### Response to Reviewer #2:

In the present manuscript, Guangyao Dai and coauthors track a Saharan dust plume across the Atlantic Ocean and calculate the dust horizontal fluxes. The novel approach in their manuscript is the combination of two satellites (CALIPSO and Aeolus) measuring at different wavelengths. To bridge the gap between the overpasses of the two satellites ERA5 model reanalysis and HYSPLIT trajectories are used. However, the satellite data are not treated in a correct manner with the result that the whole proposed method is not valid. Therefore, I have to reject the manuscript.

AR: Thanks. The train of thought of this work is using CALIPSO and Aeolus aerosol optical properties to capture and describe a long-range Sahara dust transportation event which occurred from 15 June 2020 to 27 June 2020. ERA5 model reanalysis wind field data and HYSPLIT trajectories are used as tools to verify the whole transportation. Finally, dust mass concentration derived from <u>five</u> aerosol optical properties, which are **backscatter coefficients, extinction coefficients at 532nm,1064nm from CALIPSO** and <u>extinction coefficient at 355nm</u> from Aeolus, combined with ERA5 relative humidity, wind field data which assimilate Aeolus L2B HLOS wind data to implement calculation of dust advection, which is defined as the multiplication of dust mass concentration wind velocity.

However, it is surely a challenge of this work **based on present technology** because the time and distance gaps of CALIPSO and Aeolus overpasses, and the developing Aeolus L2A product which needs new algorithm (Flament et al., 2021). More improvements and efforts need to be implemented to acquire more precise analysis and calculation. Nevertheless, in this work, based on present technology, we utilize present data and model as carefully as possible. Therefore, we think this work is acceptable and reasonable.

Hence, after the comprehensive consideration, the treatment for the satellites' data and the proposed method are improved and updated in section 3 "Methodology" of the revised manuscript. The corresponding corrections are presented as following:

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## 3. Methodology

In the study of dust transport and advection measurement, as shown in Figure 1, the dust identification, Aeolus and CALIPSO tracks match, data analysis and the HYSPLIT model analysis are necessary and the schematic flowchart is described briefly.

## 3.1 Method used to match CALIPSO and Aeolus data

To identify the dust events and to choose the quasi-synchronization observations with ALADIN and CALIOP, the flowchart is presented in this figure. To preliminarily determine the occurrences of dust events, the "Dust score index" data provided by AIRS/Aqua are used to determine the dust plume coverage and transport route. With this given information, the VFM products from the simultaneously observations with the spaceborne lidar CALIOP are applied to cross-check the identification of dust events. Hence the vertical distributions of dust plumes are presented. To find the original sources and to predict the transport routes of dust plumes, the backward trajectory and forward trajectory is used respectively. When the dust events are determined, the simultaneous observations with ALADIN and CALIOP have to be selected. As the dust plumes can be captured by CALIPSO VFM products, hence, starting from the CALIOP observations, the nearest Aeolus footprints could be figured out. Since the orbits of Aeolus and CALIPSO are different, they cannot meet each other at the exactly same time and same location. From our study, the closest CALIPSO scanning tracks to those of Aeolus, are about 4 hours ahead of Aeolus. Based on the transport directions of dust events modelled with HYSPLIT, the tracks of Aeolus should be always downwind of the tracks of CALIPSO. When the tracks of Aeolus and CALIPSO are selected, the distances between the tracks can be calculated. Assuming the wind speed scale between CALIPSO scanning tracks and Aeolus scanning tracks is 5  $\text{m} \cdot \text{s}^{-1}$  to 15  $\text{m} \cdot \text{s}^{-1}$ , the transport distance scale of the dust plumes is 72km to 216km. Besides, during the short-time transportation of Sahara dust plume, dust optical properties maintain almost unchanged (Haarig et al., 2017). Consequently, in our study, if the distances between two satellites scanning tracks are less than 200 km and the tracks of Aeolus are downwind of the tracks of CALIPSO, it is reasonable to state that the dust plumes captured by CALIPSO are transported towards the Aeolus scanning regions in around 4 hours, hence the following procedures could be continued.

