Interannual Variability in the Source Location of North African Dust Transported to the Amazon

Anne E. Barkley, Ali Pourmand, Jack Longman, Arash Sharifi, Joseph M. Prospero, Kathy Panechou, Natalie Bakker, Nick Drake, Damien Guinoiseau, Cassandra J. Gaston

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Key Points:

- Western North African dust sources dominated transport to the Amazon in winter 2014 while Central dominated in winter 2016
- Interannual variability in dust source is likely due to differences in meteorology and precipitation
- Sources of North African dust transported to the Atlantic Ocean and Amazon could change as climate changes altering nutrient deposition

Supporting Information:

Supporting Information may be found in the online version of this article.

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Interannual Variability in the Source Location of North African Dust Transported to the Amazon

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Abstract African dust is transported to South America (SA) every winter and spring. Hypotheses suggest that either Western or Central North Africa (e.g., Bodélé Depression) is the main source of transported dust, yet these notions remain largely untested with geochemical data. Using 2 years of isotopic measurements (strontium and neodymium) of African dust collected in SA integrated into a statistical model, we identified strong interannual variability in dust source region. Central North Africa supplied 44% of long-range transported dust in winter 2016 while the Western region accounted for 53% of dust in winter 2014. We propose the variability is due to differences in the strength of the Libyan High and precipitation over the Gulf of Guinea and Atlantic Ocean between the 2 years. Our findings can improve constraints of dust nutrient deposition and predictions of how changes in climate impact the source and magnitude of dust transported to the Amazon.

Plain Language Summary Dust is blown from North Africa to the Amazon and the western Atlantic Ocean in winter and spring. Identifying the area within North Africa where dust originates is the subject of intense debate, with leading hypotheses disagreeing on whether the Bodélé Depression (Central North Africa) or Western North Africa provide dust that fertilizes the Amazon. Here, we present a more nuanced hypothesis: dust from *both* the Central and Western North African dust source regions are important with the dominate source depending on the prevailing meteorological factors and precipitation patterns. Our hypothesis is supported by isotopic fingerprinting that was used to determine the source area of dust transported to South America (SA). We then integrated our data into a statistical model, which quantifies the proportion of dust from each North African region that contributes to the dust we collected in SA. Because dust source location dictates the amount and solubility of associated nutrients, changes in dust source impacts nutrient deposition to the Amazon and Tropical Atlantic Ocean. Additionally, identifying the factors that control dust source location can provide clues into how past and future changes in climatological conditions affect dust transport.

1. Introduction

Aeolian dust is produced perennially across numerous potential source areas (PSAs) in North Africa, and it is transported to the Amazon and western Tropical Atlantic Ocean (TAO) in boreal winter and spring (Prospero et al., 1981). Transported dust is thought to alleviate nutrient limitations in the TAO and Amazon Basin, fueling net primary productivity and sequestering carbon dioxide into the biosphere. Because the source location of dust controls the magnitude, mineralogy, and solubility—a proxy for bioavailability—of associated nutrients such as phosphorus (P) and iron (Fe), it is important to identify the PSA(s) of transported dust (Barkley et al., 2019; Journet et al., 2008; Okin et al., 2004; Scheuvens et al., 2013; Swap et al., 1992).

Identification of dust PSAs can be used to validate dust transport models, which struggle to accurately predict transport to different receptor ecosystems in past and present climate regimes (Evan et al., 2014; Huneeus et al., 2011; Mahowald et al., 1999, 2010; Pu & Ginoux, 2018; Wu et al., 2020). Improved predictions would aid in understanding dust-climate interactions, not limited to biogeochemical effects. Moreover, climate change will affect atmospheric circulation as well as wind and precipitation patterns, altering the primary locations of dust emission, transport, and deposition (Goudie & Middleton, 2001; Mahowald et al., 2007; Yuan et al., 2020).

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Therefore, determining the meteorological factors that control which PSAs contribute long-range transported dust in the present may provide clues into changes to dust source region(s) in the future.

Definitive identification of the PSAs that account for wintertime trans-Atlantic dust transport to the Amazon has remained elusive with several studies disagreeing with one another (Abouchami et al., 2013; Ben-Ami et al., 2010; Gläser et al., 2015; Koren et al., 2006; Y. Yu et al., 2020). Difficulties in determining the PSA(s) of dust arise due to emission, transport, and deposition processes, which are difficult to constrain using models and remote sensing (Adebiyi & Kok, 2020; H. Yu et al., 2015; A. Zhao et al., 2022). For example, the earliest remote sensing studies identified the Bodélé Depression within Central North Africa as the largest source of dust in the world and the major dust contributor to the Amazon (Koren et al., 2006; Washington et al., 2003). However, a clear geochemical signature that would indicate dust contribution from the Bodélé has not been conclusively identified in Amazonian soil (Abouchami et al., 2013).

Western North African sources have been suggested to supersede transport from Central North Africa, potentially due to losses of Central North African dust to wet deposition (Goudie & Middleton, 2001; Knippertz & Todd, 2010; Y. Yu et al., 2018, 2020). While wintertime measurements of dust collected in the Cape Verde Islands support this assertion (Kumar et al., 2018), dust collected in the Caribbean in boreal summer suggests that southern regions of Central and Western North Africa (Sahel) prevail (Bozlaker et al., 2018; Pourmand et al., 2014; van der Does et al., 2018). In the absence of long-term isotopic analysis of wintertime African dust collected in South America (SA), it is difficult to settle this disagreement.

