## Answers to Reviewers

Manuscript: Aerosol Atmospheric Rivers: Climatology, Event Characteristics, and Detection Algorithm Sensitivities by Sudip Chakraborty et al.

We are thankful to the reviewers for their insightful comments and help improving the manuscript. Our responses are below, comments by reviewers are italicized.

## Reviewer #2 (Comments to Author (shown to authors):

Thank you very much for your suggestions and the insightful comments. We have made changes to the manuscript following your suggestions. The changes are highlighted in yellow in the manuscript.

This paper is an extension of authors' previous work on the detection of aerosol atmospheric rivers (AARs) with the MERRA-2 aerosol reanalysis. The study focuses on the characteristics of AARs derived from 18-years (1997-2014) of data record. The paper is generally well written, but some clarifications and elaborations are needed.

## General comments:

Numerous studies have used satellite remote sensing, ground-based networks, and global chemical transport models to characterize aerosol long-range transport across continents. How is this study related to previous studies? What new value will this study add? How can people use the generated datasets to advance sciences? These points need to be elaborated in the paper.

## Thank you for this valuable suggestion. We have added

"In addition, in-situ measurements have been conducted to detect aerosol transport events over various regions of the world, even in the remote polar regions (Gohm et al., 2009a; Tomasi et al., 2007a; Wang et al., 2011a; Rajeev et al., 2000a; Bertschi et al., 2004a; Qin et al., 2016a; Ackermann et al., 1995; Fast et al., 2014a). Many studies have previously investigated the longrange aerosol transport events between various regions of the world (Prospero, 1999; Sciare et al., 2008; Abdalmogith and Harrison, 2005; Swap et al., 1996; Kindap et al., 2006a; Weinzierl et al., 2017) including inter-continental transport events. Many regions have been studied including the transport events from the Sahara Desert to the United States (Prospero, 1999), Europe to Istanbul (Kindap et al., 2006b), East Asia to California (Fan et al., 2014), and South Africa to the South Atlantic region (Swap et al., 1996). Many other studies have investigated aerosol aging and chemical processes during the transport events (Febo et al., 2010; Kim et al., 2009; Mori et al., 2003; Markowicz et al., 2016; Song and Carmichael, 1999) including the secondary organic aerosol formation and depicted the impact of the long-range aerosol transport on clouds (Wang et al., 2020a; Garrett and Hobbs, 1995), precipitation (Fan et al., 2014), radiation (Ramanathan et al., 2007), and air quality events including the PM2.5 level (Han et al., 2015; Chen et al., 2014; Prospero et al., 2001; Febo et al., 2010). Apart from the studies using satellite and in-situ measurements, climate models have often been used to understand aerosol transport (Takemura

et al., 2002; Chen et al., 2014; Ackermann et al., 1995; Fast et al., 2014). As mentioned above, many of these studies used different approaches and methodologies and thus comparing one region to the another around the globe or depicting one species' character of extreme events to another species with a common framework is difficult. Although these studies identify aerosol transport events across the globe, a clear picture about the identification of the extreme aerosol transport events (see methods) using long-term climatological observational data sets, their climatology and major transport pathways, and fractional contribution of those extreme transport events to the global annual transport were lacking.

Leveraging the concept of atmospheric rivers (ARs) (Ralph et al., 2020; Zhu and Newell, 1994) and a widely used global AR detection algorithm (Guan and Waliser, 2019, 2015; Guan et al., 2018), our previous study developed an aerosol atmospheric rivers (AARs) detection algorithm (Chakraborty et al., 2021a). As with ARs that were studied around the globe using different algorithms in different places, including global change studies, it was hard to get a consistent assessment based on one homogeneous method of identifying the transport events. A value of this study comes from the extension of a well-developed algorithm and applied uniformly around the globe and across species."

