL	$2^{\circ} \times 2.5^{\circ}$	Jan 2001	Dec 2100	100
GISS	$4^{\circ} \times 5^{\circ}$	Jan 2000	Dec 2099	100
IAP	$3^{\circ} \times 2.8125^{\circ}$	Jan 2000	Dec 2099	100
IPSL	$2.5^{\circ} \times \ 3.75^{\circ}$	Jan 2000	Dec 2100	101
MIROC (mres)	$2.8125^{\circ} \times \ 2.8125^{\circ}$	Jan 2001	Dec 2100	100
MIROC (hires)	$1.125^{\circ} \times 1.125^{\circ}$	Jan 2001	Dec 2100	100
MPI	$1.875^{\circ} \times 1.875^{\circ}$	Jan 2001	Dec 2100	100
MRI	$2.8125^{\circ} \times \ 2.8125^{\circ}$	Jan 2001	Dec 2100	100
UKMO	$1.25^{\circ} \times 1.875^{\circ}$	Jan 2000	Nov 2099	99

Table 5.2: Overview of socioeconomic conditions contributing to SRES emission scenarios used. Adapted after *Solomon et al.* [2007].

	Scenario A1B	Scenario B1
Population growth	low	low
GDP growth	very high	high
Energy use	very high	low
Land- use changes	low	high
Resource availability	medium	low
Pace and direction of technological	rapid	medium
change favoring	balanced	efficiency and dematerialization

Model	Median	IQR	Skewness	AMIP Median	SRESA1 - AMIP
CCSM	12.88	4.71	0.12	14.05	-1.17
CNRM	12.39	6.56	-0.20	12.07	0.32
GFDL	12.99	4.48	-0.14	14.48	-1.48
GISS	13.19	4.28	-0.45	15.15	-1.96
IAP	13.36	7.61	-0.04	14.82	-1.45
IPSL	13.10	4.78	0.14	13.20	-0.10
MIROCm	11.91	6.36	-0.44	12.63	-0.72
MIROCh	13.74	3.65	-0.41	14.24	-0.50
MPI	12.73	5.52	-0.21	12.70	0.03
MRI	12.36	5.13	-0.13	11.86	0.50
UKMO	11.86	3.29	-0.42	12.60	-0.74
remean	13.03	5.22	-0.25	13.40	-0.36
remedian	13.02	5.01	-0.27	13.36	-0.33

Table 5.3: Overview of W_{ϕ} for annual means in SRESA1 over period 2001–2099.

Table 5.4: Summary of statistics associated with the bootstrap analysis of linear trend of annual means for SRESA1 over the period 2001 to 2099. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . $r_{0.5}$ is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a *t*-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
CCSM	5.57×10^{-3}	0.28	0.58	0.44	5.88×10^{-7}
CNRM	4.21×10^{-3}	0.25	0.58	0.43	4.37×10^{-6}
GFDL	7.92×10^{-3}	0.46	0.72	0.60	8.60×10^{-15}
GISS	-3.53×10^{-3}	-0.44	-0.08	-0.26	2.89×10^{-3}
IAP	7.57×10^{-3}	0.56	0.77	0.67	2.67×10^{-20}
IPSL	3.93×10^{-3}	0.39	0.68	0.56	1.49×10^{-10}
MIROCm	7.80×10^{-3}	0.58	0.77	0.68	3.28×10^{-19}
MIROCh	7.19×10^{-3}	0.75	0.88	0.82	8.16×10^{-46}
MPI	6.22×10^{-3}	0.47	0.72	0.61	1.60×10^{-14}
MRI	4.52×10^{-3}	0.29	0.62	0.47	2.30×10^{-8}
UKMO	4.29×10^{-3}	0.43	0.69	0.57	1.59×10^{-13}
remean	4.64×10^{-3}	0.82	0.92	0.87	4.97×10^{-63}
remedian	5.17×10^{-3}	0.69	0.85	0.78	7.84×10^{-33}

Model	Median	IQR	Skewness	AMIP Median	SRESA1 - AMIP
CCSM	15.84	0.88	0.08	17.07	-1.22
CNRM	15.25	0.61	-0.27	17.79	-2.53
GFDL	14.94	0.83	-0.25	17.80	-2.85
GISS	14.67	1.00	0.14	18.49	-3.81
IAP	17.71	0.63	-0.48	19.32	-1.61
IPSL	16.25	0.43	0.15	16.74	-0.48
MIROCm	14.36	0.67	0.03	15.05	-0.68
MIROCh	15.42	0.44	-0.39	16.61	-1.18
MPI	14.82	0.57	-0.01	17.09	-2.26
MRI	15.36	0.47	-0.14	15.46	-0.09
UKMO	13.17	0.53	0.46	15.04	-1.86
remean	15.29	0.29	-0.20	16.90	-1.60
remedian	15.24	0.31	-0.36	16.9	-1.65

Table 5.5: Overview of W_{ϕ} for summer means in SRESA1 over period 2001–2099.

Table 5.6: Summary of statistics associated with the bootstrap analysis of linear trend of summer means in SRESA1 over the period 2001 to 2099. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . $r_{0.5}$ is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a t-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
CCSM	3.57×10^{-3}	-0.03	0.35	0.17	4.41×10^{-2}
CNRM	5.88×10^{-3}	0.20	0.54	0.38	4.96×10^{-5}
GFDL	7.83×10^{-3}	0.20	0.54	0.38	2.14×10^{-5}
GISS	-2.99×10^{-3}	-0.31	0.09	-0.12	8.89×10^{-2}
IAP	9.27×10^{-3}	0.36	0.65	0.52	9.25×10^{-11}
IPSL	3.57×10^{-3}	0.18	0.53	0.37	4.67×10^{-5}
MIROCm	1.11×10^{-2}	0.62	0.80	0.72	1.81×10^{-22}
MIROCh	4.83×10^{-3}	0.28	0.59	0.45	2.97×10^{-7}
MPI	8.01×10^{-3}	0.42	0.67	0.60	7.98×10^{-10}
MRI	2.71×10^{-3}	0.56	0.41	0.24	2.11×10^{-2}
UKMO	6.42×10^{-3}	0.36	0.63	0.51	6.72×10^{-9}
remean	5.49×10^{-3}	0.71	0.86	0.79	3.45×10^{-35}
remedian	5.88×10^{-3}	0.56	0.78	0.68	1.51×10^{-19}

Model	Median	IQR	Skewness	AMIP Median	SRESA1 - AMIP
CCSM	10.35	0.72	-0.15	10.19	0.15
CNRM	8.42	0.57	-0.19	7.37	1.04
GFDL	9.92	0.52	-0.10	9.72	0.20
GISS	9.63	0.70	0.31	9.79	-0.15
IAP	8.49	0.54	0.08	9.69	-1.19
IPSL	10.79	0.30	-0.07	8.83	1.95
MIROCm	7.21	0.64	-0.08	7.69	-0.48
MIROCh	10.69	0.56	0.39	10.11	0.57
MPI	8.36	0.57	-0.71	7.43	0.92
MRI	9.20	0.76	0.18	8.70	0.49
UKMO	8.96	0.41	-0.52	9.15	-0.19
remean	9.28	0.29	-0.15	8.99	0.28
remedian	9.28	0.38	0.19	9.14	0.14

Table 5.7: Overview of W_{ϕ} for winter means in SRESA1 over period 2001–2099.

Table 5.8: Summary of statistics associated with the bootstrap analysis of linear trend of winter means in SRESA1 over the period 2001 to 2099. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . $r_{0.5}$ is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a t-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
CCSM	7.10×10^{-3}	0.21	0.57	0.40	1.57×10^{-6}
CNRM	4.43×10^{-3}	0.10	0.44	0.27	9.02×10^{-4}
GFDL	4.67×10^{-3}	0.09	0.46	0.28	2.32×10^{-4}
GISS	-2.42×10^{-3}	-0.29	0.08	-0.11	1.11×10^{-1}
IAP	4.76×10^{-3}	0.14	0.51	0.33	2.51×10^{-4}
IPSL	4.06×10^{-3}	0.29	0.61	0.46	1.18×10^{-8}
MIROCm	5.60×10^{-3}	0.16	0.49	0.33	1.35×10^{-4}
MIROCh	9.67×10^{-3}	0.52	0.75	0.64	2.46×10^{-17}
MPI	5.73×10^{-3}	0.15	0.51	0.34	1.10×10^{-4}
MRI	7.97×10^{-3}	0.24	0.57	0.42	2.45×10^{-7}
UKMO	4.22×10^{-3}	0.12	0.48	0.31	1.19×10^{-4}
remean	4.90×10^{-3}	0.46	0.77	0.68	3.55×10^{-20}
remedian	5.48×10^{-3}	0.45	0.71	0.59	3.71×10^{-14}

Model	Median	IQR	Skewness	AMIP Median	SRESB1 - AMIP
CCSM	12.98	5.01	0.05	14.05	-1.06
CNRM	12.44	6.56	-0.21	12.07	0.37
GFDL	12.91	4.56	-0.14	14.48	-1.57
GISS	14.32	5.11	-0.46	15.15	-0.83
IAP	13.19	7.67	-0.03	14.82	-1.63
IPSL	13.12	4.54	0.12	13.20	-0.08
MIROCm	11.80	5.93	-0.51	12.63	-0.83
MIROCh	13.74	3.65	-0.41	14.24	-0.50
MPI	12.61	5.29	-0.25	12.70	-0.08
MRI	12.33	5.27	-0.12	11.86	0.47
remean	13.11	5.42	-0.23	13.51	-0.39
remedian	13.06	5.22	-0.25	13.53	-0.47

Table 5.9: Overview of W_{ϕ} for annual means in SRESB1 over period 2001–2099.

Table 5.10: Summary of statistics associated with the bootstrap analysis of linear trend of annual means in SRESB1 over the period 2001 to 2099. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . r_{0.5} is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a t-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
CCSM	1.76×10^{-3}	-0.06	0.33	0.14	4.57×10^{-2}
CNRM	1.52×10^{-3}	-0.06	0.36	0.16	4.53×10^{-2}
GFDL	2.59×10^{-3}	0.06	0.41	0.24	3.29×10^{-3}
GISS	-1.27×10^{-3}	-0.28	0.07	-0.11	9.26×10^{-2}
IAP	2.92×10^{-3}	0.23	0.56	0.40	1.30×10^{-4}
IPSL	2.04×10^{-3}	0.19	0.52	0.37	1.61×10^{-5}
MIROCm	6.79×10^{-3}	0.37	0.61	0.53	3.01×10^{-10}
MIROCh	7.20×10^{-3}	0.75	0.88	0.82	3.39×10^{-46}
MPI	3.80×10^{-3}	0.25	0.58	0.43	3.56×10^{-7}
MRI	3.51×10^{-3}	0.25	0.57	0.42	3.15×10^{-6}
remean	3.09×10^{-3}	0.68	0.86	0.79	3.97×10^{-35}
remedian	3.36×10^{-3}	0.55	0.78	0.67	1.06×10^{-19}

Model	Median	IQR	Skewness	AMIP Median	SRESB1 - AMIP
CCSM	15.86	0.80	-0.04	17.07	-1.20
CNRM	15.17	0.51	-0.15	17.79	-2.61
GFDL	15.01	0.80	-0.26	17.80	-2.79
GISS	16.28	0.75	0.14	18.49	-2.20
IAP	17.50	0.53	0.02	19.32	-1.81
IPSL	16.15	0.34	0.10	16.74	-0.58
MIROCm	14.22	0.60	-0.38	15.05	-0.82
MIROCh	15.42	0.44	-0.39	16.61	-1.18
MPI	14.66	0.62	-0.08	17.09	-2.43
MRI	15.25	0.52	0.30	15.46	-0.20
remean	15.54	0.19	0.02	17.07	-1.52
remedian	15.47	0.30	0.38	17.13	-1.66

Table 5.11: Overview of W_{ϕ} for summer means in SRESB1 over period 2001–2099.