In this work, we aim to resolve this issue with a multi-year analysis of the isotopic composition of dust transported to South America (SA). Aerosol samples were collected at our field site in Cayenne (Figure 1) and can be used to extrapolate transport to the Amazon because air masses that pass over Cayenne continue into the interior of Amazon (Barkley et al., 2019; Pöhlker et al., 2019), though the magnitude of dust may decrease substantially (Prospero et al., 2020). Using radiogenic strontium (Sr) and neodymium (Nd) isotopes in combination with a Bayesian mixing model, we quantified the relative importance of each PSA to dust transported to Cayenne. The isotopic ratios (e.g., $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$, denoted as ε_{Nd}) of each PSA is controlled by the geochemical evolution of the underlying bedrock, which can be associated with the history of its formation (Banner, 2004; Grousset & Biscaye, 2005; Grousset et al., 1992; Scheuvens et al., 2013). Our analysis makes it possible to describe seasonal and interannual variability of dust sources, and we propose a hypothesis for the annual change in dust source. Our results can be used to validate both nutrient deposition estimates from biogeochemical models and dust emission in transport models.

2. Materials and Methods

2.1. Field Site and Sample Collection

Our field site is located north of Cayenne, French Guiana (4.92°N, 52.31°W) on a peninsula 67 m above sea level that receives minimal local emissions (Barkley et al., 2019; Prospero et al., 2014). Approximately daily samples were collected on Whatman-41 cellulose filters using a high-volume sampler with an average air flow rate of 0.75 m^3 min^{-1}. This study uses 2 years of samples from 2014 to 2016. Samples collected in 2015 were not used in this study because of gaps in the measurement record.

2.2. Dust Mass Quantification and Radiogenic Isotope Analysis

Dust mass concentrations were determined by rinsing a portion of the filter with Milli-Q (MQ) to remove soluble material, ashing the samples at 500°C overnight, and quantifying the ash gravimetrically (Prospero, 1999). Due to the daily resolution of our sampling protocol, there was often not enough dust on the daily samples to quantify Sr and Nd, resulting in samples from multiple days needing to be combined. Samples for isotope analysis were acid digested (Baker et al., 2006) and a two-stage chromatographic extraction scheme separated Sr and Nd from interfering matrix elements using Sr and TODGA cartridges from Eichrom Inc (Pourmand et al., 2014). Results are reported as a ratio of the radiogenic isotope (^{87}Sr) to the stable Sr isotope (^{86}Sr) to yield $^{87}Sr/^{86}Sr$. Samples were bracketed with measurements of SRM987 standard solutions, and they were adjusted relative to the accepted value to compare to other literature values. Nd ratios were also bracketed with two JNdi-1 standard solutions. The ratio of the radiogenic Nd isotope (^{143}Nd) was normalized to the stable Nd isotope (^{144}Nd) to yield $^{143}Nd/^{144}Nd$,

