

Response to the reviews for Kramer et al., “Apparent dust size discrepancy in aerosol reanalysis in north African dust after long-range transport”, acp-2020-1

We would like to thank Jasper Kok and anonymous referee #2 for taking the time and focus to provide their thoughtful reviews during what has been a difficult spring for everyone. These reviews are greatly appreciated. We include each one below, with responses provided in a blue font.

Jasper Kok:

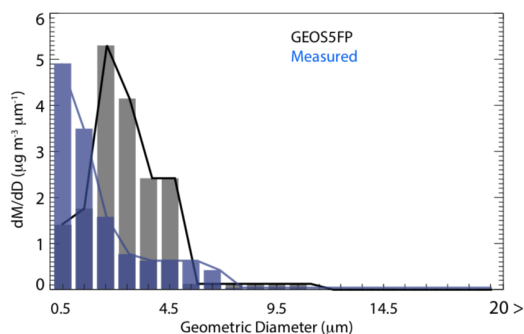
This paper investigates the size distribution of longrange transported African dust at Miami, Florida and compares in situ measurements of the (size-resolved) concentration against aerosol reanalysis data. They bring to bear a wide variety of experimental and numerical tools, and the breadth of the analysis is impressive, making for overall robust results. The authors find that the surface concentration of dust at Miami is somewhat overestimated by the reanalysis products and is also finer than represented in these (highly similar) reanalysis products. This last conclusion is counter to findings by several other studies, including my own, so is surprising and possibly important. The article is well written and is overall excellent science. I do have some comments the authors should address before publication:

#### Major comments:

1. The conclusion regarding the overestimation of dust surface concentration by the reanalysis products is confusing in the abstract. Line 7 of the abstracts notes: “Measured near-surface dust mass concentrations slightly exceed model values” whereas line 13 notes: “model dust mass concentrations near the surface are higher than those measured”. Please resolve these seemingly contradictory statements.

This is an oversight. The first statement is rewritten as “Most of the modeled dust mass resides in diameters between 2-6 micron, while most of the size-resolved measured dust mass is in diameters less than 2 micron.” (also to avoid repetition).

2. A methodological problem is that the authors are comparing modeled dust size distributions in terms of geometric (volume-equivalent) diameter  $D_g$  against measured aerodynamic diameters  $D_a$ . But because dust is both very aspherical (e.g., Okada et al., 2001; Kandler et al., 2007; Huang et al., 2020) and has a much larger density than water, a particle’s geometric diameter is smaller than its aerodynamic diameter and a correction should be made. I recommend using the simple correction based on shape data in Kok et al. (2014), namely  $D_g = 0.75 D_a$ .



Thank you very much for drawing our attention to this oversight! We have applied this simple correction, and have also replotted Fig. 9 as  $dM/dD$  - which we should have done for the initial submission of course. The new figure is shown to the left. The correction of the aerodynamically-measured diameters to a geometric

Figure 9. 17-day-average dust mass concentration per  $(\text{one } \mu\text{m})^{-1}$  diameter bin from measurements (blue) and GEOS-5 FP (black).

diameter increases the difference between the observed and modeled size-resolved dust mass concentrations. We have also rewritten section 4.4 to better describe the new figure.

3. I'm confused about GEOS-5 FP versus MERRA-2. The abstract describe these as "closely related" but section 2.4 ("GEOS-5 FP/MERRA-2") only includes a description of GEOS-5 FP and does not discuss MERRA-2. Please clarify the distinction (if any?) between these two products.

We mostly used the GEOS-5FP product for this project, as it was available prior to the MERRA-2 reanalysis. Both apply aerosol assimilation within the same model, but the GEOS-5FP aerosol product is at a 25km spatial grid spacing, while MERRA2 is global with a grid spacing of 50km. Some of the distinction is discussed on p. 2, lines 13-15 (*"The operational GEOS-5 Forward Processing (FP) model relies on a slightly more mature version of the GEOS-5 model used to produce the global Modern-Era Retrospective analysis for Research and Application-Version 2 (MERRA-2; Randles et al., 2017)."*)

To better emphasize GEOS-5 FP/MERRA-2 distinctions within section 2.4, some language has been restructured into that section (*"The GEOS-5 FP/MERRA-2 products are similar enough that they are used interchangeably within this study, reflecting their incorporation at different times of the study"*) and an additional sentence incorporated (*"Aerosol products from the operational GEOS-5 FP model are available at a 25 km grid spacing, slightly finer than the 50 km grid spacing of the global MERRA-2 reanalysis."*)

4. Data availability – the authors note data is available from the first author, but these data are valuable to the community and really ought to be posted on a publicly available repository.