Table 5.12: Summary of statistics associated with the bootstrap analysis of linear trend of summer means in SRESB1 over the period 2001 to 2099. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . r_{0.5} is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a t-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
CCSM	-7.10×10^{-5}	-0.21	0.17	-0.02	2.12×10^{-1}
CNRM	1.96×10^{-3}	-0.01	0.39	0.20	8.86×10^{-2}
GFDL	2.56×10^{-3}	-0.07	0.30	0.12	7.99×10^{-2}
GISS	2.61×10^{-3}	-0.06	0.32	0.13	8.47×10^{-2}
IAP	3.97×10^{-4}	-0.17	0.23	0.03	2.10×10^{-1}
IPSL	2.56×10^{-3}	0.09	0.46	0.28	1.51×10^{-3}
MIROCm	5.43×10^{-3}	0.19	0.53	0.37	1.60×10^{-5}
MIROCh	4.84×10^{-3}	0.27	0.59	0.44	2.89×10^{-7}
MPI	1.78×10^{-3}	-0.11	0.29	0.10	7.89×10^{-2}
MRI	3.43×10^{-3}	0.09	0.46	0.28	1.05×10^{-3}
remean	2.55×10^{-3}	0.34	0.64	0.50	9.42×10^{-9}
remedian	2.92×10^{-3}	0.26	0.57	0.43	1.17×10^{-6}

Model	Median	IQR	Skewness	AMIP Median	SRESB1 - AMIP
CCSM	10.11	0.56	-0.01	10.19	-0.08
CNRM	8.34	0.54	0.25	7.37	0.97
GFDL	9.77	0.46	0.35	9.72	0.04
GISS	10.10	0.87	-0.25	9.79	0.31
IAP	8.38	0.44	0.45	9.69	-1.31
IPSL	10.67	0.33	0.40	8.83	1.84
MIROCm	7.29	0.66	0.77	7.69	-0.40
MIROCh	10.69	0.56	0.39	10.11	0.57
MPI	8.31	0.56	0.01	7.43	0.87
MRI	9.13	0.57	0.39	8.70	0.42
remean	9.29	0.24	0.06	8.95	0.33
remedian	9.30	0.30	0.36	9.12	0.18

Table 5.13: Overview of W_{ϕ} for winter means in SRESB1 over period 2001–2099.

Table 5.14: Summary of statistics associated with the bootstrap analysis of linear trend of winter means in SRESB1 over the period 2001 to 2099. Trend_{0.5} is the median of the linear trend (°yr⁻¹). $r_{\rm min}$ and $r_{\rm min}$ are the 95 % confidence interval of the correlation between W_{ϕ} . r_{0.5} is the median correlation coefficient. $p_{0.5}$ is the median probability to reject the hypothesis that a trend exists using a t-test.

	Trend _{0.5}	r_{\min}	$r_{\rm max}$	$r_{0.5}$	$p_{0.5}$
CCSM	8.87×10^{-4}	-0.18	0.21	0.02	1.85×10^{-1}
CNRM	1.86×10^{-3}	-0.06	0.30	0.13	1.02×10^{-1}
GFDL	2.88×10^{-3}	0.02	0.40	0.22	1.12×10^{-2}
GISS	-3.30×10^{-3}	-0.36	0.03	-0.17	3.16×10^{-2}
IAP	1.88×10^{-3}	-0.08	0.30	0.12	6.86×10^{-2}
IPSL	1.62×10^{-3}	0.02	0.38	0.20	1.33×10^{-2}
MIROCm	7.93×10^{-3}	0.17	0.52	0.36	6.34×10^{-6}
MIROCh	9.69×10^{-3}	0.51	0.75	0.65	1.28×10^{-17}
MPI	5.10×10^{-3}	0.08	0.48	0.28	6.16×10^{-4}
MRI	5.71×10^{-3}	0.20	0.52	0.38	1.68×10^{-5}
remean	3.44×10^{-3}	0.49	0.74	0.63	3.48×10^{-16}
remedian	3.98×10^{-3}	0.33	0.62	0.48	1.54×10^{-9}

Table 5.15: Side-by-side summary of median W_{ϕ} in AMIP or 20th Century, SRES-A1B and SRES-B1 scenarios for various reanalysis and CMIP GCMs. Median W_{ϕ} shown for summer mean (JJAS) and winter mean (DJFM).

	JJAS			DJFM		
Model	AMIP or 20th	SRES-A1B	SRES-B1	AMIP or 20th	SRES-A1B	SRES-B1
NCEP	15.03	-	-	9.26	-	_
ERA-40	14.66	-	-	8.46	-	_
ERA-Interim	16.35	-	-	8.88	-	-
CCSM	14.96	15.84	15.86	9.74	10.35	10.11
CNRM	16.09	15.25	15.17	7.73	8.42	8.34
GFDL	14.87	14.94	15.01	10.37	9.92	9.77
GISS	17.35	14.67	16.28	9.56	9.63	10.10
IAP	16.24	17.71	17.50	10.17	8.49	8.38
IPSL	14.87	16.25	16.15	9.13	10.79	10.67
MIROC-mres	14.13	14.36	14.22	7.69	7.21	7.29
MIROC-hres	15.71	15.42	15.42	9.96	10.69	10.69
MPI-ECHAM	16.23	14.82	14.66	8.24	8.36	8.31
MRI	14.97	15.36	15.25	9.23	9.20	9.13
UKMO-HADGEM	14.47	13.17	-	9.39	8.96	-

Table 5.16: Side-by-side summary of iqr W_{ϕ} in AMIP or 20th Century, SRES-A1B and SRES-B1 scenarios for various reanalysis and CMIP GCMs. IQR of W_{ϕ} shown for summer mean (JJAS) and winter mean (DJFM).

	JJAS			DJFM		
Model	AMIP or 20th	SRES-A1B	SRES-B1	AMIP or 20th	SRES-A1B	SRES-B1
NCEP	0.45	-	-	0.37	-	_
ERA-40	0.52	-	-	0.30	-	_
ERA-Interim	0.54	-	-	0.48	-	-
CCSM	0.41	0.88	0.80	0.24	0.72	0.56
CNRM	0.74	0.61	0.51	0.57	0.57	0.54
GFDL	0.47	0.83	0.80	0.44	0.52	0.46
GISS	0.55	1.00	0.75	0.61	0.70	0.87
IAP	0.29	0.63	0.53	0.68	0.54	0.44
IPSL	0.36	0.43	0.34	0.34	0.30	0.33
MIROC-mres	0.21	0.67	0.60	0.34	0.64	0.66
MIROC-hres	0.34	0.44	0.44	0.37	0.56	0.56
MPI-ECHAM	0.37	0.57	0.62	0.71	0.57	0.56
MRI	0.46	0.47	0.52	0.72	0.76	0.57
UKMO-HADGEM	0.39	0.53	-	0.40	0.41	-

	JJAS		DJFM			
Model	AMIP or 20th	SRES-A1B	SRES-B1	AMIP or 20th	SRES-A1B	SRES-B1
NCEP	0.013	_	-	-0.029	_	-
ERA-40	0.026	-	-	-0.011	-	-
ERA-Interim	0.042	-	-	-0.007	-	-
CCSM	0.052	0.004	0.000	-0.019	0.007	0.001
CNRM	0.014	0.006	0.002	0.007	0.004	0.002
GFDL	0.023	0.008	0.003	0.048	0.005	0.003
GISS	-0.004	-0.003	0.003	0.011	-0.002	-0.003
IAP	-0.023	0.009	0.000	-0.012	0.005	0.002
IPSL	0.002	0.004	0.003	-0.014	0.004	0.002
MIROC-mres	-0.003	0.011	0.005	0.018	0.006	0.008
MIROC-hres	0.001	0.005	0.005	-0.014	0.010	0.010
MPI-ECHAM	0.006	0.008	0.002	-0.022	0.006	0.005
MRI	0.024	0.003	0.003	-0.033	0.008	0.006
UKMO-HADGEM	-0.015	0.006	-	-0.010	0.004	-

Table 5.17: Side-by-side summary of the linear trend (°yr⁻¹) in W_{ϕ} in AMIP or 20th Century, SRES-A1B and SRES-B1 scenarios for various reanalysis and CMIP GCMs. Linear trend of W_{ϕ} shown for summer mean (JJAS) and winter mean (DJFM).

Table 5.18: Projection of expected change in mean seasonal mineral dust ($\mu g m^{-3}$) in Barbados from 2000 to 2099 based on median of all model runs for SRES-A1B and SRES-B1 scenarios via linear regression. Expected change calculated from the product of linear regression of 20th century between observed mineral dust at Barbados and WACZ Latitude index and the expected change in position of the WACZ from the year 2000 to the year 2099. Expected change shown for 2.5th and 97.5th percentiles to represent at 5% confidence interval about the median (50th percentile) of linear trend of median bootstrapped seasonal value of all model runs.

	SRE	S-A1B	SRES-B1		
	JJAS DJFM		JJAS	DJFM	
2.5 th Percentile	-3.0	-1.3	-1.1	-1.0	
$50^{\rm th}$ Percentile	-3.8	-2.2	-1.9	-1.5	
97.5 th Percentile	-4.7	-2.4	-2.7	-2.0	
²⁵⁶² Chapter 6

Source Apportionment of Collected Mineral Dust in the Southeastern United States and Verification With Atmospheric Data

2567 1

2568 6.1 Introduction

That the composition of mineral dust could inform as to the origins of the dust is a long established principle, dating back to *Darwin* [1846] who noted the presence of fossils from aquatic micro-organisms as evidence for the origins of the dust likely being lake bed sediment. Despite growing attention to mineral dust over the Tropical North Atlantic in the late 1960's [*Prospero et al.*, 1970; *Carlson and Prospero*, 1972] Darwin's simple hypothesis on the sources of mineral dust in North Africa was not much improved upon until higher resolution satellite

¹This work is presented as a dissertation chapter but is expected to be published with co-authors A. Beck, J.K. Cochran and E.T. Rasbury.

²⁵⁷⁵ imagery began to become available and key source regions from mineral dust in North Africa
²⁵⁷⁶ clearly identified.

While mineral dust is often treated as a homogeneous, spherical object with a simple size 2577 distribution in global climate and theoretical models, in reality the composition, shape and 2578 size vary greatly from source region to source region and on occasion within source regions. 2579 Furthermore such compositions, shapes and sizes can change rapidly in the atmosphere as 2580 the airmass ages, with heavier mineral dust particles settling near to the source and lighter 2581 mineral dust particles persisting in the atmosphere for long periods of time. Changes in 2582 composition of mineral dust have been shown to be of great importance for both atmospheric 2583 and marine chemistry processes, in particular the solubility of iron has been shown to both 2584 vary greatly in time and location and be of critical importance to biogeochemical processes 2585 in the surface ocean. Much attention has been paid to the solubility of iron accounting to 2586 its critical importance as a trace nutrient necessary for primary productivity, however other 2587 metals may undergo similar chemical changes in the atmosphere (e.g. titanium) highlighting 2588 the complexity of the composition of mineral dust in the atmosphere. Similar to composition, 2580 the size and shape of mineral dust can be an important control on both atmospheric and 2590 oceanographic chemical processes. 2591

While the complexity of mineral dust as a function of its source creates challenges for 2592 environmental chemists, the complexity represents an opportunity to apportion source of 2593 mineral dust observed long distances from its source; mineralogical, elemental and isotopic 2594 composition of mineral dust can all be used as tracers for mineral dust aerosols source. For 2595 example, as elemental analysis was being developed it was nearly immediately applied to 2596 determine the composition of dusts. Baron Nordenskiold in 1869 [Hartley and Ramage, 2597 1901] used the elemental composition of dust in snowpack in Upsala, Sweden to determine 2598 if dust deposited in snowpack may have originated from a meteorite shower that occurred 2599 concurrently with its detection. More recently *Perry et al.* [1997] have shown the utility of 2600 using elemental ratios to identify the source of mineral dust. In intervening years analyses 2601

have been performed on surface soils and sands, ocean and ice core sediments and mineral
dust collected in the atmosphere; including major international aerosol sampling projects
(e.g ACE-Asia and SHADE). Much of this work has been summarized in two review papers
by Formenti et al. [2011] and Grousset and Biscaye [2005].