Also, we have added the following in the conclusion section:

"To our knowledge, this study significantly advances the understanding of global aerosol transport events. This study is the first to provide a common detection and analytical basis for studying extreme transport events, heavily leveraging the heritage and methodologies by the (water vapor) atmospheric river community. Moreover, this study identifies the extreme transport events by selecting grid cells with IAT values greater than the 85<sup>th</sup> percentile of their climatological values and retaining the stronger 50% of those objects detected based on the object-mean IAT values. This study creates a database of the AAR events between 1997-2014 that will be expanded till 2020 and will provide a valuable platform for aerosol transport-related research including their impacts on the climate and air quality. Furthermore, the algorithm developed to detect AARs can be used to detect the real-time AAR events using the nature run of the GOES FP system that provides analyses and forecasts produced in real-time, using the most recent validated GEOS system

(https://gmao.gsfc.nasa.gov/GMAO products/NRT products.php).

Discussion of potential uncertainties associated with the analysis: MERRA-2 assimilates satellite observed aerosol optical depth (clear-sky radiance associated with it). This offers strong constraint to the AOD in MERRA-2. However, aerosol components (e.g., sulfate, dust, sea-salt, and carbonaceous aerosol) are not fully constrained. How will this lack of constraint in the aerosol components affect your detection and quantification of AARs? MERRA-2 aerosol vertical distribution is also not constrained by satellite-based lidar observations, which constitutes of another uncertainty. You could use CALIPSO lidar observations to evaluate the MERRA-2 aerosol extinction profiles in major AAR regions (e.g., those defined in Figure 5).

We agree with the reviewer. There is an attempt to use CALIPSO and HSRL examine the vertical structure of aerosols in MERRA-2; see the Buchard et al. 2017 paper. Regarding speciation: single channel AOD data has no information about speciation. The model, informed

by specified emissions, comes up with this speciation. We acknowledge this limitation, there is no good way to quantify it, except perhaps by using lidar typing as in the paper below. They found that "despite having the column AOT constrained by MODIS, comparison to the CALIOP VFM reveals a greater occurrence of dusty aerosol layers in our MERRAero-CALIOP VFM due to errors in MERRAero aerosol speciation."

Nowottnick, E. P., P. R. Colarco, E. J. Welton, and A. da Silva, 2015: Use of the CALIOP vertical feature mask for evaluating global aerosol models. Atmospheric Measurement Techniques Discussions, 8, 1401–1455, <u>https://doi.org/10.5194/amtd-8-1401-2015</u>.

As a result, owing to the importance of the fact that MERRA-2 aerosol vertical distribution is also not constrained by satellite-based lidar observations, we are currently investigating the CALIPSO profiles as a part of a new analysis. We have added:

"It is important to keep in mind that the aerosol vertical structure in MERRA-2 is not directly constrained by measurements, and are chiefly determined by the injection height of the emissions as well as turbulent and convective transport processes parameterized in the model. Evaluation of the vertical structure of MERRA-2 aerosols appears in Buchard et al. (2017). Buchard et al. (2017) has attempted to use Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) to examine the vertical structure of aerosols in MERRA-2. In section 4.3

We acknowledge in the conclusion section that

"We acknowledge a source of uncertainty in the aerosol mass mixing ratio and IAT profiles shown in this study from the MERRA-2 data since MERRA-2 aerosol vertical distribution is also not constrained by satellite-based lidar observations. A recent study found that comparison to the CALIOPO VFM data detects a greater occurrence of DU aerosols in MERRA-2 due to errors in MERRAero aerosol speciation (Nowottnick et al., 2015). In a future study, an in-depth analysis will be performed using the observation from the ground-based and air borne measurements (wherever available) and CALIPSO. For example, ORACLES mission and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation can be used to to study the subsidence that depresses the plumes once they reach the Atlantic Ocean off the west African Coast (Das et al., 2017)."

We acknowledge the importance of the question and it will be addressed in a future study We show one example here from our current analysis here,



**Figure.** (A) Mean aerosol mass mixing ratio for all the dust AARs observed between 1997-2014 over the locations marked in the map shown in the inset. (B) DU AARs detected and CALIPSO overpass in the daytime on 25<sup>th</sup> June, 2008. (C) CALIPSO vertical feature mask over the region where a DU AAR (encircled in green) is detected to transport aerosols from the Sahara Desert to the Caribbean region. Color bar represents: **1: Clear air, 2: cloud, 3: tropospheric aerosols, 4: stratospheric aerosols, 5: surface, 6: subsurface, 7 attenuated, and L: low confidence**. The aerosol layer is encircled in black.