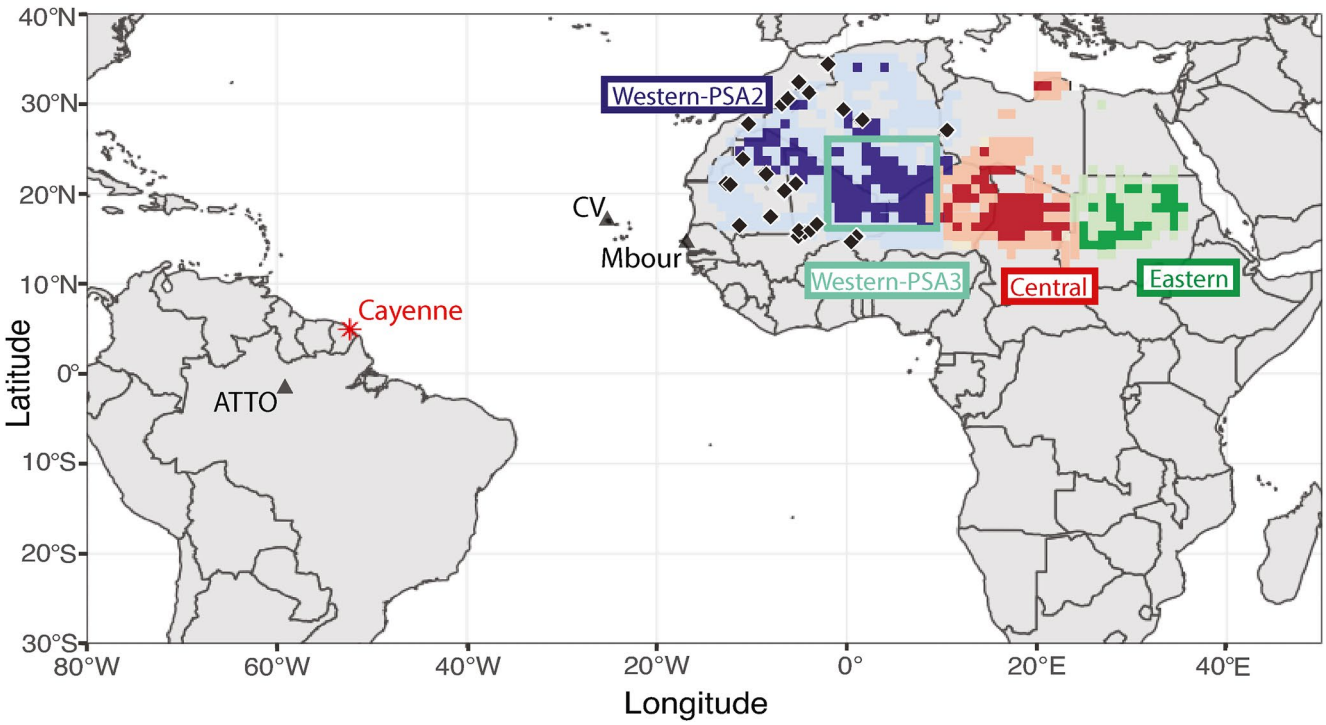

Figure 1. Map showing potential source areas (PSAs) of dust emission in North Africa. The Eastern, Central, and Western-PSA2 were adapted from Jewell et al. (2020) and Western-PSA3 from Kumar et al. (2014), Skonieczny et al. (2013), and Bakker et al. (2019). The bolder color represents locations (1° × 1°) where dust source activation is >5% and the lighter color represents <5% (Schepanski et al., 2012). The red asterisk shows the location of the field site where our samples were collected in South America. The gray triangles represent the locations of the Amazon Tall Tower Observatory, Cape Verde Atmospheric Observatory, and Mbour, Senegal.

which was normalized to the Chrondrite Uniform Reservoir value of 0.512638 (Jacobsen & Wasserburg, 1980) and reported in epsilon notation ($\varepsilon_{\mathrm{Nd}}$).

$$\varepsilon_{Nd} = \left[\frac{^{143}Nd/^{144}Nd_{\mathrm{Sample}}}{^{143}Nd/^{144}Nd_{\mathrm{CHUR}}} - 1\right] \mathrm{x}\ 10^4$$

All analysis was conducted using a Thermo Scientific Neptune Plus High-performance Multi-Collector Inductively Coupled Plasma Mass Spectrometer. Certified reference materials AGV-2 and SGR-1b were analyzed for analytical accuracy and precision. All measurements are reported in the data repository (ds01; Barkley et al., 2019; Prospero et al., 2020).

2.3. MixSIAR: A Bayesian Mixing Model

MixSIAR is a Bayesian mixing model developed for use in stable isotope studies (Stock et al., 2018; Stock & Semmens, 2016). By considering the dust compositions in aerosol samples as mixtures and the isotopic signatures of North African PSAs as sources, MixSIAR is able to estimate the mixture of dust from different North African PSAs that satisfy isotopic mass balance in our dust samples (Longman et al., 2022; see Figure S1 in Supporting Information S1). Through incorporating standard deviations of the sources, the model considers variation in the isotopic signature of PSAs when considering likely model fits. All model simulations were run using uninformative priors and assuming equal likelihood of all sources contributing to the collected dust. Because Nd and Sr isotopes are not linearly related in crustal rocks and sediments (Winter, 2001), Sr and Nd concentrations from the source PSAs (Castillo et al., 2008) are included in MixSIAR runs following Phillips and Koch (2002). A total of 3,000 Markov Chain Monte Carlo simulations were produced, resulting in potential outcomes that are plotted in Figure 3. More information is available in Supporting Information S1.