Previous work has assessed the mineral composition of mineral dust near to sources in 2606 Africa [Glaccum and Prospero, 1980; Paquet et al., 1984; Avila et al., 1997; Caquineau et al., 2607 1998; Falkovich et al., 2001; Caquineau et al., 2002; Khiri, 2004; Alastuey et al., 2005; OHara 2608 et al., 2006; Kandler et al., 2009] and Asia [Biscaye et al., 1997; Svensson et al., 2000; Shen 2609 et al., 2005; Wang et al., 2005; Shen et al., 2006; Li et al., 2007; Jeong, 2008; Wang et al., 2610 2008]. Samples in these analyses vary in collection methods from mechanically operated 2611 surface impactors, passive collection on surfaces and collection from near-surface, tower-based 2612 and aircraft retrievals. Furthermore methods to determine mineral composition vary widely, 2613 from single particle techniques such as x-ray diffraction or microscopy to bulk analytical 2614 methods. Additional differences between studies originate from minerals studied and how 2615 mineral composition was reported, (e.g. percentage of weight, percentage of volumes or ratios 2616 of minerals). In some cases single particle analysis is not quantitative but rather results are 2617 reported in a semi-quantitative manner (e.g. counts or relative counts) [Falkovich et al., 2618 2001; Reid et al., 2003; Zhang et al., 2005; Kandler et al., 2007; Jeong, 2008; Matsuki et al., 2619 2010]. Differences in sample collection, analysis techniques and what minerals were reported 2620 lead to difficulties in direct comparisons to study to study. A summary of mineralogical 2621 composition by region from *Formenti et al.* [2011] is given in Table 6.1. 2622

Elemental composition of mineral dust near to sources has also been extensively studied in both Africa [*Chester et al.*, 1984; *Bergametti et al.*, 1989a,b; *Borg and Banner*, 1996; *Chiapello et al.*, 1997; *Guieu et al.*, 2002; *Formenti et al.*, 2003; *Alastuey et al.*, 2005; *Formenti et al.*, 2008] and Asia [*Alfaro and Gomes*, 2001; *Makra et al.*, 2002; *Zhang et al.*, 2003; *Zhang et al.*, 2003; *Xu et al.*, 2004; *Cheng et al.*, 2005; *Sun et al.*, 2005; *Arimoto et al.*, 2006; *Shen et al.*, 2007]. Elemental composition is commonly determined by x-ray fluorescence, or mass

spectroscopy. Many spectroscopic analyses, such as the commonly used inductively coupled 2629 plasma mass spectrometry (ICP-MS) require chemical digestion of mineral dust via strong 2630 acids. Chemical digestion methods vary widely between studies and the choice of chemi-2631 cal digestion methods can impact results. For example hydrofluoric acid (HF) is necessary 2632 to completely break down aluminosilicates. However HF in the presence of silicon dioxide 2633 (SiO_2) can react to produce silicon tetrafluoride (SiF_4) which is a gas at room temperature 2634 and can thus artificially reduce measurements of Si, an element which is an important geo-2635 chemical marker. Furthermore for analysis involving rare earth elements (REE) fluorine ions 2636 are known to attach to and remove REE from solution. Thus choice of digestion methods 2637 can affect elemental ratios, making comparisons between studies challenging. Despite afore-2638 mentioned challenges Formenti et al. [2011] summarized a wide array of elemental analysis 2639 of mineral dust from North Africa and Asia and these are summarized in Tables 6.2. 2640

Isotopic analysis of Nd and Sr (e.g. Borg and Banner [1996]; Grousset and Biscaye 2641 [2005]; Cole et al. [2009]) has been used to identify mineral dust source regions in marine 2642 sediments, ice cores, soils, and in transported aerosols. Earth-derived particles found in the 2643 atmosphere as mineral dust originate from soils and rocks generally of crustal origin. The 2644 isotopes ⁸⁷Sr and ¹⁴³Nd are radiogenic and are produced continuously from their naturally 2645 occurring parents, ⁸⁷Rb and ¹⁴⁷Sm, respectively. The proportion at which these two isotopes 2646 are found is a function of age of a rock, and the initial $\frac{Rb}{Sr}$ and $\frac{Sm}{Nd}$ ratios imprinted by chemical 2647 processes affected by temperature and pressure at the time of rock formation. The isotopic 2648 composition of Sr and Nd varies from the mantle-derived material to the crustal material. 2649 Thus $\frac{^{87}Sr}{^{86}Sr}$ and $\frac{^{143}Nd}{^{144}Nd}$ ratios vary from place to place on Earth's surface as a consequence of 2650 the unique ages and composition of local rocks. While isotopic ratios are strong indicators 2651 of provenance there are challenges with such an analysis. In general isotopic ratios are more 2652 resistant to issues associated with digestion of mineral dust, however isotopic ratios are often 2653 paired with elemental concentrations so digestion techniques can be important. Furthermore 2654 the isotopic ratio has been shown to change with the size of mineral dust Grousset and 2655

Biscaye, 2005], in turn the size of mineral dust has been shown to change with distance from
source [Pye and Tsoar, 1987].

Having outlined challenges associated with using geochemical from previous studies next 2658 we examine if it is possible to use simple mineralogical and elemental ratios to apportion 2659 sources. [Formenti et al., 2011] in a review paper present a useful regional framework and 2660 overview of mineralogy literature for much of the previous work done to apportion source. 2661 North Africa is subdivided into regional basins that are identified as key sources of mineral 2662 dust, such as the aforementioned Bodele Depression shown as NAF-5 in Figure 6.1a. Asia is 2663 similarly treated in Figure 6.1b. Next we present a brief overview of mineralogy, elemental 2664 and trace metal composition and isotopic ratios observed in the literature that can be utilized 2665 to separate source regions analytically; first we look for differences between continents and 2666 second we determine if differences in North African sources can be assessed. 2667

Some differences between Asian and African dust are evident in simple mineralogical 2668 and elemental ratios. Asian dust sources show higher illite to kaolinite ratios (1.1 to 13.9)2669 than African dust sources (0.2 to 2.0, Table 6.1). Similarly the ratio of chlorite to kaolinite is 2670 higher in Asia (1.3 to 3.3) than African dust sources (0.0 to 2.6). In contrast Asian dust (0 to 3.3)2671 35%) appears to have lower carbonate content per unit weight than African dust sources (0) 2672 to 70%). We note a wide range in carbonate content on sub-continental scales and between 2673 studies. The ratio of Ca to Al has been used by Perry et al. [1997] and others to assess 2674 sources of mineral dust: Asian mineral dust (0.31 to 4.14) is enhanced relative to African 2675 sources (0.36 to 0.92) as also shown in Table 6.1. For K to Al, Asian dust (0.20 to 1.00) is 2676 also shown to be enhanced relative to African sources (0.11 to 0.3). The ratio of Fe to Al is 2677 similar between Asia (0.36 to 3.53) and Africa (0.43 to 1.08). 2678

As the Bodele depression is thought to be the source of nearly one-quarter the total dust emitted from North Africa [*Koren et al.*, 2006] much attention has been paid to the mineralogy of the depression [*Mounkaila*, 2006; *Bristow et al.*, 2010]. Previously Chapters 2 and 3 of this dissertation have highlighted the key role that the Bodele depression plays

in responding to changes in the position of the WACZ. The ratios of carbonate and illite to 2683 kaolinite have been used to fingerprint sources previously in the Bodele [Schütz and Sebert, 2684 1987; Caquineau et al., 2002]. Fe has in general higher values in southern Africa including 2685 the Sahel and Bodele than northern locations [Lázaro et al., 2008; Formenti et al., 2008]. 2686 While the ratio of Si to Al is in many places not useful in discriminating between sources, 2687 the relatively constant high values of Si to Al in the Bodele (near 4) may make it a good 2688 tracer of Bodele originated mineral dust relative to other North African sources (near to 2689 2.3) [Formenti et al., 2008]. Cole et al. [2009] note that plagorskite is common in sediments 2690 originally formed in lakebeds, such as that of the Bodele Depression. Plagorskite is rich in 2691 Mg, so elemental ratios in which Mg is enriched could suggest a lakebed source. 2692

While mineralogy and elemental ratios appear to be good indicators of source, it is not 2693 clear if samples taken thousands of kilometers from source regions will maintain source 2694 region signals. For example Trapp et al. [2010] analyzed mineral dust collected in Miami 2695 and Barbados and found that although trace metal concentrations varied tightly with the 2696 concentration of mineral dust, the ratios between metals did not change greatly with time. 2697 It is unclear from this work if this implies a constant source region or if the fine mode of 2698 mineral dust that remains as a mineral dust layer ages does not maintain geochemical tracers 2699 with time and distance from the source regions. 2700

The above literature review, Tables 6.1 and 6.2 and Figures 6.1a and 6.1b highlight 2701 both the challenges associated with assimilating multiple analysis performed with differing 2702 sample collection and analytical techniques and the potential utility of such analysis in 2703 apportioning sources. Variability between measurements in a region or continent is often of 2704 similar magnitude as the variability between regions or continents. In particular we note 2705 that previous studies have focused either on a specific region (e.g. Bodele Depression or 2706 Gobi Desert) or on an individual continental source, and no studies perform analysis of 2707 samples from varying sources. To address the issue associated with sample and analytical 2708 technique disparities in previous analyses, we collect samples from four different continents 2709

(North Africa, Europe, Asia and Australia) and present a unified analysis via a qualitative mineralogy and a quantitative elemental composition of the mineral dust samples. As a test of this analysis five samples of mineral dust with unknown origins collected in the Southeastern United States are analyzed and their sources assessed. This study seeks answers to three main questions: (1) Is continental source apportionment possible with trace metal analysis? (2) Is regional source apportionment possible with trace metal analysis? (3) Do North African regional sources change as a function of the position of the WACZ?

The structure of the chapter is as follows. In Section 6.2 we introduce the samples 2717 acquired and analyzed in this project. In Section 6.3 the digestion, analysis and statistical 2718 techniques used in this project are introduced. In Section 6.4 the results of this project are 2719 shown. Sections 6.5 and 6.6 we summarize and discuss our findings in the context of prior 2720 results, linking these results to findings in Chapters 2 and 3 of this dissertation in which 2721 the role of the WACZ in controlling mineral dust emission and transport was established. 2722 In addition we discuss the potential utility of isotopic analysis, how this future work will 2723 augment our findings and how this analysis can be used in near-real time to determine the 2724 sources of mineral dust to the southeastern United States. 2725

2726 6.2 Materials and Data

Samples of mineral dust were collected from Africa, Europe, Australia, North America and
Asia at varying distances from source regions. To characterize mineral dust sources by
continent we acquired samples near to major source regions of Asia, Africa and Australia.
To test our characterization scheme we include filter samples of mineral dust of unknown
origins in the Southeastern United States.

²⁷³² 6.2.1 Source Sample Collection

Table 6.3 summarizes the origins of mineral dust samples acquired for this study. All samples were collected at the Earth's surface in the form of deposited mineral dust. In most cases samples were simply swept off a surface (e.g. board on a rooftop for samples collected in Spain or car window in the case of samples collected in Australia) and sent for analysis. All samples captured in Europe are assumed to be of North African origin, as North African dust storms impacting Spain and the coastal Mediterranean have been well established in the literature.

2740 6.2.2 SEARCH Project

Our filters for analysis come from Pensacola, Florida collected as part of the South-Eastern 2741 Aerosol Research and Characterization Study experiment (SEARCH project: http://www. 2742 atmospheric-research.com/studies/SEARCH/index.html.) sponsored by the EPA. Fig-2743 ure 6.2 shows the location of aerosol filter collection stations in the SEARCH project. 2744 Atmospheric-Research, a subcontractor with the EPA provided five - 47mm Tefon filters 2745 with mineral dust loadings of 25-125 μ g of mineral dust. Pensacola, FL receives mineral 2746 dust periodically during the spring and summer months. The location of the sample site is 2747 shown in Figure 6.2. 2748

2749 6.3 Methods

Here we focus on the two analytical techniques used to determine mineralogy and elemental composition of mineral dust samples. Before being chemically digested, samples were analyzed at the National Synchrotron Light Source (NSLS) at Brookhaven National Lab to determine mineralogy. Samples were then chemically digested in the clean-room of the Isotope Laboratory under the direction of Dr. Troy Rasbury at the Department of Geosciences at Stony Brook University, then analyzed by ICP-MS in the lab of Dr. Aaron Beck at the ²⁷⁵⁶ Virginia Insitute of Marine Science. Next we detail the analytical methods utilized in each²⁷⁵⁷ aforementioned step.