### 3.2 Datasets and quality control

In this study, the extinction coefficient at 355 nm from ALADIN, at 532 nm and 1064 nm from CALIOP are collected as the useful dataset. The extinction coefficients at 355 nm correspond to the "Aeolus Level 2A Product" retrieved by SCA (standard correct algorithm). In this study, we choose SCA instead of ICA (iterative correct algorithm) because the extinction coefficients from ICA are noisy and the assumption of "one

single particle layer filling the entire range bin" in SCA is reasonable and is met in the situation of the heavy dust events observation. Additionally, we use the mid bin product (sca optical properties mid bins) of SCA instead of the normal product of SCA, because the mid-bin algorithm provides more robust results (Baars et al., 2021; Flament et al., 2021). The extinction coefficients, which are more sensitive to noise and are the significant inputs of the dust advection calculation, are better retrieved through this "mid bin" averaged version of the algorithm. In terms of quality control, negative extinction coefficient values of L2A are excluded while the "bin 1 clear" flag and the "processing qc flag" of L2A are used to eliminate invalid data. The backscatter coefficients and extinction coefficients at 532 nm and 1064 nm are the "Total Backscatter Coefficient 532", "Extinction Coefficient 532", "Backscatter Coefficient 1064" and "Extinction Coefficient 1064". Moreover, "Extinction QC Flag 532" and "Extinction QC Flag 532" from CALIPSO Level 2 products are used to conduct quality control of CALIPSO data. Since the footprints of Aeolus and CALIPSO are not exactly matched, the missing wind data between their tracks have to be filled in using the ERA5 wind field data. There are two purposes on the usage the ERA5 wind field data between Aeolus and CALIPSO tracks. One is that the ERA5 wind speed and direction data provides the evidence of dust transporting from CALIPSO tracks towards Aeolus tracks. Besides, the ERA5 wind field data between the tracks of Aeolus and CALIPSO at all height surfaces are smoothly distributed and the values are stable. It means that the Aeolus L2C data can be used at the location of the CALIPSO track.

## 3.3 Dust advection calculation

In Figure 2, the flowchart of dust mass advection calculation procedure is provided. Based on the dataset consists of the backscatter coefficients and extinction coefficients at the wavelengths of 1064 nm and 532 nm from CALIOP and the extinction coefficients at the wavelength of 355 nm from ALADIN, the aerosol volume concentration distribution can be calculated based on regularization method which was performed by generalized cross-validation (GCV) from Müller et al. (1999).

The advantage of this method is that it does not require prior knowledge of the shape of the particle size distribution and the estimate uncertainty of aerosol volume concentration is on the order of 50% if the estimated errors of the input is on the order of 20%. For the accuracy of the CALIPSO-retrieved extinction and backscatter coefficients: for the backscatter coefficient at 532 nm, during the daytime, the average difference between collocated CALIPSO and HSRL measurements is  $1.0\%\pm3.5\%$  in V4 (Getzewich et al., 2018); for the backscatter coefficient at 1064 nm, the CALIOP V4 1064 nm calibration coefficients are accurate to within 3 % (Vaughan et al., 2019); for the extinction coefficients, the uncertainty in the V4 dust lidar ratio of 20 % (30 %) at 532 nm (1064 nm) (Kim et al., 2018), thus it is considered that the estimate errors of the extinction coefficients from CALIPSO are on the order of 20%. Consequently, we think that the uncertainties of CALIPSO-retrieved extinction and backscatter coefficients are on the order of 20%. According to Flament et al. (2021), because of the lack of cross-polarized light, 355nm backscatter coefficients of Aeolus are underestimated, especially for dust aerosol. Nevertheless, the extinction is not affected. In this work, Aeolus-retrieved backscatter coefficients at 355nm are not applied for the calculations of the dust volume concentration distribution and mass concentration. For the accuracy of the Aeolus-retrieved extinction coefficient, the simulation extinction coefficients fit the inputs well mostly, especially when the altitude is larger than 2km (Flament et al, 2021). Hence, we think that after rigorous quality control, Aeolus L2A extinction coefficient could be the input parameters of the regularization method. In conclusion, we think that the estimated errors of the five input parameters we used to calculate the aerosol volume concentration are on the order of 20%. The estimate errors of dust advection are the combination of mass concentration estimate errors (~50%) and Aeolus L2C wind vector estimate errors.