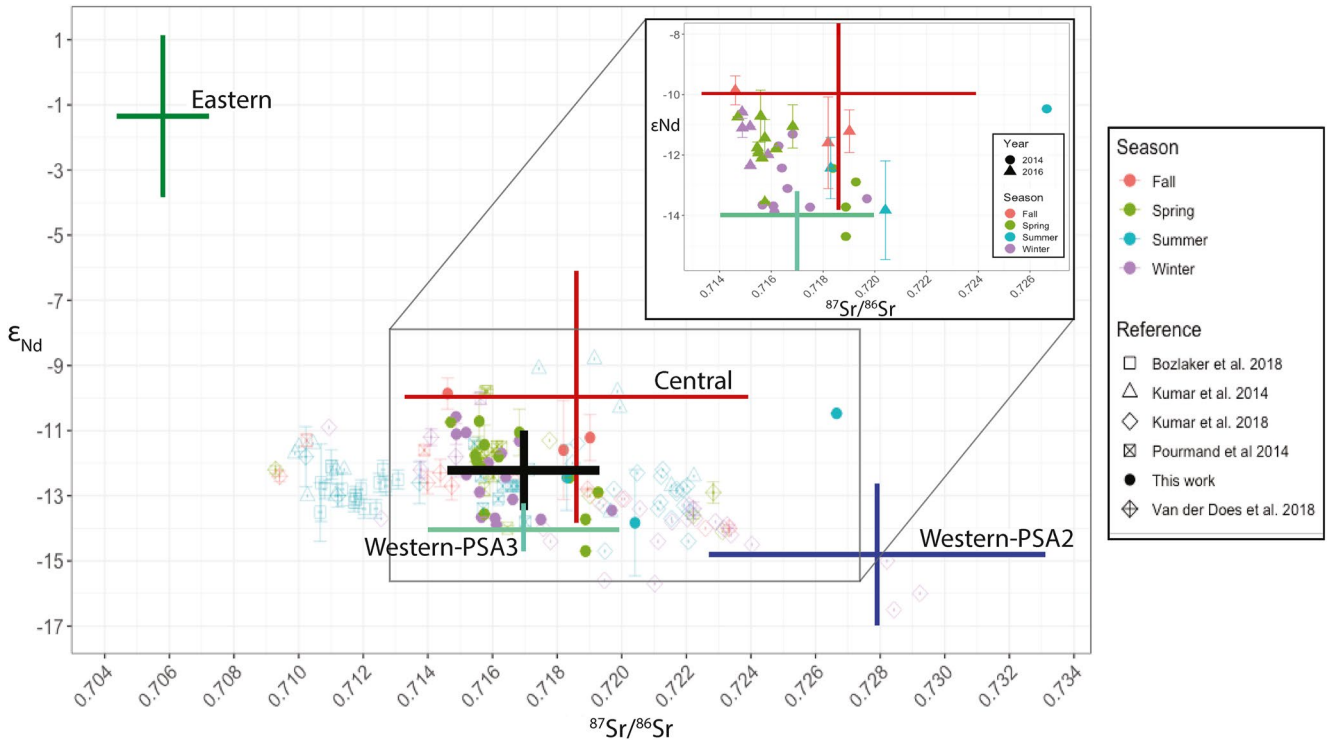

Figure 2. Results showing the $^{87}Sr/^{86}Sr$ and ε_{Nd} isotopic signature of samples collected in South America (This work; filled circles). The green, red, blue, and turquoise crosses correspond to the average $^{87}Sr/^{86}Sr$ and ε_{Nd} and one standard deviation (±1SD) and correspond to the source locations (potential source areas [PSAs]) shown in Figure 1. The isotopic signature of the Eastern, Central, and Western-PSA2 were adapted following Jewell et al. (2020). Due to the lack of samples from the Western-PSA3, its isotopic signature was inferred from samples collected nearby in Cape Verde (Kumar et al., 2018) and Mbour, Senegal (Skonieczny et al., 2013). The black cross shows the average and ±1SD of our data. Data from previously published literature of dust collected at receptor sites are shown in unfilled markers with colors that correspond to the season in which they were collected. Error bars on data points represent ±1SD, except for Pourmand et al. (2014) and this study, which shows 95% confidence intervals. When error bars are not present, SDs are smaller than their marker size. Winter is DJF, spring is MAM, summer is JJA, and fall is SON. The insert shows the section of the plot inside the gray box and further subdivides our data by year.

3. Results

3.1. Isotopic Fingerprinting of Dust Transported to the Amazon and Tropical Atlantic Ocean

The Western, Central, and Eastern North African PSAs are represented by the shaded boxes in Figure 1, which were geographically defined by Jewell et al. (2020). The isotopic domains of each of these sources are shown in the crosses in Figure 2 (Jewell et al., 2020). We further subdivide the Western PSA into two regions based on their distinct ages and geology and name them to be consistent with previous terminology (Formenti et al., 2011; Scheuvens et al., 2013). One is the Western-PSA2, which contains Precambrian outcrops and is defined as the only area inside the Western PSA from which there are samples with known Sr and Nd values (diamonds in Figure 1; Guiraud et al., 2005). The second region is the Western-PSA3, which contains younger, Paleozoic terrains. It is located within the turquoise box in Figure 1 but has no published Sr or Nd isotope values for soil material despite being previously identified as a considerable dust source using remote sensing and back trajectory analysis (Bakker et al., 2019; Feuerstein & Schepanski, 2019; Kumar et al., 2018; Pourmand et al., 2014; Schepanski et al., 2012; Skonieczny et al., 2013; Y. Yu et al., 2020). Western-PSA3 contains erodible silt-rich alluvium deposits derived from wadis inside a closed depression (Goudie & Middleton, 2001). The isotopic domain of the Western-PSA3 source (Figure 2) was inferred using dust collected off the western coast of Africa in Cape Verde and Mbour, Senegal, where air mass back trajectory analysis pointed to the Western-PSA3 as the source of dust (Kumar et al., 2018; Skonieczny et al., 2013). Though there is overlap in the Sr domain of the Central and Western-PSA3 sources, the Nd ratios are significantly different (p-value < 0.005). PSA source data are summarized in Table S1 in Supporting Information S1.

The isotopic composition of dust presented in Figure 2 (filled circles) is a multi-year geochemical fingerprinting analysis of aerosol samples collected in Cayenne. Previously published $^{87}Sr/^{86}Sr$ and ε_{Nd} values (open markers)