$_{2758}$ 6.3.1 μ X-ray Diffraction and Fluorescence

A major advantage of μ x-ray diffraction (XRD) and μ x-ray fluorescence (XRF) performed 2759 at the National Synchrotron Light Source (NSLS) Brookhaven National Lab is that the 2760 analyses do not require destruction, meaning that the mineral dust samples (in particular on 2761 filters) could be later digested and further analyzed. XRD measures x-rays scattering from a 2762 sample that has been bombarded with x-rays or gamma rays containing high levels of energy. 2763 X-rays of different energy levels are scattered from the individual mineral dust particle and 2764 re-emitted from the bombarded sample, then both the rate and shape of scatter are measured. 2765 The intensity and pattern of scattered radiation are determined by the crystalline physical 2766 structure of atoms, thus individual minerals have unique scattering properties that can aid 2767 in identification. XRF requires bombardment with high energy particles, in the process 2768 dust molecules are excited and based on elemental composition emit radiation at discrete 2769 wavelengths and intensities which can be used to identify composition of the dust. While 2770 both processes require high energy radiation and then measure reflectance/scatter (XRD) 2771 or emission (XRF), the analytical techniques for the two are different. XRF analysis of 2772 a sample produces a two dimensional map of the sample, showing changes in elemental 2773 concentrations. In contrast XRD is a single particle analysis, requiring that each individual 2774 mineral dust grain be analyzed individually. 2775

Samples were handled as follows. Pure dust samples were mounted in a thin layer onto kaptron adhesive tape, chosen for its lack of diffraction. In an attempt to produce a layer of one particle thick samples were mounted from vials onto the tape, then tapped repeatedly to dislodge excess dust. XRF mapping was performed on the samples, along with XRD on individual grains. As mineral dust was spread evenly across the tape XRD was performed randomly on discrete spots along the kaptron tape. Because the beam of radiation was on the order of 15 μ m in width it was impossible to always identify and analyze individual grains, often multiple mineral dust particles were captured in one image. In contrast to mineral dust mounted on kaptron tape, mineral dust on collected filters was not evenly distributed but rather preferentially settled into depressions on filter surface (see Figure 6.4). In this case XRD could not be performed randomly, but rather in a semi-random location. XRF was used to identify locations in which metals associated with mineral dust were present and XRD was performed at this location.

Once a location was selected for XRD, the standard method for collection an analysis 2789 of XRD patters for NSLS-BNL beamline x26 was followed. Here briefly we highlight the 2790 approach. The standard software, Fit-2D was used for diffraction imaging. Two standards 2791 were first analyzed to calibrate the software alumina (Al₂O₃, NIST X-Ray Powder Diffraction 2792 Intensity Set SRM 674a) and silver behenate $(AgC_{22}H_{43}O_2)$. Using Fit-2d, power diffration 2793 patterns were calculated for each sample. A kaptron blank was taken and subtracted from 2794 observed intensities for pure dust samples. For filters a Teflon filter blank was taken and 2795 subtracted, however the blank signal was large and often overwhelmed the signal of the 2796 mineral itself. Calibrated, blank-corrected outputs were imported into Match! software 2797 for comparison to a mineralogical database; samples that did not have a clear match were 2798 discounted. We consider the results from this analysis to be semi-quantitative at best, and 2799 are probably best characterized as qualitative. Filter concentrations of mineral dust were low 2800 and diffraction signals were weak while the diffraction patterns from the Teflon filter were 2801 often quite strong. Formenti et al. [2011] note challenges in limited diffraction databases for 2802 mineral dust particles as observed in previous studies [Moore et al., 1989; Caquineau et al., 2803 2002. That the results of this analysis are qualitative rather than quantitative is not rare 2804 for diffraction studies [Falkovich et al., 2001; Reid et al., 2003; Zhang et al., 2006; Kandler 2805 et al., 2007; Jeong, 2008; Matsuki et al., 2010] because of low abundance of atmospheric 2806 aerosols and limited diffraction database resources. In this study, the mineralogy serves as 2807 useful supplemental information to the metal analysis. 2808

²⁸⁰⁹ 6.3.2 Elemental Analysis via ICP-MS

Analysis of elemental composition is performed via ICP-MS, which requires complete chem-2810 ical digestion of samples so that metals are dissolved into an acid solution which can be 2811 injected for analysis. Si is thought to be a key element for identifying mineral dust originat-2812 ing in the Bodele depression and in digestions utilizing HF Si can be lost via evaporation of 2813 SiF_4 in a gaseous compound. We performed digestions without HF in order to preserve Si. 2814 However without HF we cannot be assured of complete digestion of aluminosilicate clays, 2815 potentially reducing yields of elements commonly contained in these materials (such as: Al, 2816 Si, REE) and making comparison of our results to other analyses difficult. We used two di-2817 gestion methods, one to digest pure dust samples and the other to digest filters that contain 2818 mineral dust. Both digestions were performed in the clean lab of the Isotope Laboratory in 2810 the Geosciences Department of Stony Brook University. Unless otherwise noted all materials 2820 used were acid washed and subjected to sequential HCl, HNO_3 and DI water baths for over 2821 a month before utilization. All solvents used in this analysis were trace metal grade, low 2822 blank, SEASTARTM rated acids. 2823

Pure mineral dust samples were weighed on a Mettler micro-balance (Table 6.3). Sample 2824 masses were chosen to be of the order of mineral dust expected to be on the filters (25 to 2825 100 μ g). Dust was placed in Teflon vials for digestion (see Figure 6.3a). Initially 3mL aqua 2826 regia was applied to the sample for two days, then the sample was brought to dryness on 2827 a hotplate. After cooling, 2mL of HNO_3 (16 N) were added to the samples, after two days 2828 the samples were again brought to dryness on a hotplate. Lastly 1.6 mL of HNO₃ (1 N) was 2829 used to pick up dissolved metals and centrifuged to remove non-dissolved refractory material 2830 (not quantified). 2831

Filter samples were treated as similarly as possible to the pure mineral dust samples. However in contrast to the pure dust samples, the mass of mineral dust on the filters is unknown, so it is impossible to calculate elemental concentration in parts per million or similar notation. Filters were placed in Teflon vials for digestion and 6mL aqua regia was ²⁸³⁶ applied to the samples for two days, then the sample was brought to dryness on a hotplate. ²⁸³⁷ After cooling the filters were flipped with acid washed tongs (see Figure 6.3c) and 3mL of ²⁸³⁸ HNO₃ (16 N) were added to the samples, then after two days the samples were again brought ²⁸³⁹ to dryness on a hotplate. To allay concerns of metal ions attaching to the non-Teffron edges ²⁸⁴⁰ of of filters, they were rinsed with 2 to 3 mL of HNO₃ (1 N) then dried to completeness ²⁸⁴¹ before being treated as pure-dust samples for the remaining steps, including pick-up in 1.6 ²⁸⁴² mL followed by centrifuging.

²⁸⁴³ While samples were treated to reduce trace metal contamination, some contamination is ²⁸⁴⁴ unavoidable. For example we expect contamination would be possible during collection and ²⁸⁴⁵ μ x-ray analysis at BNL. All other digestion and analysis steps were performed in clean-lab ²⁸⁴⁶ environments, however despite this precaution blanks can be encountered in non-properly ²⁸⁴⁷ cleaned vessels and in the acids used in digestion. To account for blank or contamination ²⁸⁴⁸ during digestion, three blanks were run for both the pure dust and filter examples, and ²⁸⁴⁹ treated exactly as described above for actual samples.

Fully digested, centrifuged samples were then taken to the Virginia Institute of Marine 2850 Science for ICP-MS analysis. While elements such as Fe, Al, Ca, K and Si are commonly 2851 found in crustal material and are in relatively high abundance [Taylor et al., 1983] other 2852 elements which can be used to apportion source via natural variability such as REE or 2853 through trace metal indicators of anthropogenic activity such as As, Cd, Cu, Cr, Ni, Pb, 2854 V, and Zn [Trapp et al., 2010] are often many orders of magnitude lower in abundance. To 2855 account for such a large range of natural variability, standards were run over six orders of 2856 magnitude (a range of 0.003 ppm to 31.9 ppm). The calibration curves are generally reliable 2857 at over 5 orders of magnitude higher than the highest value of the calibration curve. Based on 2858 calibration curves we discount values less than 0.001 ppm (or 1 ppb) as unreliable, although 2859 for some metals higher cutoffs are applied where necessary based on blank problems. 2860

To account for instrument drift, indium was used as an internal standard, as it is not found in nature. We cannot quantify contamination associated with sample collection, or contamination encountered during x-ray analysis, but blanks were measured for machine/instrument
(25), digestion of pure dust (3), digestions with filters (3). Digestion variability was further
assessed, quantified and corrected for by use of digestion replicates (3) and matrix issues
addressed via standard additions (6). Table 6.4 summarizes the blanks calculated in this
analysis.

In general instrument blanks were small compared to signal from pure dust samples (Table 6.4). Only Si had a machine blanks in excess of 10% (11.7%) of the median pure dust signal. As the signals on the filters were lower than those of pure dust, the ratio of instrument blank to filter signal is higher. However only three elements had banks of between 10% and 50% (Si, Eu, Gd). Other elements exhibited an instrument blank between 1% and 10% (U, Mg, S, Ca, Rb, Sr, Ba, Sm, Ho, Er, Tm, Yb, Lu, Th).

However, in general, instrument blanks were small compared to digestion blanks. In all 2874 cases digestion blanks were high for Fe, Ni, Cu, Zn many of which have been historically 2875 used for provenance. The digestion blank was calculated individually for pure dust and 2876 filter, as the digestion process differed slightly in the quantity of acid used in digestion. For 2877 pure dust a number of key elements had a digestion blank between 1% and 10% of the dust 2878 signal (Si, S, V, Mn, Fe, Cu), no elements fell into the range of 10% to 50% and two others 2879 far exceeded 50% (Ni, Zn). Rare earth element blanks were extremely low, well below 1%2880 for all elements, and often well below 0.1%. The filter digestion process introduced large 2881 blanks, likely coming from the filter itself. Few elements had filter digestion blanks between 2882 1% and 10% (Si, S, Rb). Many major elements commonly found in crustal materials had 2883 filter digestion blanks between 10% and 50% (Pb, U, Mg, Al, Ti, V, Sr, Ba). Multiple 2884 elements had filter digestion blanks in excess of 50% (Ca, Mn, Ni, Cu, Zn). In particular the 2885 filter digestion process provided more metal than the sample did (ratio greater than 100%) 2886 for many elements which are used to establish provenance (Ca, Mn, Ni, Cu, Zn). Blanks 2887 represent a challenge to the analysis, in particular Ni and Zn appear to be likely unusable. 2888 Thus for these filter samples the list of elements which could be reliably estimated is greatly 2889

2890 reduced.

2891 6.4 Results

2892 6.4.1 Mineralogy

Table 6.5 to Table 6.5 shows minerals identified in pure dust samples from two African, two Asian and one Australian sample. Samples that did not have a match in the mineral database are included to give a sense of mineral representation.

Two samples from North Africa were analyzed (POLS 3, 4GOST) and 16 diffraction patterns were taken and analyzed. Five of 16 patterns were matched to known minerals, the remaining 11 patterns in general had no match in the database. It has been previously noted that mineral dust diffraction patterns, in particular North African samples are under represented. Of the five patterns identified, calcite, muscovite, kaolinite, and dolomite were dominant.

Three samples from Asia were analyzed (Nanjing, Inner Mongolia, Nandon) for a total of 2902 17 diffraction patterns. Twelve of 17 patterns were matched, a much higher ratio than for the 2903 North African minerals. Dolomite and calcite were observed 3 of 17 times. Aluminosilicates 2904 (biotite, muscovite and generic aluminosilicate, aluminum oxide) were observed 4 of 17 times. 2905 Iron and titanium oxides were common in the Asian samples (iron-titanium hydrate, iron 2906 oxide, titanium oxide, rutile, magnetite); identified in 7 of 17 patterns. Additionally quartz 290 with iron impurities was observed. This is largely different from North African mineral dust 2908 where iron and titanium oxides were not observed. Silica and quartz were observed in 2 of 2909 17 measurements. 2910

The Australian sample yielded six diffraction patterns. Muscovite and a generic aluminosilicate were observed in 2 of 6 diffractions, titanium oxide and magnetite were observed in 2 of 6 diffractions and silica was observed in 1 of 6 diffractions.