It should be emphasized that due to the different vertical resolution and horizontal resolution between Aeolus data and CALIPSO data, a common pixel grid should be conducted before calculation. For vertical resolution, 23 data bins of Aeolus L2A mid bin optical property products are interpolated to 399 data bins of CALIPSO according to the altitude information of two products. For horizontal resolution, both Aeolus and CALIPSO products are averaged along every integer latitude to acquire a common horizontal pixel grid. After integrating and multiplying an assuming typical dust particle

density which is set as 2.65 g·cm<sup>-3</sup> referring to previous studies (e.g., Schepanski et

al., 2009; Hofer et al., 2017; Mamouri and Ansmann, 2017), the particle mass concentration would be estimated as Engelmann et al. (2008) introduced. For ECMWF wind field data, wind speed data, wind direction data and RH data between Aeolus and CALIPSO scanning tracks are averaged along longitude and averaged along every integer latitude, while, vertically, they are interpolated to CALIPSO data bins to match the common pixel grid. Besides, when the RH is larger than 90%, the dust aerosol will be influenced by the hygroscopicity effect and its properties could change. Then the mass concentration calculation method does not make sense any more (Engelmann et al., 2008). For the cloud screening, aside the RH data, we use Level 2 5km aerosol profile of CALIPSO, which only provide aerosol optical properties so the cloud can be screened. Therefore, relative humidity data provided by ECMWF is used to filtrate unavailable data. Ultimately, combining with the particle mass concentration and the horizontal wind speed provided by Aeolus and ECMWF, the dust mass advection is defined as Eq. (1), to represent the transportation of dust aerosol quantificationally.

Advection<sub>aerosol-mass</sub> = 
$$m \cdot v$$
,

where *m* is the aerosol mass concentration and  $\vec{v}$  is the horizontal wind velocity.

(1)



Figure 1. Dust identification, Aeolus and CALIPSO tracks match and data procedures.





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# <u>Reference:</u>

Flament, T., Trapon, D., Lacour, A., Dabas, A., Ehlers, F., and Huber, D.: Aeolus L2A Aerosol Optical Properties Product: Standard Correct Algorithm and Mie Correct Algorithm, Atmos. Meas. Tech. Discuss. [preprint], https://doi.org/10.5194/amt-2021-181, in review, 2021.

The following points underline my decision and may help the authors to improve their work:

Aeolus is providing the circular co-polarized component of the backscatter and not the total backscatter coefficient. The missing cross-polarized component is not negligible

in dust cases as used in the manuscript. You are missing a significant part of the backscatter coefficient at 355 nm.

### AR: Thank you for your suggestion.

As reported in Flament et al. (2021), it is clearly stated that "Designed as a wind lidar, ALADIN does not have the ability to measure depolarization. The UV laser beam is linearly polarized, and analyzed only along the parallel direction. Any cross-polarized light is rejected. This means that, in order to compare Aeolus observations to other instruments, only the co-polar component must be considered. Not going through this extra step before comparing would make it seem that the total backscatter of highly depolarizing targets such as ice crystals or dust is largely underestimated by Aeolus. **Because the extinction is not affected,** the corresponding Aeolus lidar ratio is going to be larger than the total lidar ratio.". It can be concluded that because of the missing cross-polarized component, Aeolus backscatter coefficient at 355nm is underestimated, especially for dust aerosol. **Nevertheless, it has to be emphasized that the extinction coefficient at 355nm during the calculation of volume concentration and mass concentration.** 

As described in the section 3 of the revised manuscript, we only use five aerosol optical properties to estimate dust volume concentration and mass concentration, which are backscatter coefficients and extinction coefficients at 532nm and 1064nm from CALIPSO and extinction coefficient at 355nm from Aeolus. In our study, Aeolus-retrieved backscatter coefficients at 355nm are not applied for the calculations of the dust volume concentration distribution and mass concentration. Thus, we insist that the usage of Aeolus-retrieved extinction coefficient in calculating the dust volume concentration and mass concentration is reasonable.

Besides, Aeolus can provide valuable information thanks to its HSRL design. Flament et al. (2021) also proves that Aeolus has the ability to capture dust aerosol layers, which are from the same dust transportation event as this work (as shown below).