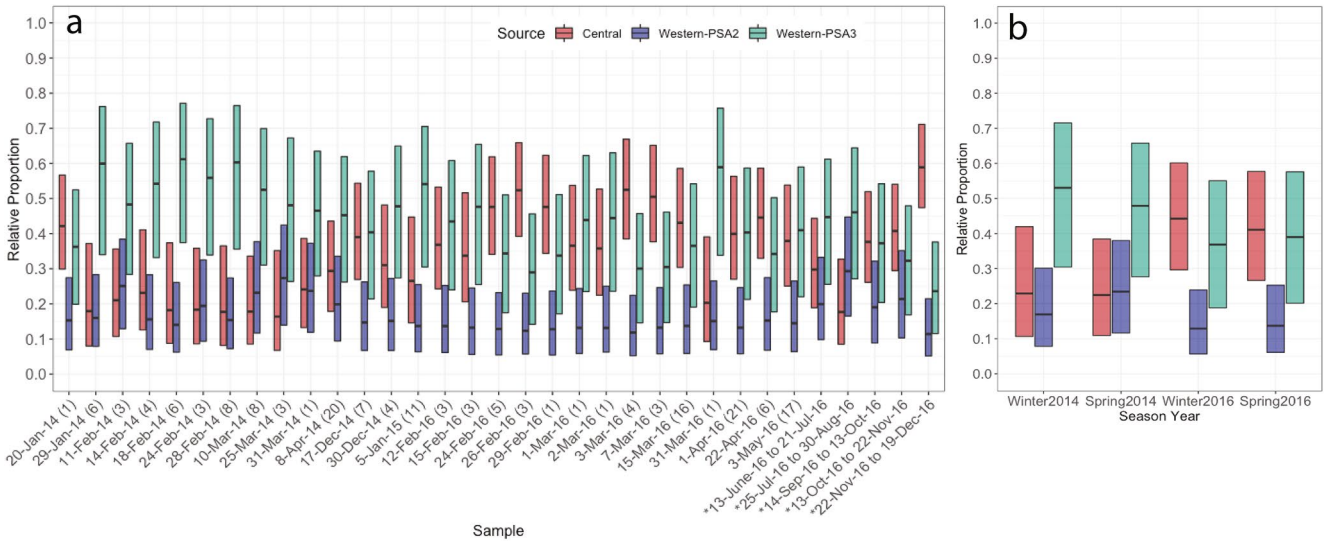

Figure 3. Panel (a) timeseries showing the proportion of each potential source area (PSA; Western-PSA2, Western-PSA3, Central) modeled by MixSIAR that explains our geochemical results measured in each aerosol sample ($n = 33$). The number in parentheses in the x-axis denotes the number of days dust was collected for each sample (e.g., sample collection occurred for 1 day for sample "20 January 2014"). Samples with an asterisk indicate approximately a month of sampling and the range of collection dates is listed. Panel (b) The relative proportion of each dust source PSA contributing to our aerosol samples collected during different years and seasons in Cayenne modeled by MixSIAR (same legend as panel a). Winter is DJF and spring is MAM.

are also plotted for reference and categorized by season of collection. Our sample values fall within the domain of the Central PSA; however, mixing with dust from other regions of North Africa cannot be ruled out because nearly all of our samples fall below the mean Sr and Nd isotopic composition of the Central PSA. Our data broadly agree with data collected at receptor sites in Barbados (Bozlaker et al., 2018; Pourmand et al., 2014; W. Zhao et al., 2018), other eastern Caribbean islands (Kumar et al., 2014), and sediment traps in the TAO (van der Does et al., 2018). Samples collected in winter and spring in the TAO are likely the closest comparison to our data because these samples were collected in the same season at a similar latitude (10°N). Indeed, these data show similar values to our measurements.

Samples analyzed in this study are not isotopically similar to measurements of dust from the Western-PSA2, but we cannot rule out mixing with this region. Previous studies have shown that strong winds pass over parts of the Western-PSA2 (Knippertz & Todd, 2012), which led to the hypothesis that this region can emit large quantities of dust (Kumar et al., 2018; Y. Yu et al., 2018). The Eastern PSA, likely an important source of dust for eastern Mediterranean and North African regions (Falkovich et al., 2001; Laskin et al., 2005), does also not appear to contribute dust to the western TAO at any point during the year (Figure 2).

Small amounts of dust reach the Amazon in the summer (JJA) and fall (SON) as detected in samples collected at the Amazon Tall Tower Observatory (ATTO) in the Central Amazon (Nogueira et al., 2021). A shift toward more radiogenic ε_{Nd} values in transported dust collected at Amazon Tall Tower Observatory (ATTO) in summer and fall is observed and the authors suggest this dust originated from southern Africa (Nogueira et al., 2021). In our samples, however, we see no comparable shift in ε_{Nd} values, which suggests dust transport to Cayenne is not from southern Africa during summer and fall, in agreement with findings from previous modeling and remote sensing studies (Kok et al., 2021; Prospero et al., 2020). The reason for the seasonal differences between Amazon Tall Tower Observatory (ATTO) and Cayenne is likely due to the location of Cayenne, approximately 1,000 km northeast of ATTO (Figure 1). Therefore, while Cayenne and ATTO likely receive dust from the same dust events in winter (DJF) and spring (MAM; Barkley et al., 2019; Pöhlker et al., 2019; Prospero et al., 2020), ATTO is affected by different sources than Cayenne in the summer and fall.

When winter and spring samples were categorized by collection year, the isotopic signatures suggest interannual variability in dust source region (inset of Figure 2). For example, samples analyzed in 2016 showed a more radiogenic ε_{Nd} signature (average $\varepsilon_{Nd} = -11.5$, $\pm 1SD = 0.83$, $n = 13$) than samples collected in 2014 (average $\varepsilon_{Nd} = -13.1$, $\pm 1SD = 0.89$, $n = 11$) and the difference between the 2 years were statistically different ($p < 0.005$).