²⁹¹⁴ While the mineralogy cannot be considered to be quantitative, it does allow us to place

the source minerals three loose groupings, a calcium carbonate group (calcite, dolomite), a titanium-iron oxide group (iron oxides, titanium oxides, iron-titanitum hydrates, magnetite, rutile) and an aluminosilicate group (kaolinite, muscovite, aluminosiciate (generic) and aluminum oxide). Silica and quartz were also identified but may not be dissolved in our digestive approach, thus may not contribute to our elemental analysis.

²⁹²⁰ 6.4.2 Trace Metal Chemistry of Mineral Dust

In Figures 6.5 and 6.6 elements of interest normalized by their concentration of Al are plotted. 2921 Normalization to Al is performed for two reasons; continuity to prior studies *Formenti et al.*, 2922 2011] and as filters contain unknown masses of dust, elemental concentrations $\left(\frac{\text{massofelement}}{\text{massofsample}}\right)$ 2923 cannot be directly determined. The utility of Ca, Fe and Si are described in our review of 2924 [Formenti et al., 2011]. V and Pb have been shown to be tracers of anthropogenic activity 2925 [Sedwick et al., 2007; Trapp et al., 2010] and in turn sources as African mineral dust passes 2926 through fewer industrialized locations than does Asian dust. Mn has been shown to track 2927 tightly with the total quantity of mineral dust in aerosols collected near to the Caribbean 2928 [Trapp et al., 2010]. In each figure the mineral dust source is shown by color: Asian dust is 2929 blue, African dust is red and Australian dust is green. 2930

Figures 6.5 and 6.6 are complimentary figures acting as a matrix of each individual 2931 ratio plotted against each other to best visually separate continental sources. Ca/Al shows 2932 clear separation in continental dust sources; African mineral samples exhibit higher ratios, 2933 generally in excess of 4. Asian samples have significantly lower ratios of Ca to Al, with values 2934 below 4. Ti to Al also is able to parse continents. Asian samples exhibit Ti/Al ratios in 2935 excess of 0.02, while the majority of African samples fall below 0.01 (although some do fall 2936 in the Asian range). Other ratios are less successful in separating sources. In both Mn/Al 2937 and V/Al, Asian and African samples overlap each other. For both Si/Al and Pb/Al some 2938 discrimination is evident, but enough samples are in mixed ranges that cause problems. 2939 Plotting Ca/Al vs Ti/Al is the most successful in separating Asian and African sources 2940

(Figure 6.5). Asian dust is found in the upper left quadrant of the plot, corresponding to
Ti/Al ratios in excess of 0.02 and Ca/Al ratios below 4 ((Figure 6.5)). African dust is clearly
visible outside of the narrow Asian window. Australian dust is also clearly separated, falling
nearest to the origin.

However due to concerns with possible contamination during the digestion process with 2945 Al, it is not clear if ratios normalized with Al to appropriate. We repeat the analysis normal-2946 izing with Ti, which had significantly less contamination (Table 6.4). While normalization 2947 to Ti is less common in the literature, doing so yields clearer separation between continents. 2948 Figures 6.7 and 6.8 present the matrix of ratios normalized by Ti. The ratio of Pb to Ti is 2940 not helpful in separating Asian and African sources, but Australian dust is easily identified 2950 by its high concentration of Pb. Asian dust appears to be depleted in both Ca and Mn 2951 relative to African dust, with ratios of Ca/Ti of an two orders of magnitude less than Asian 2952 dust and ratios of Mn/Ti less than 2, while the majority of African samples show ratios in 2953 excess of 2. Al/Ti, Si/Ti and V/Ti all show the same general trend: Australian dust shows 2954 high ratios, African dust has medium ratios and Asian dust has low ratios. Values of 1 for 2955 Si/Ti, 50 for Al/Ti and 0.25 for V/Ti can roughly be used to separate African-Asian source 2956 regions. 2957

Cole et al. [2009] suggest that Mg may be a useful element in identifying dust originating in dried lakebeds, such as the Bodele Depression. However we do not show Mg Figures 6.5, 6.6, 6.7 and 6.8 as Mg is not useful in separating continental sources. All Asian dust is depleted in Mg (ratios near to zero), while Mg ratios in African dust ranges over orders of magnitude. Here we hypothesize that Mg could be useful in separating regional sources, however it is not useful in separating continental sources.

We have identified a number of ratios by which continental sources can be separated, however to a large extent this approach is subjective and only includes analysis of six of the thirty elements analyzed for. To better quantify the relationships and include all elements analyzed, we use a principal component analysis.

²⁹⁶⁸ Principal Component Analysis Using Metal Data

principal component analysis (PCA) can be used to identify which elements are coupled together over varying source regions. We first perform PCA using elemental ratios normalized to Al, which is the most common normalized element. We repeat the analysis using ratios normalized to Ti to account for potential Al contamination.

Figure 6.9 shows the results of the PCA for ratios normalized by Al. Figure 6.9 a shows the 2973 magnitude first principal component (PC1) and second principal component (PC2) with the 2974 sample value plotted as a color blue for Asia, red for Africa and green for Australian. Asian 2975 dust samples are found only in the top right quadrant of the figure, corresponding to values of 2976 PC1 below -1 and PC2 values above 0. In contrast African samples are uniformly distributed 2977 across PC1, but generally fall below a values of PC2 of 1. Figure 6.9b shows the second and 2978 third principal (PC3) components plotted against each other. In summary Asian dust can 2970 be identified by PC1 values above 1, PC2 values above 0 and PC3 values near 0. African 2980 dust can be identified by PC2 values below 1 and PC3 values above 0.5 (with a notable 2981 outliers). Figure 6.9c attempts to visualize the three-dimensional relationship; separation 2982 between sources is visible. Figure 6.9d shows the biplot of elements that correspond to the 2983 principal components. Weight of the individual element is shown in blue by the distance from 2984 the origin. Each element is labeled on the biplot. Red dots represent the samples. Three 2985 key groupings are identified; Ti-Fe-Pb-Mn are grouped together, possibly representing an 2986 iron-oxide grouping. A Ca-Rb-Sr-Si group is seen possibly corresponding to a carbonate 298 group. To the right REE elements are grouped together tightly. 2988

Figure 6.10 shows the results of the PCA for ratios normalized by Ti. The first and second principal components are featured in Figure 6.10a with sample values again plotted as color blue for Asia, red for Africa and green for Australian. PC2 discriminates little between African and Asian dust, as most values are near to a value of zero. In PC1 however Asian dust is shown to have values of less than -3, and African dust has values greater than -3. Figure 6.10b shows the third principal component which also can be used to separate Asian and African dust. In PC3 Asian dust have values less than -0.5, while African dust has values greater than -0.5. Figure 6.10c combines all three principal components, and visually African and Asian dust can be discriminated. Australian dust is disparate to Afrian and Asian dust in all three PC. Figure 6.10d shows the biplot for the principal components normalized by Ti, again three groups are evident a Ca-Sr group, a Fe-Pb-Mn group and a REE-Al-Si group.

We have highlighted two approaches to separate African and Asian dust, using elemental composition of a few select metals and principal component analysis of multiple metals. In the next section we seek to test our separation methods on filters of mineral dust with unknown origins.

3005 6.4.3 Apportionment of Unknown Filters

First we consider metals ratios normalized by Al. Ca/Al and Ti/Al were both shown to separate Asian and African sources. The Ti/Al of all five filters fall within the range of African samples, with all filters having ratios near 0.01. Ca quantities measured on filters were low, with 3 of 5 falling below detection limits. Of the two remaining filters the Ca/Al ratios fell into the Asian range. Little can be concluded from this analysis as normalizing by Al yields conflicting results.

More elemental ratios were able to separate continental sources when ratios were normal-3012 ized by Ti than Al; Ca/Ti, Mn/Ti, Al/Ti, Si/Ti and V/Ti all were able to clearly separate 3013 Asian and African dust sources. Ca/Ti ratios for filters were low, but as previously noted 3014 3 of 5 filter samples had Ca levels below detection. Mn/Ti filters ratios were low, similar 3015 to Asian signals. Al/Ti ratios of filters fell in medium range, closer to African dust than 3016 Asian dust. Si/Ti values for filters varied widely and had values near both Asian and African 3017 ranges. V/Ti values for filters fell in the middle of the African source values. Two metal ra-3018 tios (Al/Ti, V/Ti) suggested African sources and one ratio (Mn/Ti) suggested Asian sources 3019 for the filters. This analysis, similar to the results using ratios normalized to Al, yielded 3020

3021 conflicting results for sources.

Next we repeat the PCA, including filter samples normalized by both Al and Ti. In this analysis the loading patterns are re-defined as the variance of all samples is greatly changed by the inclusion of filter samples. To avoid biases from variable concentrations all samples are normalized before the PCA is performed. Of interest here are two questions; can source regions still be separated and do the filter samples group with any source region.

First we examine the PCA for elements normalized to Al as shown in Figure 6.11. Figure 3027 6.11a shows the first and second principal components, with filter samples shown in black. 3028 Filter samples are separated from source samples along the first principal component. The 3020 majority of source samples are bunched together in the lower right quadrant of Figure 6.11a 3030 with negative values of the 2nd principal component, and positive values of the 1st principal 3031 component. It is difficult to use Figure 6.11a to separate sources, as African and Asian 3032 mineral dust appear to be intermingled tightly. The 2nd and 3rd principal components are 3033 plotted on Figure 6.11b. Here sources are better separated: Australian dust have higher 3034 values of the 3rd principal component than others, while Asian dust has higher values of 2nd 3035 principal component compared to African dust. However filters values range greatly over the 3036 3rd and 2nd principal components, not matching with any source group clearly. Combining 3037 all three components in Figure 6.11c filter samples are clearly distinct from other source 3038 Additionally no clear geochemical explanation for principal component loading regions. 3039 patterns (Figure 6.11d) is evident. Rare earth elements do group tightly in the lower right 3040 quadrant, but other previously identified groupings such as Fe-Ti (iron and titanium oxides) 3041 and Ca-Sr-Rb (carbonates) are now disparate and randomly grouped. 3042

Figure 6.12 is as Figure 6.11 but with data normalized to Ti. Figure 6.12a shows the first and second principal components, where each major source region is clearly separated along the first principal component with Asian samples falling below -3, African samples falling between -3 and 4 and Australian samples falling above values of 8. Figure 6.12b shows the 2nd and 3rd components, where additional separation is evident along both the 2nd

and 3rd components. Along the 2nd component, values increase from Australian samples 3048 (-2 to -1.5) to Asian samples (-1 to -0.5) to African samples (-0.75 to 5). Along the 3rd 3049 component, values decline from Australian samples (near to 1.5), towards Asian samples (0 3050 to (0.5) and into African samples (0 to -2). However along each principal component, filter 3051 samples are clearly distinct and separate from nearly all source signals, as summarized in 3052 Figure 6.12c. Geochemical groupings by loading pattern are not as clear when filter samples 3053 are included as seen in Figure 6.12d. Rare earth elements appear to be tightly clustered 3054 again, with aluminum suggesting an aluminosilicate source. However carbonates (Ca, Sr) 3055 and iron oxides (Fe) are not clearly separated. 3056

In summary principal component analysis shows that with filters included separation 3057 of sources is possible, particularly when looking at data normalized to Ti. Signals are 3058 not distinct when looking at data normalized to Al, possibly due to variability induced by 3059 extremely large blanks in Al in filter analysis (blank is 37% of Al median filter signal). In 3060 comparison Ti had relatively small blanks (11%) and showed more skill in separating source 3061 regions. Additionally while we are able to separate continental sources, we note that our 3062 clear geochemical separation by aluminosilicate vs. carbonate vs. iron oxide disappears, 3063 possibly due to high blanks associated with filter digestion in key tracers such as Fe (84%). 3064 Ca (256%), Sr (48%). 3065

3066 6.5 Discussion

3067 6.5.1 Back Trajectory Analysis

Geochemical sourcing of mineral dust is a useful tool to apportion source, however it not the only tool available. In this section we couple geochemical tracers with back-trajectory analysis and satellite data.