Figure A shows the elevated DU aerosol mixing ratio for DU over the Sahara to Caribbean pathway (orange). (B) shows the AARs detected on 25<sup>th</sup> June 2008 and the CALIPSO track on the same day overpassing a DU AAR. And (C) shows the CALIPSO vertical feature mask overpass on the same day observing the cross section of the DU AAR (Fig. B; circled) travelling from the Sahara Desert to the Caribbean region/Southern US/ North of the South America. From (C), it is clear that CALIPSO can see that the aerosol layers are elevated up to 6 km between 12–23° N (exactly over the AAR) and is consistent with the DU profile (orange color, The Sahara Desert to the Caribbean pathway) in (A) or Fig. 5B in the manuscript. It is to be noted that CALIPSO can also detect aerosols outside the AAR boundary since AAR detection is restrained by the IAT values, not the aerosol mass that is not being extremely transported (IAT values < 85<sup>th</sup> percentile). We acknowledged the importance of the question and we have added the importance of this in the conclusions section and it will be addressed in a future study.

An 18-years data record of AARs has been generated. But this study only analyzed climatology and seasonal variations of AARs. I strongly suggest that the authors analyze and present interannual variations of AARs in the paper.

We agree with the reviewer and thanks for the great suggestion. This manuscript is intended to provide more details regarding the algorithm development and sensitivities so that the database can be released and used by others, and subsequently we, and others, can undertake this sort of variability studies. We are working on the influence of the events like ENSO, MJO, emissions etc on the variations in AARs' occurrences, interannual variations, and also the trends. That warrants a separate manuscript and will be provided once we extend the data up to 2020. We are currently extending the datasets so that we have a longer period of time for the trend analysis.

Specific comments:

In the abstract: aerosol components DU, CA, SU, and SS are used but not defined.

Thanks for pointing this out.

Lines 56-58: not sure about what you mean here.

We have clarified in lines 56-62:

"Aerosols can influence a plant's health in two ways. Firstly, aerosols are known to increase the amount of diffuse radiation (Xi & Sokolik, 2012). The implications for plants are that along with decreasing the direct beam photosynthetically active radiation (PAR), the presence of aerosols would lead to greater diffused PAR, which means illumination of a greater portion of plant canopies, including shaded leaves, for which direct PAR was not accessible previously (Gu et al., 2003; Knohl & Baldocchi, 2008; Niyogi et al., 2004)."

Lines 61-64: there have been numerous studies of aerosol intercontinental transport, based on satellites, global models, and even ground-based networks. What is a relationship between your study and these previous studies?

Thank you for pointing this out, and we agree with your assessment. We have provided a more complete literature review in the introduction to connect our work with previous work. Please see the introduction section and lines 590-602 of the conclusion section.

Line 70: here you define CA (carbonaceous aerosol) as a sum of organic and black carbon. Later when discussing CA AARs, you present organic carbon (OC) and black carbon (BC) separately. To be consistent, it is better to combine OC and BC.

We have combined OC and BC in Figures 1 and 2 that show the mean IAT and emission of different species of aerosols to remove redundancy. They have almost similar geographical distributions in terms of sources and IAT value around the globe. In other figures we have separated OC and BC particularly because of the significance of the impact of BC on the

radiative forcing. We will provide BC AARs in the data base separately for future studies on the impact of BC AARs on the radiative forcing. Also, OC rivers generally carry a higher mass of aerosols than BC rivers. We have mentioned that in lines 312-314.

"Herein forth, we separately show the CA rivers as OC and BC rivers, especially because BC AARs can significantly impact the radiative forcing compared to OC AARs and the differences in the mean mass of aerosols being transported by the OC and BC AARs (Fig. 4). "

Lines 94-95: no need to define DU, SU, SS and CA again. They are already defined in lines 69-70.