Sr isotope ratios between the 2 years were also statically different ($p < 0.005$) with more radiogenic Sr isotope signature in samples collected in 2016 (average $^{87}Sr/^{86}Sr = 0.7176$, $\pm 1SD = 0.0006$ $n = 13$) than in 2014 (average $^{87}Sr/^{86}Sr = 0.7155$, $\pm 1SD = 0.0014$, $n = 11$).

3.2. Interannual Variability in the Source Location of Transported Dust

To quantify the contribution of dust transported to South America (SA) from each North African PSA, we used MixSIAR. The model requires all source PSAs are identified to apportion our samples. This *a priori* step is done by connecting the $^{87}Sr/^{86}Sr$ and ε_{Nd} values of each source PSA (endmember) to each other in Sr-Nd space, resulting in a mixing envelope (Figure S1 in Supporting Information S1). Samples that fall inside the mixing envelope are likely mixtures of the known PSAs while samples that fall outside indicate an unknown contributor.

Because the Central PSA and Western-PSA2 have been characterized using samples collected in these regions, we first used only these PSAs as endmembers. However, approximately one-third of our samples fell outside the mixing envelope, indicating a missing source(s) PSA containing unradiogenic Sr and Nd values is responsible for dust transport to Cayenne. We propose that the Western-PSA3 is the missing source (turquoise box, Figure 1; turquoise cross, Figure 2). Dust samples collected at receptor sites in Senegal and Cape Verde have been suggested to be from the Western-PSA3 (Kumar et al., 2018; Skonieczny et al., 2013), and samples collected in these locations have a similar isotopic signature as our missing source ($^{87}Sr/^{86}Sr = 0.716$; $\varepsilon_{Nd} = -14.5$). By including the Western-PSA3 source, 33 of 34 of our sample mixtures fall within the mixing envelope.

To validate our modeling approach, we compared our results to products from a suite of high-resolution remote sensing products (Schepanski et al., 2012). We investigated a dust storm initiated on 19 February 2016 in the Bodélé Depression (Figure S2 in Supporting Information S1) that reached Cayenne on 29 February 2016 (Figures S3a–S3k in Supporting Information S1), likely resulting in an increase in dust from the Central PSA. Our modeling results agreed with remote sensing products and indicated that approximately 48% of dust collected on 29 February 2016 was from the Central PSA, which suggests that MixSIAR can accurately discern PSAs. To further validate our results, we performed two sensitivity analyses using MixSIAR to demonstrate our results were not biased by the use of remote sensing (test 1; see Figures S4–S6 in Supporting Information S1) nor by particle grain size (test 2; see Figures S7–S9 in Supporting Information S1).

Looking first at the results in 2016, Figure 3a shows that the Central PSA dominated transport to SA in winter and spring on most days. The seasonal median of dust from the Central PSA was 44% (Figure 3b), a result that is in line with previous studies that suggest Central PSA dust sources active during winter contribute to dust deposition in the Amazon (Koren et al., 2006; Washington & Todd, 2005; Washington et al., 2006). We also include pie plots of our model results in Sr-Nd space (a scatter-pie plot) in Figure S10 in Supporting Information S1 to enable better visualization of the model results. Wintertime dust emission from the Central PSA is due to a confluence of synoptic-scale meteorological conditions including intensification of anticyclonic systems that occur over the North African mid-latitudes, fueling stronger, drier, and colder northeasterlies (Engelstaedter et al., 2006; Knippertz & Todd, 2012; Stuut et al., 2005). Simultaneously, the ITCZ begins to shift northward, which brings winds from the southeast that meet the northeasterlies, enhancing surface convergence and gustiness (Engelstaedter & Washington, 2007). The strongest anticyclone over North Africa during this season is the Libyan High (Knippertz & Todd, 2012), located north of the Bodélé Depression, resulting in strong dust emissions from this region. Previous remote sensing work has suggested that dust from the Central PSA was the major contributor to dust to SA (Koren et al., 2006), and our geochemical results from 2016 support this, as does our detection of species of freshwater diatoms from African paleolakes (e.g., *Aulocoseria*) in winter and spring of 2016 at Cayenne (Barkley et al., 2021; Skonieczny et al., 2011).

In contrast, the majority of long-range transported dust collected in Cayenne in 2014 originated from the Western-PSA3. For example, on 29 January 2014, and for all samples until 8 April 2014, the Western-PSA3 contributed 54% to our samples (Figure 3a). During winter 2014, more dust originated from the Western-PSA3 on average (53%) than the Central PSA (23%). This trend continues into spring 2014 (Figure 3b), when the Western-PSA3 again provides more dust (48%) to Cayenne than the Central PSA (23%). Our results suggest that the Western-PSA3 is also an important wintertime dust source transported to the Amazon Basin in agreement with other studies (Kumar et al., 2018; Skonieczny et al., 2013). Generally, dust emission from the Western-PSA3 region in boreal winter is less well understood but is again likely related to anticyclonic activity over North Africa. This

activity provides a strong pressure-gradient differential between northern and southern North Africa, fueling the convergence that drives dust transport from the Western PSA (Engelstaedter et al., 2006; Evan et al., 2016; Knippertz et al., 2011).