Figure 6.13a and 6.13b are aerosol absorbing data from TOMS/Earth-Probe for the day before and day of collection respectively. A pulse of dust is evident near Bermuda, extending

southward toward Hispanola in Figure 6.13a, the next day the dust appears to move over 3073 Florida towards the collection site. Figure 6.13c and 6.13d show back trajectory analysis 3074 during morning (12z) and early afternoon (18z) during the day of collection. Trajectories 3075 were run at three heights at 1,000 (red), 10,000 (blue) and 15,000 (green) meters above 3076 ground level and run back in time for ten days prior to sample collection. In both morning 3077 and afternoon, air from near the surface has its origins over the Tropical North Atlantic 3078 Ocean. For air at highest heights in both morning and afternoon the air originates from 3079 over the Pacific Ocean. Air at intermediate height switches from Atlantic to Pacific origins. 3080 Satellite measurements of dust appear to suggest a Saharan source, as does near-surface air. 3081 however back trajectory analysis of air aloft leaves open the potential of Asian influence. 3082

Filter 03-T9329 shows similar satellite imagery to the last case, on the day prior to 3083 collection Figure 6.14a dust particles appear over the Bahamas south towards Puerto Rico 3084 extending into South Florida. The next day dust appears to spread west over the remainder 3085 of Florida. Trajectory analysis is shown in Figure 6.14c and 6.14d for morning (12z) and 3086 afternoon (18z) the day of collection. Low level flow again shows a source over the Tropical 3087 Atlantic near to the coast of Africa. Upper level flow again shows a source over the Pacific, 3088 with middle levels swapping sources over the duration of the day. Based on satellite mea-3089 surements a North African source would be assumed, supported by low level flow, however 3090 again air arriving aloft, suggests the possibility of Asian aerosols. 3093

In contrast to the previous two cases satellite imaginary for the day prior to (Figure 6.15a) and of collection (Figure 6.15b) for Filter 04-T11260 no clear dust plumes are evident approaching Florida, although Figure 6.15b suggests that some dust may passed over Florida between images. Figure 6.15c and 6.16d show that both in morning and afternoon near surface air has origins in the Tropical North Atlantic, and air aloft has origins over the Pacific.

³⁰⁹⁸ Filter 04-T11261 has the least indication of mineral dust from satellite imagery as shown ³⁰⁹⁹ in Figure 6.16a and 6.16b. On the day of collection (Figure 6.16b) some indication of dust may be approaching Florida, but the signal is weak. Trajectories are shown for the morning and afternoon of collection in Figures 6.16c and 6.16d. Low level air has its origins near to Africa and air aloft has origins from near to Asia in the Pacific.

Satellite images associated with Filter 04-T9429 are shown in Figures 6.17a and 6.17b for the day preceding collection and the day of collection. On both days a pulse of dust appears to be near to Florida, either just south and west of the collection station or to the east on respective days. Trajectories for Filter 04-T9429 are shown in Figures 6.17c and 6.17d for morning and afternoon of the day of collection respectively. Here no clear source of air masses is evident, suggesting something of a stagnant wind pattern. No Asian of African influence of air masses is evident.

In general satellite imagery confirms the expectation that mineral dust appears to be coming from points east in its path passing over Florida. Trajectory analysis suggest that air at low levels is originating over the Tropical Atlantic near to Africa, however air aloft appears to have its origins in most cases over the Pacific.

The above analysis comes with a number of caveats. The satellite analysis as presented 3114 is both qualitative and subjective. The satellite in use here has known calibration issues over 3115 this time-period [Kiss et al., 2007] and is likely over-estimating aerosol load. The trajectory 3116 analysis is run using low resolution NCEP reanalysis data, the only data available for analysis 3117 over this period. As such there is a large degree of uncertainty of air mass source, and this 3118 uncertainty likely grows over the ten-day back trajectory presented here. None the less, this 3119 analysis suggests that the Sahara is the likely source of mineral dust to Florida but Asian 3120 influence cannot be ruled out. 3121

3122 6.5.2 Geochemical Tracers and the WACZ

Previous chapters introduced the hypothesis that changes in the position of the WACZ changes the quantity of Saharan mineral dust transported across the Atlantic, specifically by activating preferred hot spots such as the Bodele Depression in years in which the WACZ

is south. Filters were subjectively selected to include the largest mineral dust events over 3126 the period 2003 to 2004, thus the events included in this analysis are like major Saharan 3127 dust events. Previous studies have indicated timespans of approximately a week to reach 3128 the Caribbean, and thus additional time to reach Florida. As such here we assume the dust 3129 took between 7 and 14 days to reach Florida. To examine if the position of the WACZ 3130 plays a role in the quantity of mineral dust reaching Pensacola, FL we calculate the mean 3131 position of the WACZ over the period 7 to 14 days preceding the sample collection. This 3132 information is summarized in Table 6.6 along with the anomaly of the week long period for 3133 the trend corrected climatological mean. In four of five events the WACZ was south of its 3134 climatological position. 3135

3136 6.6 Conclusions

In this work we analyze dust samples collected over four continents for mineralogy via diffraction and elemental ratios via ICP-MS. Principal component analysis, a novel approach in geochemistry, is applied to elemental data and, combined with elemental ratios, clear separation of source regions is demonstrated, representing a validation of the method.

Blanks, for the filter samples are problematic. High blanks in elements previously identi-3141 fied via PCA as being important in discriminating between key mineral groupings, e.g. (Fe, 3142 Ca, Sr and Al) reduce the ability to apportion source of filter samples. Similarly in all treat-3143 ments blanks on elements which could be used as anthropogenic signal identifiers, such as 3144 Zn, Cu and Ni are high and reduce the usefulness of these elements to discriminate sources. 3145 Instrument blanks associated with Si may be reduce its utility in identifying alumniosilicate 3146 rich materials. However, Ti is shown to be a resistant element with low blanks and is useful 3147 in each analysis. 3148

The elemental composition of pure dust samples expressed as ratios to Al and Ti showed the ability to separate African and Asian dust sources for specific elemental ratios. However

when many of these elemental ratios were examined for filter samples of unknown origin, 3151 they showed ratios dissimilar to both African and Asian samples, or some ratios were similar 3152 to African and others similar to Asian samples. This ambiguity leaves open that possibility 3153 that problems with the digestion method could be the cause. As the initial quantities of 3154 mineral dust on filters are unknown, recovery during the digestion process is not known. 3155 Metals could be dissolved into acid and then resorb onto the edges of the filter which were 3156 not Teflon. It is also possible that mineral dust could be deposited deep into the Teflon and 3157 thus not exposed to the acid, or simply not fully digested. If the digestion process was not 3158 the same between pure dust and filter samples, comparisons made here are not appropriate. 3150 Only three samples from Asian sources were used in this analysis, and less than the 3160 seven African sources utilized. African samples in general exhibited wider spread than Asian 3161 samples, possibly due to under-representation of Asian sources in this study. An additional 3162 four samples from Asia have been procured but these samples arrived after this analysis was 3163 performed. Results would have been strengthened by including theses additional samples. 3164

3165 6.6.1 Filter Handling

Blanks associated with filter samples were unacceptably high. This experimental set up 3166 does not allow for us to quantify or account for blanks arising from sample collection in 3167 the field or handling during μ -XRF and μ -XRD analysis. Filter samples were collected in 3168 impacters exposed to the elements for prolonged period of times. After collection it is unclear 3169 how filters were handled or processes. In future studies sample collection blanks must be 3170 accounted for. Furthermore the advantage of non-destructive techniques such as μ -XRF 3171 and μ -XRD is offset by possible contamination induced during processing. Future analyses 3172 should be designed for two samples to be taken, one directly sent for μ -XRF and μ -XRD 3173 analysis and the other for trace metal analysis. 3174

3175 6.6.2 North American Mineral Dust

While samples were procured from four continents, no North American dust samples were 3176 used in this analysis. The Sahara is the dominant global source of mineral dust aerosols, 3177 with the Asian continent the second largest source of mineral dust aerosols. In general 3178 North America is not a major source of mineral dust on the global scale, but regionally 3179 North American dust sources can be important. We cannot rule out North American dust 3180 sources as unimportant for the Southeastern United States so samples from North American 3181 desert bases should be acquired and included in future analysis. Local sources of mineral 3182 dust mixing in with long-range dust on filters could explain why observed ratios varied from 3183 "pure" end-member samples. 3184

3185 6.6.3 Isotope Analysis

As discussed in the introduction, isotopic analysis is a powerful tool to apportion source. Samples used in this study have been treated in a way that isotopic analysis is the next step in the analysis, however due to multiple lab setbacks this analysis cannot be included this dissertation. We anticipate performing this isotopic analysis in the near future, as isotopic ratios should complement findings from the principal component analysis.

3191 6.6.4 Rare Earth Elements

Analysis of rare earth elements (REE) may represent a strong method for apportioning source, as shown in *Taylor and McLennan* [1985] and subsequent work. Blanks associated with instrument and digestion techniques are low for rare earth elements. Although REE are found in low abundance in crustal material and thus signals are low on filters that contain only milligrams of mineral dust, signals were detectable for most REE for most filter samples and were well above detection for pure dust samples. *Trapp et al.* [2010] highlighted the potential utility of such elements in an analysis of mineral dust at Miami, noting that elemental ratios change little between dust storms with the exception of REE. As such REE likely have utility in apportioning source, potentially on subcontinental scales. In this analysis not all REE were available for analysis in available calibration fluids, so a complete analysis was not possible. In future analyses REE should be fully run.

3203 6.6.5 SEARCH Utilization

The ability to use principal component analysis to separate source regions on elemental ratios will be applied near real time to samples from the SEARCH network in 2013 during mineral dust outbreaks in the Southeastern United States. Aliquots from pure dust samples used in this study are being sent to Atmospheric-Research in Cary, NC for complete digestion and analysis via ICP-MS receiving the same treatment that field filters samples receives. This data will be analyzed and PCA performed and a predictive schemed develop to identify dust sources in near real time as ICP-MS analysis is performed on field samples.



Figure 6.1: Regions as separated in *Formenti et al.* [2011] and shown in Table 6.1 and 6.2.



Figure 6.2: Position of sampling stations for the SEARCH project.



Figure 6.3: (top left) Digestion set-up both pure dust and filter samples placed in acidwashed Teflon beakers covered with Teflon eye glasses. (top right) Pure dust sample in aqua regia digestion phase. (bottom left) Pure dust sample after dry down, black carbon shown as residual. (bottom right) Teflon filter after acid digestion before final rinse.



Figure 6.4: Sample images from NSLS at BNL. (top) Example of intensity patterns from μ x-ray diffraction. (bottom) Examples of μ x-ray refraction of sample Teflon filters (left) Fe, (right) Pb.



Figure 6.5: Matrix of scatterplots showing elemental ratios normalized to Al. Asian samples are blue, African samples are red and Australian samples are green.



Figure 6.6: Matrix of scatterplots showing elemental ratios normalized to Al. Asian samples are blue, African samples are red and Australian samples are green



Figure 6.7: Matrix of scatterplots showing elemental ratios normalized to Ti. Asian samples are blue, African samples are red and Australian samples are green



Figure 6.8: Matrix of scatterplots showing elemental ratios normalized to Ti. Asian samples are blue, African samples are red and Australian samples are green



Figure 6.9: PCA on elemental ratios normalized to Al. (a) The first and second principal components. (b) The second and third principal components. (c) A three-dimensional visualization of the principal components. (d) A biplot of the relative magnitudes of the principal components and their grouping. The three principal components express 75% of the total variance of the elemental data.



Figure 6.10: PCA on elemental ratios normalized to Ti. (a) The first and second principal components. (b) The second and third principal components. (c) A three-dimensional visualization of the principal components. (d) A biplot of the relative magnitudes of the principal components and their grouping. The three principal components express over 95% of the total variance of the elemental data.


Figure 6.11: As in Figure 6.9 PCA on elemental ratios normalized to Al, with filter samples included. The three principal components express over 68% of the total variance of the elemental data.



Figure 6.12: As in Figure 6.9 PCA on elemental ratios normalized to Ti, with filter samples included. The three principal components express over 90% of the total variance of the elemental data.