<u>Reference:</u>

Flament, T., Trapon, D., Lacour, A., Dabas, A., Ehlers, F., and Huber, D.: Aeolus L2A Aerosol Optical Properties Product: Standard Correct Algorithm and Mie Correct Algorithm, Atmos. Meas. Tech. Discuss. [preprint], https://doi.org/10.5194/amt-2021-181, in review, 2021.

CALIPSO measures the backscatter coefficient at 532 and 1064 nm, but not the extinction. The extinction provided by CALIPSO is retrieved by multiplying the backscatter coefficient with the aerosol-type-dependent lidar ratio. Therefore, the extinction coefficient is not an independent quantity. For your inversion calculation, you need independent measurements of the extinction coefficient, either by high spectral resolution (HSRL) or Raman lidar measurements.

AR: Thank you for your suggestion. Because of the detection principle, CALIPSO can only derive 532nm and 1064nm backscatter coefficient directly. The extinction retrieval of CALIPSO definitely needs more complex and aerosol-type-dependent algorithms. After the launch of CALIPSO, the retrieval algorithm of extinction had been established and developed, which was named as Hybrid Extinction Retrieval Algorithms (HERA) (Young et al., 2009). Uncertainty and error sensitivity analyses of this HERA algorithm were implemented to evaluate the propagation of uncertainty errors and bias errors (Young et al., 2013). To further evaluate the errors of extinction and improve the extinction products quality, a large amount of validation campaigns and experiments are implemented (Kacenelenbogen et al., 2011; Misra et al., 2012; Mioche et al., 2010). Recently, Abdoul et al. (2020) use the CALIPSO extinction observations to assess the performance of dust extinction coefficients modeled by the Weather Research and Forecasting model with Chemistry (WRF-Chem). Besides, Xing et al. (2021) combine aerosol extinction vertical profiles from the CALIPSO and assimilated multi-layer wind profiles from the MERRA-2 to calculate aerosol extinction flux.

Meanwhile, as reported in Getzewich et al. (2018): "Extensive validation data acquired by NASA's airborne high spectral resolution lidar (HSRL) shows that during the daytime the average difference between collocated CALIPSO and HSRL measurements of 532 nm attenuated backscatter coefficients is reduced from 3.3%±3.1% in V3 to 1.0%±3.5% in V4.". In Vaughan et al. (2019): "By evaluating calibration coefficients derived using both water clouds and ocean surfaces as alternate calibration targets, and through comparisons to independent, collocated measurements made by airborne high spectral resolution lidar, we conclude that the CALIOP V4 1064 nm calibration coefficients are accurate to within 3%.". And in Kim et al. (2018): "The uncertainty in the V4 dust lidar ratio of 20% (30%) at 532 nm (1064 nm) accounts for the regional variability.". Therefore, we think that although the CALIPSO extinction coefficients are not independent quantities, but thanks to the numerous validation campaigns and the algorithm update, the CALIOP-retrieved extinction coefficients can be the credible parameters in the mass concentration calculation.

Besides, since the observation objects in our study is Sahara dust plumes, the dust lidar ratios are well-studied, e.g., Ansmann et al., 2011; Haarig et al., 2017 and the citations in these papers. With the CALIOP-retrieved backscatter coefficients, combining the Sahara dust lidar ratios, the Sahara dust extinction coefficients should be trustable.

#### <u>Reference:</u>

Ansmann, A., Petzold, A., Kandler, K., Tegen, I. N. A., Wendisch, M., Mueller, D., Weinzierl, B., Mueller, T., and Heintzenberg, J.: Saharan Mineral Dust Experiments SAMUM–1 and SAMUM–2: what have we learned?. Tellus B: Chemical and Physical Meteorology, 63(4), 403-429. https://doi.org/10.1111/j.1600-0889.2011.00555.x, 2011. Chaibou, A. A. S., Ma, X., Kumar, K. R., Jia, H., Tang, Y., & Sha, T.: Evaluation of dust extinction and vertical profiles simulated by WRF-Chem with CALIPSO and AERONET over North Africa. Journal of Atmospheric and Solar-Terrestrial Physics, 199, 105213. https://doi.org/10.1016/j.jastp.2020.105213, 2020.