Our model results shown in this work do not support the Western-PSA2 as a consistent source of dust to SA. Results in Figure 3a show enhanced transport from the Western-PSA2 is episodic in nature, with three periods of time where Western-PSA2 contributes 20%–30% of dust (early February 2014, late March 2014, and summer 2016). Early flight studies and meteorological analysis suggest that dust from the coastal region of the Western-PSA2 is lofted into the atmosphere but stays constrained below the inversion layer in the MBL and, therefore, would not be expected to undergo extensive long-range transport (Carlson & Prospero, 1972; Prospero et al., 2020, 2021).

3.3. Possible Role of North African Climate in Controlling the Source of Transported Wintertime Dust

We suggest that the strength of the Libyan High over North Africa and its associated zonal winds affect dust emission while precipitation results in losses of dust during transport, modulating the relative importance of each dust PSA transported to the Amazon.

In 2016, the Libyan High was stronger than in 2014 (Figures S11 and S12 in Supporting Information S1), causing stronger northeasterlies to pass over the Central PSA. This is evidenced by Figure 4a, which, using NCEP/NCAR reanalysis monthly composites, shows that the zonal winds at 925 hPa were up to 2 m s^{-1} stronger in 2016 than in 2014 over the southern part of the Libyan High, represented by the warmer colors below the "L." Additionally, the Trade Winds were up to 5 m s^{-1} stronger in 2016 across the eastern TAO. The stronger winds over Africa and the TAO could have more efficiently transported dust across these regions. While there is evidence of intra-seasonal variability in the amount of dust transport that has been linked to the strength of the Libyan High (Washington & Todd, 2005), we suggest that the variation in the strength of the Libyan High can also modulate the relative amount of dust transport from different PSAs to the Amazon.

Precipitation has been suggested to dominate dust deposition across the TAO, particularly from the Central PSA (Goudie & Middleton, 2001; van der Does et al., 2020). Using the Tropical Rainfall Measuring Mission product 34B2, Figure 4b shows the difference in precipitation rate for January to April in 2014 and January to April in 2016 with warmer colors indicating more precipitation in 2016 than in 2014. The plot shows that there was more precipitation in the Gulf of Guinea and TAO in 2014, which we suggest caused enhanced wet deposition of dust from the Central PSA in 2014 compared to 2016. This conclusion is in agreement with a recent study in which the authors used a trajectory model to show that a majority of the dust from the Central PSA was deposited over the Gulf of Guinea and TAO as a result of their location underneath or within precipitating clouds (Y. Yu et al., 2020). An analysis of individual days also confirms our hypothesis; these results are shown in Figures S13–S18 in Supporting Information S1.

Our finding that there is interannual variability in the source of dust transported to Cayenne agrees with a recent modeling study showing Cayenne is located near (within ∼1°) a transition zone whereby both Western and Central African dust sources contribute equally to dust deposition in the Amazon (Kok et al., 2021). Model runs in Kok et al. show that the Central PSA (which in their study includes the Sahel region) can contribute between 16% and 53% of dust deposition to the Amazon while the Western PSA (which in their study combined the Western-PSA2 and Western-PSA3) can contribute between 28% and 53% of the annual deposition flux. The spread in their results, which are the average of 4-year runs, may be due to the interannual variability of dust transport from each PSA.

4. Implications

Previous work has suggested that either the Bodélé Depression (Ben-Ami et al., 2010; Koren et al., 2006) or parts of the Western North African PSAs (Y. Yu et al., 2020) dominate long-range dust transport to SA and the TAO. Our results, which are based on radiogenic Sr and Nd isotopes combined with a mixing model, reveal a more nuanced picture. We proport that interannual variability in the transported dust PSA(s) is controlled by the prevailing meteorology. Additionally, differences in our results and those reported at ATTO point to gradients in the dust PSAs transported to different regions of the Amazon Basin during different seasons. Because the