Figure 6.13: (top) Absorbing Index (AI) from TOMS/Earth-Probe for July 29 and July 30, 2003. (bottom) Back-trajectories for 12z July 29, 2003 (left) and 18z July 29, 2003 (right). These images correspond to filter 03-T10914.



Figure 6.14: (top) Absorbing Index (AI) from TOMS/Earth-Probe for June 29 and June 30, 2003. (bottom) Back-trajectories for 12z June 29, 2003 (left) and 18z June 29, 2003 (right). These images correspond to filter 03-T9329.



Figure 6.15: (top) Absorbing Index (AI) from TOMS/Earth-Probe for August 22 and August 23, 2004. (bottom) Back-trajectories for 12z August 22, 2004 (left) and 18z August 22, 2004 (right). These images correspond to filter 04-T11260.



Figure 6.16: (top) Absorbing Index (AI) from TOMS/Earth-Probe for August 25 and August 26, 2004. (bottom) Back-trajectories for 12z August 25, 2004 (left) and 18z August 25, 2004 (right). These images correspond to filter 04-T11261.



Figure 6.17: (top) Absorbing Index (AI) from TOMS/Earth-Probe for June 23 and Jun 24, 2004. (bottom) Back-trajectories for 12z June 23, 2004 (left) and 18z June 23, 2004 (right). These images correspond to filter 04-T9429.



Figure 6.18: Example of separation of continental sources (top) and regional sources (bottom) using isotopic ratios of Nd and Sr after *Grousset and Biscaye* [2005]

North Africa			Asia				
Source	<u>Illite</u> kaolinite	<u>Chlorite</u> kaolinite	Carbonate	Source	<u>Illite</u> kaolinite	<u>Chlorite</u> kaolinite	Carbonate
area	ratio	ratio	$(\mathrm{wt}\%)$	area	ratio	ratio	$(\mathrm{wt}\%)$
NAF-1	1.0 - 2.0	1.5	10 - 50	EAS-1	12.5 - 13.9	3.3	6-35
	[PA, C2]	[PA]	[PA]		[S5]	[S5]	[W5, LI,
							W8]
NAF-2	> 1.6	0.0-0.8	5 - 70	EAS-2			0-10
	[AV, C2,	[GL, AV,	[AV, KH,				[W5, S6, LI,
	KA]	KA]	KA]				W8]
NAF-3	0.3 - 0.7	0.2 - 0.9	0 - 55	EAS-3	7.0 - 8.0	2.0 - 2.2	4-13
	[PA, C8]	[PA, AL]	[PA, FA,		[S6]	[S6]	[W5, S6, LI,
			A5]				W8]
NAF-4	0.2 - 1.9	0.0 - 2.6	1 - 25	EAS-4	1.1 - 6.0	1.3 - 3.0	0–5
	[OH]	[OH]	[OH]		[BI, SV]	[BI, SV]	[W5, JE]
NAF-5	0.0 - 0.4	b.d.	b.d	EAS-5	5.0 - 5.6	1.4	0-10
					[S5]	[S5]	[W5, LI,
							JE, W8]
NAF-6	0.7			EAS-6	7.5 - 9.7	1.7 - 2.0	< 1
	[C2]				[S5]	[S5]	[W5, LI, JE]

Table 6.1: Table of mineralogy by region from [Formenti et al., 2011]. The references are: AL: Alastuey et al. [2005], AV: Avila et al. [1997], BI: Biscaye et al. [1997], C8: Caquineau et al. [1998], C2: Caquineau et al. [2002], FA: Falkovich et al. [2001], GL: Glaccum and Prospero [1980], JE: Jeong [2008], KA: Kandler et al. [2009], KH: Khiri [2004], LI: Li et al. [2007], OH: OHara et al. [2006], PA: Paquet et al. [1984], S5: Shen et al. [2005], S6: Shen et al. [2006], SV: Svensson et al. [2000], W5: Wang et al. [2005], W8: Wang et al. [2008].

North Africa			Asia				
Source	Ca/Al	Fe/Al	K/Al	Source	Ca/Al	Fe/Al	K/Al
NAF-1		0.43 - 0.70		EAS-1	2.04 - 4.14	0.87 - 1.74	0.54 - 0.94
		[CS, B2]			[Z2, MA]	[Z2, MA]	[Z2, MA]
NAF-2	0.60 - 0.92	0.54 - 1.24	0.1 – 0.3	EAS-2	1.69	0.48	0.43
	[B1, CI,	[B1, B2,	[B1, CI,				
	F3, F8]	CI, F3, F8]	F3, F8]				
NAF-3	0.36 - 0.40	0.35 - 1.69	0.22	EAS-3	0.94 - 1.80	0.65 - 3.53	0.26 - 0.44
	[CI, AL]	[B2, CI,	[CI, AL]		[Z1, Z2]	[Z1, Z2]	[Z1, Z2]
		AL]					
NAF-4		0.55 - 0.65		EAS-4	0.42	0.55	
		[GU]			[SU]	[SU]	
NAF-5	0.4	0.6	0.3	EAS-5	0.67 - 2.43	0.36 - 1.60	0.20 - 1.00
	[F8]	[F8]	[F8]		[AF, Z1,	[AF, Z1,	[AF, Z1,
					Z2, AR,	Z2, AR,	Z2, AR,
					XU, SU]	XU, SU]	XU, SU]
NAF-6	0.75	1.08	0.24	EAS-6	0.31 - 0.94	0.70 - 1.07	0.36 - 0.80
	[EL]	[EL]	[EL]		[CG, SH]	[CG, SH]	[CG, SH]

Table 6.2: Table of elemental ratios by region from Formenti et al. [2011]. The references are: AL: Alastuey et al. [2005], AF: Alfaro and Gomes [2001], AR: Arimoto et al. [2006], B1: Bergametti et al. [1989a], B2: Bergametti et al. [1989b], CG: Cheng et al. [2005], CS: Chester et al. [1984], CI: Chiapello et al. [1997], EL: Eltayeb et al. [1993], F3: Formenti et al. [2003], F8: Formenti et al. [2008], GU: Guieu et al. [2002], MA: Makra et al. [2002], SH: Shen et al. [2007], SU: Sun et al. [2005], XU: Xu et al. [2004], Z1: Zhang et al. [2003], Z2: Zhang et al. [2003].

Sample	Sample Mass (g)	Sample Location	Sample Origin
Australia	0.0191	Australia	Australia
Australia Rep	0.01114	Australia	Australia
BCN	0.01554	Europe	N. Africa
GOST	0.01707	Europe	N. Africa
Green	0.01119	Europe	N. Africa
Green Rep	0.01065	Europe	N. Africa
Handan	0.01691	Asia	Asia
Inner Mongolia	0.01198	Asia	Asia
Inner Mongolia Rep	0.01774	Asia	Asia
Nanjing	0.01369	Asia	Asia
POLS1	0.01423	Europe	N. Africa
POLS2	0.01152	Europe	N. Africa
POLS3	0.01598	Europe	N. Africa
RED	0.01259	Europe	N. Africa
Sahara Bulk	0.02128	Africa	N. Africa
03-T10914	-	N. America	Unknown
03-T9329	-	N. America	Unknown
04-T11260	-	N. America	Unknown
04-T11261	-	N. America	Unknown
04-T9429	-	N. America	Unknown

Table 6.3: Table of samples analyzed. Columns are sample names as used in this study, mass of sample used in digestion (g), where the sample was acquired and the assumed source of the sample. Bottom five rows represent filters of unknown provenance.

	Instrument	Instrument	Pure Dust Di-	Filter Diges-
	Blank to Pure	Blank to	gestion Blank	tion Blank to
	Dust	Filter	to Pure Dust	Filter
Pb	0.00%	0.06%	0.40%	27.28%
U	0.03%	1.81%	0.14%	17.58%
Mg	0.01%	1.03%	0.28%	37.27%
Al	0.00%	0.09%	0.05%	36.65%
Si	11.71%	45.54%	4.44%	9.57%
\mathbf{S}	0.42%	1.41%	1.94%	4.94%
Ca	0.01%	1.97%	0.20%	265.56%
Ti	0.02%	0.17%	0.42%	10.92%
V	0.02%	0.17%	2.87%	27.57%
Mn	0.00%	0.03%	6.85%	544.82%
Fe	0.00%	0.01%	9.34%	83.80%
Ni	0.00%	0.00%	93.02%	261.57%
Cu	0.01%	0.08%	9.47%	116.42%
Zn	0.00%	0.01%	71.26%	134.56%
Rb	0.07%	2.32%	0.09%	5.24%
Sr	0.05%	3.66%	0.21%	48.22%
Ba	0.04%	1.50%	0.25%	21.61%
Ce	0.00%	0.18%	0.03%	4.83%
\Pr	0.01%	0.36%	0.03%	4.77%
Nd	0.00%	0.29%	0.03%	4.87%
Sm	0.01%	1.10%	0.03%	4.22%
$\mathbf{E}\mathbf{u}$	0.81%	41.69%	0.77%	21.23%
Gd	0.41%	16.18%	0.15%	-3.71%
Dy	0.01%	0.67%	0.03%	4.95%
Но	0.04%	2.88%	0.03%	4.94%
Er	0.02%	1.15%	0.04%	4.59%
Tm	0.09%	6.29%	-0.01%	1.65%
Yb	0.03%	2.06%	0.04%	9.63%
Lu	0.07%	5.33%	0.03%	9.07%
Th	0.03%	5.65%	0.05%	6.24%

Table 6.4: Table of procedural and analytical blanks by metal. The second and third columns shows the ratio of instrument blanks to median pure dust and filters samples as a percentage respectively. The fourth and fifth columns show the ratio of the digestion blanks to median pure dust and filter samples as a percentage respectively.

Sample Source	Dust ID	Mineral
North Africa	1	no match
	2	no match
	3	no match
	4	no match
	5	no match
(POLS 3)	6	calcite
	7	muscovite
	8	no match
	9	no match
	10	no match
N. Africa	11	kaolenite
	12	no match
	13	no match
(4GOST)	14	no match
	15	dolomite
	16	dolomite
Asia	17	no match
	18	FeTi hydrate
	19	FeO
	20	no match
(Naniing)	21	no match
(Ivanjing)	22	biotite
	23	dolomite
	24	Al oxide
	25	Quartz (w/ Fe impurities)
	26	no match
	27	muscovite
(Australia)	28	aluminosilicate
(mastralia)	29	silica
	30	TiO
	31	magnesite
Asia	32	no match
	33	no match
(Inner Mongolia)	34	Aluminosilicate, TiO
(innor wongona)	35	dolomite
	36	iron oxide
Asia	37	no match
	38	calcite
(Handon)	39	rutile (TiO)
	40	magnetite

Table 6.5: Table of mineralogy from μ -XRD. Columns are samples, sample number, and mineral as identified by MATCH! software. A value of "no match" indicates a pattern which could not be matched to any known mineral diffraction pattern.

Samples	W_{ϕ}	W_{ϕ}
T9329	18.031	1.55
T10914	18.13	-0.08
T11260	17.56	-1.26
T11261	18.49	-0.33
T9492	17.86	-0.36

Table 6.6: Position of WACZ (W_{ϕ}) for each sample over the period 7 to 14 days prior. The anomaly (W_{ϕ} ') calculated relative to trend corrected climatological position of WACZ over 20th century.