Haarig, M., Ansmann, A., Althausen, D., Klepel, A., Groß, S., Freudenthaler, V., <u>Toledano, C., Mamouri, R.-E., Farrell, D. A., and Prescod, D. A.: Triple-wavelength</u> <u>depolarization-ratio profiling of Saharan dust over Barbados during SALTRACE in</u> <u>2013 and 2014, Atmos. Chem. Phys., 17, 10767-10794, https://doi.org/10.5194/acp-17-</u> <u>10767-2017, 2017.</u>

Kacenelenbogen, M., Vaughan, M. A., Redemann, J., Hoff, R. M., Rogers, R. R., Ferrare, <u>R. A., Russell, P. B., Hostetler, C. A., Hair, J. W., and Holben, B. N.: An accuracy</u> <u>assessment of the CALIOP/CALIPSO version 2/version 3 daytime aerosol extinction</u> <u>product based on a detailed multi-sensor, multi-platform case study, Atmos. Chem.</u> <u>Phys., 11, 3981–4000, https://doi.org/10.5194/acp-11-3981-2011, 2011.</u>

Mioche, G., Josset, D., Gayet, J. F., Pelon, J., Garnier, A., Minikin, A., & <u>Schwarzenboeck, A.: Validation of the CALIPSO-CALIOP extinction coefficients from</u> <u>in situ observations in midlatitude cirrus clouds during the CIRCLE-2 experiment.</u> <u>Journal of Geophysical Research: Atmospheres, 115(D4).</u> <u>https://doi.org/10.1029/2009JD012376, 2010.</u>

Misra, A., Tripathi, S. N., Kaul, D. S., & Welton, E. J.: Study of MPLNET-derived aerosol climatology over Kanpur, India, and validation of CALIPSO level 2 version 3 backscatter and extinction products. Journal of Atmospheric and Oceanic Technology, 29(9), 1285-1294. https://doi.org/10.1175/JTECH-D-11-00162.1, 2012.

Xing, Z., Li, S., Xiong, Y., & Du, K.: Estimation of cross-boundary aerosol flux over theEdmonton-Calgary Corridor in Canada based on CALIPSO and MERRA-2 data during2011–2017.AtmosphericEnvironment,246,118084.https://doi.org/10.1016/j.atmosenv.2020.118084, 2021.

Young, S. A., & Vaughan, M. A.: The retrieval of profiles of particulate extinction from Cloud-Aerosol Lidar Infrared Pathfinder Satellite Observations (CALIPSO) data: Algorithm description. Journal of Atmospheric and Oceanic Technology, 26(6), 11051119. https://doi.org/10.1175/2008JTECHA1221.1, 2009.

Young, S. A., Vaughan, M. A., Kuehn, R. E., & Winker, D. M.: The retrieval of profiles of particulate extinction from Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) data: Uncertainty and error sensitivity analyses. Journal of Atmospheric and Oceanic Technology, 30(3), 395-428. https://doi.org/10.1175/JTECH-D-12-00046.1, 2013.

Following point 1 and 2, the main part of your data procedure, the calculation of the dust volume concentration is not correct. It can not be done in the presented manner. This is not an easy point to correct and leads to my decision to reject the paper.

AR: As replied above, for the point 1 and 2, we have explained/addressed them in detail. We insist that the datasets from ALADIN/Aeolus and CALIOP/CALIPSO and the updated methodology in the revised manuscript should be solid. We kindly ask for your reconsideration. Thanks.

The horizontal flux is not well defined. The horizontal velocity is a vector with two components (East-West, North-South), so the horizontal flux should have a direction. If you just take the absolute value of the velocity, your flux may have different directions at every point. What does this help us in understanding the dust transport?

AR: Yes, thanks for your suggestion. Firstly, it should be emphasized that, in the revised manuscript, we define "dust advection" instead of "mass flux" to describe dust transportation quantificationally. The "dust advection" is the multiplication of the mass concentration (m) and the horizontal wind velocity (v), which means it is a vector. In Fig. 7 and Fig. 10 of the revised manuscript, we plot the dust advection directions of every cross-section on panel (b) to explain the dust transport. Fig. 7 and Fig. 10 are shown below. It can be seen from panel (b)s of Fig. 7 and Fig. 10 that the dust advection directions at every point of every cross-section are shown clearly.