Thanks for pointing this out.

Lines 103-104: Given it assimilates satellite observations of aerosol optical depth, MERRA-2 should be able to "capture the global aerosol optical depth reasonably well". This is obvious.

While aerosol data is only assimilated twice daily under clear sky conditions, the MERRA-2 aerosol reanalysis data capture the global aerosol optical depth reasonably well and are well-validated by many studies using observation.

Figure 2: what do you mean SU emission here? Do you mean emitted sulfate or precursor gases (e.g., SO2 and DMS)? Direct emissions of sulfate are very small. Then what is unit for the sulfur emission? What is unit for carbonaceous aerosol (CA) emission?

Yes, we have used SO<sub>2</sub>. We have modified the text accordingly from SU to SO<sub>2</sub>. The units (kg m<sup>2</sup> s<sup>-1</sup>) are provided in the figure title and the caption. Thanks for pointing this out.

Line 217: can "boreal forests" be referred to as "rainforests"?

We have corrected as "Globally, boreal and rainforests are the most significant contributors to the CA aerosols (Fig. 2D). "Please see lines 268 and 625.

Lines 252-254: But if satellite observations (e.g., AOD) captured the ash plume, the ash signal would be evident in the MERRA-2 product to some extent.

This is correct. However, since ash is not explicitly included as a species in MERRA-2, the correction implied by the ash signal in AOD would be erroneously ascribed to another species, and without having the correct vertical structure, subsequent transport would likely be wrong as well. Successfully, the algorithm detects many SU AAR over there.

Lines 259-260: "in terms of AOD" - do you really need these four words?

Deleted.

Line 288: are you sure that MERRA-2 don't account for biogenic sources like Carbonyl Sulfide and Dimethyl Sulfate? I cannot believe that an aerosol model dealing with sulfur cycle doesn't account for DMS.

We have rectified the sentence. MERRA-2 includes sources and transports Dimethyl Sulfate (DMS), with mechanisms for creating sulfate aerosols from DMS. While OCS is included in the current development version of the GEOS model, it was not available in the earlier version of the model used in MERRA-2. Please see the modified sentence in line 349.

Lines 318-319 (and Figure 5A): There is a secondary peak at about 960 mb, which should be discussed. I assumed that the two peaks are associated with seasonal variations of trans-Atlantic dust transport.

Thank you very much for pointing this out. This warrants further study on the roles of different jet streams on the IAT profiles- their seasonal variabilities. We hope to address that in another study.

However, we have added in lines 385-395: "There is also a secondary peak at the 960 hPa (Fig. 5A). The aerosol mixing ratio (Fig. 5B) slightly decreases from the surface up to a height of 960 hPa and then increases above it. This might suggest the influence of the gravitational force and settlement on the aerosol mixing ratio as observed in many of the aerosol mixing ratio profiles of other species over other regions (for example, the red line showing the aerosol mixing ratio profile for the Sahara-European pathway). However, a smaller peak in the wind profile (Fig. 5C) around 950 hPa indicates the influences from the low-level jets and the seasonal variations of trans-Atlantic dust transport that have not been studied yet. A detailed seasonal analysis is required to understand the existence of the smaller peak at 960 hPa. However, the presence of the African easterly jet-north or AEJ-N above, centering 600-700 hPa, further lifts the aerosols and attain a peak around 750 hPa."

*Figure 5: why do you use aerosol mixing ratio? It is more suitable to show mass concentration?* 

To be consistent with the classic AR study where we use specific humidity as the moisture variable, we use mixing ratio here as well.

Line 379: "compoment" should be "component", right?

Corrected.

*Lines* 517-518: *if CA AARs are associated with burning of boreal forests, can we still call them "rainforests"*?

Thanks for pointing this mistake. We have added boreal and rain forests in lines 268 and 625.

*Lines 519-520: How can the presence of SU AARs all year around in the northern hemisphere be explained by the biomass burning and biogenic activities?* 

We have removed that speculation from the sentence.