³²¹¹ Chapter 7

3212 Conclusion

3213 7.1 Summary of Key Findings

The reasons for the inter-annual variability of dust transport from the Sahara across the 3214 Atlantic were previously not well-understood. This work has provided insight into the mech-3215 anisms that govern the interannual variability of mineral dust transport. To obtain these 3216 results it was necessary to develop a new climate index that tracks the north and south 3217 migration of the ITCZ over West Africa. The utility of this index was demonstrated by 3218 applying it in three different ways; (1) to the record of mineral dust load at Barbados, (2) to 3219 reanalysis and GCM products of the 20th century and (3) to projections of the 21st century 3220 in GCM output. We found a; (1) robust relationship between the latitude of the ITCZ and 3221 dustload at Barbados, (2) varying degrees of skill at GCM at recreating the 20th century 3222 record as seen in reanalysis and a northward trend in the latitude of the ITCZ in summer 3223 and (3) a strong northward trend in all seasons in the 21st century, with implications for 3224 a reduction in mineral dust transport. Last (4) using mineralogical and elemental ratios 3225 of collected mineral dust samples from multiple continents we constructed a framework for 3226 separating continental sources of mineral dust. 3227

In this work we addressed this issue of variability by creating a new climate index that

captures the position and intensity of the zone of near-surface convergence over West Africa, 3229 a part of the global Intertropical Convergence Zone (ITCZ). We then related this index to 3230 a 38-year record of mineral dust concentrations at Barbados first focusing on the winter 3231 season. The results showed that the latitudinal displacement of the ITCZ over West Africa 3232 and the dust load in Barbados are statistically significantly correlated with a correlation 3233 coefficient of r = -0.69. A southward movement of the ITCZ corresponds to an increased 3234 dust load at Barbados. This correlation represented an improvement upon previous results, 3235 which focused on traditional teleconnection indices such as the North Atlantic Oscillation 3236 or the El-Niño-Southern Oscillation. From analyzing composites of wind and precipitation 3237 we concluded that for the winter season, the inter-annual variability of the Barbados dust 3238 load is related to changes in near-surface northeasterly winds in semi-arid regions in North 3239 Africa coincident with the movement of the ITCZ. Changes in precipitation appear to only 3240 play a minor role. 3241

We next extended the "center of action" indices for the ITCZ over West Africa to the 3242 inter-annual variability of dust transport from the Sahara to Barbados in summer. As 3243 in the winter case, previous efforts to explain the variability of summer season mineral 3244 dust amount focused on teleconnection indices such as ENSO and the NAO. However only 3245 weak relationships between such climate indices and the abundance of mineral dust were 3246 found. The latitudinal position of the ITCZ is significantly correlated with the quantity 3247 of mineral dust at Barbados over the period 1965 to 2003 (r = -0.47) in summer. A 3248 southward displacement of the ITCZ is associated with both increased near-surface flow 3249 and decreased precipitation over the dust emitting regions of the southern Saharan desert, 3250 Sahel and Lake Chad. This in turn reduces soil moisture and vegetation furthering the 3251 potential for dust emission. In contrast, the intensity of the ITCZ is not correlated with 3252 dust concentration at Barbados. Analyzing this "null case," we conclude that the coupling 3253 of changes in near-surface winds with changes in precipitation in emission regions driven by 3254 a southward movement of the ITCZ most directly influence dust load at Barbados and the 3255

3256 Caribbean during summer.

Having shown that the latitudinal position of the ITCZ over West Africa is important 3257 for mineral dust emission and transport in both winter and summer seasons, we next exam-3258 ined how multiple reanalysis products and global climate models from CMIP3 compared in 3259 determining in the latitude of the ITCZ over the period 1978 to 2002. Reanalysis products 3260 performed similarly to each other in the abstract, but when temporal variability is compared 3261 significant differences emerged. Substantial differences in the spatial field of 925 hPa diver-3262 gence between reanalysis and GCM were identified over North Africa. The ITCZ index was 3263 calculated using the output of each model and the each reanalysis product, which allowed 3264 for a quantitative comparison. Analysis of ITCZ in both reanalysis and model showed a 3265 significant northward trend in the position of the ITCZ over the 20th century in summer. 3266

While reanalysis products were similar in their representation of the ITCZ latitude, our 3267 analysis showed that there was a large range in performance of the global climate models in 3268 representing this quantity. In particular we found global climate models were unable to accu-3269 rately quantify the interannual variability of the ITCZ and did not replicate trends observed 3270 in reanalysis. Lastly we provided an overview of each models performance and potential 3271 suitability for future projections. We identified two models that performed consistently with 3272 reanalysis products, and three models which exhibited poor skill at placing the latitude of 3273 the ITCZ over West Africa. 3274

Further, we calculated the ITCZ latitude index using CMIP GCM model output from sim-3275 ulations of the 21st century. Two scenarios were considered; SRES-A1B, in which greenhouse 3276 gases levels continued to rise steadily in the 21st century, and SRES-B1 in which greenhouse 3277 gas emission was moderated. A large southward discontinuity in the divergence field was 3278 noted when model results were compared to the 20th century, which was attributable to 3279 changes in ocean conditions in GCMs. A strong northward trend of the WACZ was iden-3280 tified for all seasons in both scenarios for the whole of the 21st century. We extrapolated 3281 the trend in WACZ to observed variability between W_{ϕ} and mineral dust at Barbados and 3282

predicted reductions of mineral dust between $-1.9 \ \mu g m^{-3}$ and $-3.8 \ \mu g m^{-3}$ in summer and $-1.5 \ \mu g m^{-3}$ and $-2.2 \ \mu g m^{-3}$ in winter. Model performance in the 20th century was not related to the trend predicted in the 21st century.

Last, we attempted to verify our hypothesis by applying geochemical techniques to min-3286 eral dust samples. Mineral dust samples from Africa, Asia, Europe and Australia were 3287 acquired and analyzed for their mineralogy and elemental composition. Filter samples of 3288 unknown provenience collected in Florida were similarly analyzed. To differentiate source 3289 regions principal component analysis was applied to elemental composition. The results 3290 showed that African, Asian and Australian dust can be separated in this manner. Principal 3291 components were shown to correspond to three mineral groupings; carbonate, aluminosili-3292 cates and iron/titanium oxides. Filter samples from Florida were shown to be dissimilar to 3293 the pure African, Asian and Australian signals, possibly because high blanks obscured the 3294 dust source signal. 3295

³²⁹⁶ 7.2 Implications of Research

Our key finding that the latitude of the WACZ controls a large fraction of the variability of 3297 mineral dust transport to Barbados in both winter and summer represents a new paradigm for 3298 explaining seasonal dust transport. It also represents a major improvement on prior results 3299 utilizing other teleconnection indices such as the NAO and ENSO. This finding confirms other 3300 previous studies; for example the improvement over ENSO is consistent with Mahowald et al. 3301 [2003] who did not find correlations in West Africa between dust and ENSO. Our results 3302 indirectly confirm the findings of *Engelstaedter and Washington* [2007] who found a spatial 3303 correlation over North Africa between 10 m-divergence and satellite estimates of dustload. 3304 Our results directly confirm the hypothesis of Schwanghart and Schütt [2008] who argued 3305 that northward movement of the ITCZ could diminish NE winds over the Sahara and Sahel. 3306 While this work focuses on the role of the WACZ in explaining mineral dust transport, 3307

such an index has been applied by Taylor et al. [2012] to biogeochemical and primary pro-3308 ductivity data in the Cariaco Basin and find meaningful relationships. This index likely can 3309 be applied to many other environmental datasets in North Africa and surrounding regions. 3310 In the summer during the 20th century a clear northward trend of the WACZ was ob-3311 served in reanalysis and GCM output; this northward trend continued in the 21st century, 3312 extending to both summer and winter over this period. This finding agrees with basic the-3313 ory surrounding the Hadley Cell, which posits a poleward expansion of the Hadley Cell in a 3314 warming climate [Polvani and Kushner, 2002; Hu and Fu, 2007]. Here our work focuses on 3315 the ascending branch of the Hadley Cell over West Africa. Our finding of poleward expansion 3316 of the ascending branch compliments the work of *Gastineau et al.* [2008] who found evident 3317 for equatorward and poleward contraction/expansion of the ascending branch of the Hadley 3318 Cell in climate models. 3319

As northward migration of the WACZ leads to reduced dust transport, and the WACZ is expected to drift northward as the climate warms in the 21st century, then mineral dust transport should be expected to diminish in the warming climate. Paleoclimatologists have long argued for the paradigm that cold, glacial periods are dusty and warm, interglacial periods are less dusty. Here our new paradigm is a direct mechanism for which this argument can be explained on a regional scale, with a proven physical mechanism.

Utilizing the mineralogy and elemental composition of dust samples procured from source 3326 regions, a framework has been developed to separate continental sources. This result sup-3327 ports the supposition made in *Formenti et al.* [2011] that mineralogy on regional and conti-3328 nental scales can be useful in identifying dust sources. Trapp et al. [2010] found unchanging 3329 elemental ratios of major metals in Barbados and Miami dust samples, our results suggest 3330 that this implies a single continental source for this dust. The framework of continental 3331 source separation has practical applications; this analysis will be repeated and applied real 3332 time to suspected mineral dust outbreaks across the Southeastern United States moving 3333 forward. 3334

3335 7.3 Future Work

³³³⁶ This work has opened up new opportunities for further research in the following areas:

3337 **7.3.1** Timescale

Mineral dust is directly emitted to the atmosphere via turbulent wind gusts, which occur on 3338 the timescale of seconds to minutes. Emitted dust is lofted above the boundary layer over 3339 the course of hours. A particularly dusty air layer can form a Saharan Air Layer over the 3340 course of days. A dust layer can be transported across the Atlantic Ocean from Africa over 3341 the course of 1 to 2 weeks. Each of these processes responsible for the emission, persistence 3342 and transport of mineral dust occurs on timescales shorter than the seasonal timescale that 3343 is the focus of this work. A natural extension of this work would be to examine how the 3344 location of the WACZ is related to physical drivers associated with short timescales such 3345 as gustiness and stability. One potential mechanism would be to examine if the WACZ is 3346 in some way related to African Easterly Waves, which represent a potential mechanism by 3347 which to relate short term variability to seasonal scale variability. 3348

³³⁴⁹ 7.3.2 Assessment of more recent CMIP model results

The global climate models analyzed in this work are products of the Climate Model Intercomparison Project 3 (CMIP3). At the time this work was initiated CMIP3 represented the best choice for analysis. In the interim CMIP5 has been released. CMIP5 utilizes, in general, higher resolution models (in both space and time) as well as an improvement on model physics. CMIP5 may include some dust-climate feedbacks which could be of importance in setting the position of the WACZ. Before this work can be prepared for publication, it should be repeated with CMIP5 model products.

³³⁵⁷ 7.3.3 Mechanism for control of WACZ in Climate Models

We have found both a large southward shift in the WACZ in 21st century model runs and large biases relative to reanalysis in many 20th century model runs. This work could be extended to the mechanisms by which these shifts and biases occur and begin to investigate how changes in model SST impact the WACZ and how cross-equatorial pressure gradients impact the WACZ.

3363 7.3.4 Geochemistry Analysis

From the beginning it was expected that the geochemistry section of this work would be an analytical challenge, accounting for the small quantities of mineral dust expected to be found on the filters of unknown provenance. Below are four suggestions for improving such a study in the future.

3368 North American Samples

Filter samples, in general, did not fall within the envelope of elemental composition as seen in Asian, African or Australian mineral dust samples. While this result could be due to method or analysis errors, it could also be indicative of a local source of mineral dust not captured by our source region. Future analysis should include mineral dust samples both from North American desert sources and local soil sources.

3374 Comparing Different Digestions

Elemental composition as measured by ICP-MS is no doubt strongly influenced by the acids and digestion methods selected. A comprehensive study should include a sensitivity analysis to digestion method. Filters should be prepared with a known quantity of mineral dust applied to the surface. In this way recovery the digestion method could be quantified.

3379 Isotopic Analysis

This work is substantially weakened by the absence of an isotopic analysis. Samples are prepared and waiting for chemical separation of Pb, Sr and Nd. Once separation is performed and a suitable instrument is found, such information will be invaluable in overcoming blank issues associated with the elemental analysis. In addition to traditional geochemical mixing curve analysis isotope data can be included as a predictor in the principle component analysis. It is anticipated that this work will be performed prior to publication of this work.

3386 Blank Reduction

Filters were taken to the National Synchrotron Light Source at Brookhaven National Lab 3387 for micro x-ray diffraction and florescence studies. These methods were chosen as they 3388 are non-destructive, the filters could subsequently be digested an analyzed for elemental 3389 composition. However in this process, the filters were exposed in a non-clean environment, 3390 and contamination could possibly have been introduced. This could represent a major source 3393 of trace metals, and in turn register erroneous elemental composition. In future work two 3392 filters should be taken side by side, one preserved in a clean environment for digestion and 3393 the other taken for diffraction studies. Alternatively captured filters could be physically split 3394 for different analyses. 3395

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