Fig. 7 The dust advection calculated with data from ALADIN, CALIOP and ECMWF(a) the dust advection values at different cross-sections of dust plumes and (b) the dust advection directions at different cross-sections of dust plumes on 19 June 2020.



Fig. 10 The dust advection calculated with data from ALADIN, CALIOP and ECMWF (a) dust advection values at different cross-sections and at different times during the dust transport and (b) dust advection directions at different cross-sections and at different times during the dust transport

Your result, that the minimum flux occurs at dust emission (line 271 and 322) is misleading. Why should the flux be lowest at emission? Looking at your back trajectories (Fig. 8a) indicates that a significant amount of dust originated from regions west of the track on 15 June. This dust is not observed on 15 June, but on 16 June leading to a greater horizontal flux.

AR: Flament's Yes, with thanks Thomas we agree you. to recommendations/suggestions in the usage of "Mid\_bin" of L2A data, we re-produced the calculation of the mass concentration and dust advection. It is figured out by the revised calculation that the mean dust mass advection are about 1.67  $mg\cdot m^{-2}\cdot s^{-1}$  on 15 June 2020, 1.88 mg  $\cdot$  m<sup>-2</sup>  $\cdot$  s<sup>-1</sup> on 16 June 2020, 1.55 mg  $\cdot$  m<sup>-2</sup>  $\cdot$  s<sup>-1</sup> on 19 June 2020,  $0.78 \text{ mg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$  on 24 June 2020 and  $0.38 \text{ mg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$  on 27 June 2020. Actually, the mean dust advection value on 15 June is not the lowest anymore, but indeed lower than that on 16 June. In case of misleading, we addressed this statement in the revised manuscript. The slightly lower mean dust advection value occurs at dust emission on 15 June than that on 16 June results from the fact that the dust plume captured by CALIPSO and Aeolus is not the entire sources of the whole dust transportation. In the revised manuscript, it is explained as "It has to be emphasized that, according to Fig. 8(a), Aeolus and CALIPSO quasi-synchronically observed the dust plumes only at part (not whole) of the emission regions. The emission part from the West Africa (perhaps stronger than that from the central Africa) is missed and thus leading to the lower mean dust advection value on 15 June than that on 16 June."

The combination of the two satellites is a great new idea. However, you should highlight

the scientific question behind. You speak about ocean fertilization, but it remains open, which amount of dust is deposited to the Ocean. With Fig. 11, you show the low chlorophyll concentration in the studied area, but you do not quantify the effect of the discussed dust event on the ocean fertility. Your description remains very general stating that dust add nutrients to the Ocean.

AR: Yes. We only observe the long-range Sahara dust transportation by Aeolus, CALIPSO and reanalysis data and attempt to describe this event quantificationally. Actually, the study focusing on the ocean fertilization affected by Sahara dust is part of our ongoing work. According to Mills et al. (2004), in the tropical North Atlantic, community primary productivity was nitrogen-limited, and that nitrogen fixation was co-limited by iron and phosphorus. Saharan dust addition stimulated nitrogen fixation, presumably by supplying both iron and phosphorus. The mineral dust contains micronutrients such as Fe and P that have the potential to act as a fertilizer, increasing primary productivity in the equatorial Atlantic Ocean, and thus leading to N<sub>2</sub> fixation and CO<sub>2</sub> drawdown (Meskhidze et al., 2007). Consequently, the dust plumes observed in this study could be the fertilizer of Atlantic Ocean and the influence of the dust plumes deposition will be considered in our future research.

### <u>Reference:</u>

Meskhidze, N., Nenes, A., Chameides, W. L., Luo, C., & Mahowald, N.: Atlantic Southern Ocean productivity: Fertilization from above or below?. Global Biogeochemical Cycles, 21(2). https://doi.org/10.1029/2006GB002711, 2007. Mills, M. M., Ridame, C., Davey, M., La Roche, J., & Geider, R. J.: Iron and phosphorus co-limit nitrogen fixation in the eastern tropical North Atlantic. Nature, 429(6989), 292-294. https://doi.org/10.1038/nature02550, 2004.