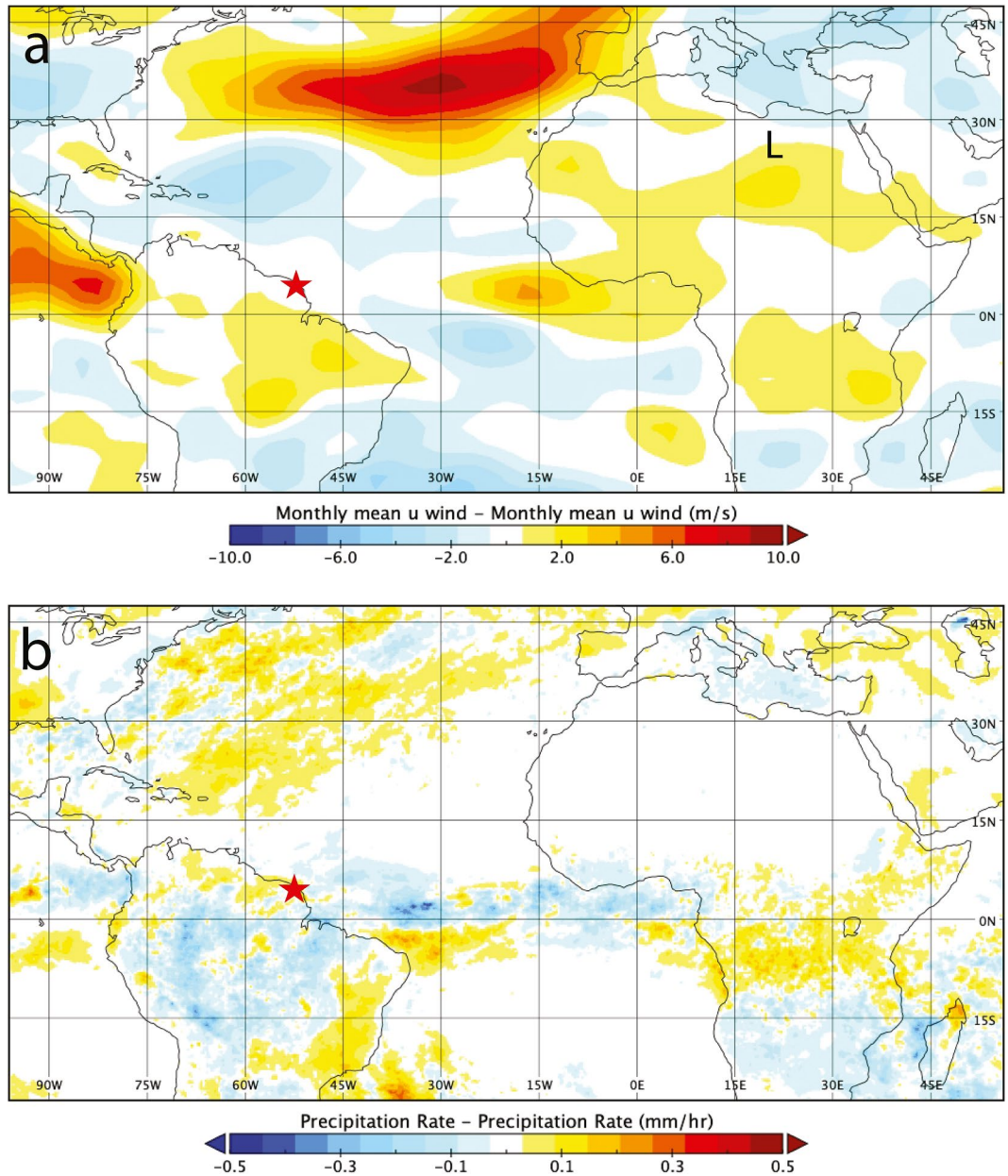

Figure 4. The difference in 925 hPa zonal wind between February 2014 and February 2016 is shown in panel a. The warmer colors indicated stronger easterly winds in 2016. The "L" denotes the location of the Libyan High in 2016. The difference in precipitation from January to April 2016 and January to April 2014 is shown in panel b. The warmer colors indicate more precipitation in 2016. The star shows the location of our sampling site in Cayenne.

atmospheric deposition of nutrients associated with dust is thought to control primary productivity in the nutrient-limited TAO and Amazon (Jickells & Moore, 2015; Okin et al., 2004), interannual variability in the dominant PSA can, in turn, modulate nutrient deposition and the biogeochemical outcome in these ecosystems. We note that more isotopic analysis from each PSA—particularly from Western-PSA3—is needed to extend the results of this work as well as fingerprinting using other isotopes such as lead (Pb) that have shown promise in distinguishing between different PSAs (Scheuvens et al., 2013).

North African dust contains iron (Fe) concentrations that are approximately uniform throughout the region (Scheuvens et al., 2013). Soil from the Bodélé Depression contains nanosized Fe oxides including magnetite that might enhance Fe solubility; however, it is unclear if soil from the Western-PSA2 and Western-PSA3 contain

these soluble forms (Moskowitz et al., 2016). Phosphorus (P) concentrations are less uniform across North Africa and the Western-PSA2 has some magmatic P "hotspots" with enhanced P solubility (Gross et al., 2015; Scheuvens et al., 2013). The Bodélé contains elevated levels of P from fishbones or scales that are more bioavailable than P from apatite, the most common P-containing mineral in North African dust (Hudson-Edwards et al., 2014). Therefore, when combined with our results, the available data suggest that there may be an enhancement in the solubility of Fe transported to the TAO and the magnitude and solubility of P transported to the Amazon in years when transport from the Central PSA dominates but more elemental and solubility analyses of Western PSA soils are needed.

Climate change is poised to affect global precipitation, wind, and atmospheric circulation patterns. There is high confidence that precipitation will increase over North Africa (Arias et al., 2021) with the largest increases projected over parts of the Central PSA and Western-PSA3 (Figure S19 in Supporting Information S1), potentially reducing dust emission from these PSAs by increasing soil moisture and vegetation. This may result in a change in the amount as well as the dominant dust PSA transported to the Amazon. Given the present-day connections between meteorological conditions and dust (Prospero & Lamb, 2003), isotopic fingerprinting of paleodust samples combined with MixSIAR could provide a window into past and future changes in meteorological patterns as well as dust emissions under a changing climate.

Data Availability Statement

Isotopic data and code for MixSIAR presented in this work can be found at the University of Miami data repository: https://doi.org/10.17604/qqq0-p310. Dust mass concentrations from Cayenne in 2014 and 2016 can be found in Prospero et al. (2020) and Barkley et al. (2019), respectively. Data used to make Figures 4a and 4b can be found at https://psl.noaa.gov/data/gridded/data.ncep.reanalysis.html and https://giovanni.gsfc.nasa.gov/giovanni/ respectively.

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