This has been done. The data availability statement now includes: *"The datasets developed for this study (lidar extinction retrievals, size-resolved dust mass concentrations and measured sodium mass concentrations) are available from the University of Miami Scholarly Repository at <https://doi.org/10.17604/1b5v-h184>. These include the sun photometer aerosol optical depths, which are also publicly available through the AERONET website."*

I think Figure 9 is the paper's most salient result – that surface dust in Miami is finer than represented in aerosol reanalysis products – but some corrections need to be made to the presentation. Panel c shows dust mass concentration versus diameter in terms of bars, and a reader would reasonably conclude that the surface area of the bar is representative of the mass of aerosol in that size class. But that is not the case here, and instead the total mass in each size class is plotted. This leads to a distortion of the data in that larger size bins will be correspondingly larger (e.g., if you divide the 4-10 um bin into two separate bins, then each bin would be only half the height of the current representation). To fix this, the authors need to normalize the mass concentration by the diameter range, which is equivalent to plotting  $dM/dD$ , as is standard (although they could also plot  $dM/d\ln D$  and use a logarithmic x-axis).

Yes! We should have done this in the original submission of course. This has now been done.

#### **Minor comments:**

Line 27-29 on p. 4: "some reduction in aerosol asphericity may

also occur from chemical aging”. Measurements indicate that chemical aging of North African dust is quite limited (Denjean et al., 2015), and Huang et al. (2020) just showed that North African dust becomes more aspherical during transport, probably because of preferential settling of spherical dust particles. So this statement should be adjusted.

This has been rewritten as “A relatively consistent  $\Delta_p$  throughout transport to the eastern Caribbean can reflect a lack of atmospheric processing (Denjean et al., 2015), in which the externally-mixed dust particles may even increase in asphericity (Huang et al., 2020), although less is known of the chemical composition and shape of dust arriving at Miami via the Gulf of Mexico and subsequent westward transport over southeastern United States (Kramer et al., 2020).”

P. 11: “transport of at least 20,000 km”. Earth’s circumference is 40,000 km, so this seems incorrect.

Changed to ‘~6000 km’

Second column of Table 1 should specify that this is diameter.

Table 1 has also been modified to include an additional column indicating the geometric diameter corresponding to the impactor aerodynamic diameter thresholds.

Table 2. Length > width and aspect ratio  $\geq 1$ , so the entries in the last column (which are all < 1) should be the reciprocal number. It’d be interesting to note how the measured aspect ratios compare to other literature data on dust shape, as compiled in Huang et al. (2020).

The Table has been corrected, and a sentence added to page 9: “Although these are only five samples, their mean aspect ratio of 1.9 is equal to the median calculated from almost 78000 samples gathered in Puerto Rico (Reid et al., 2003a, b; Huang et al., 2020). This asphericity will contribute to the survival of the particles, as aspherical particles fall at slower terminal speeds than spheres of equivalent mass (Yang et al., 2013; Huang et al., 2020).”

Figure 2. If there is no data, then no data should be plotted, so please remove the zeroes for the bulk mass concentration data (8/4-8/9). [Done](#)

Figure 3: please note units for dust mass concentrations in caption. [Done](#).

Figure 7: please note whether correlation is taken in linear or logarithmic space (it ought to be the latter since the data spans several orders of magnitude). [Done](#)

## Reviewer 2

The manuscript is clearly structured and in general terms well written. In few sections it lacks quantification of results, which requires modification. The manuscript can be accepted for publication in ACP after major revisions have been considered which are specified in the following. Revisions are classified major since the adjustment of diameters to either volume equivalent or aerodynamic equivalent may alter the main conclusions.

Thank you for the comments. Please note that the adjustment of the measured diameters to their geometric value increases the discrepancy between the modeled and measured diameters.

#### SPECIFIC COMMENTS

1. The abstract contains the conclusion “Measured near-surface dust mass concentrations slightly exceed model values, with most of the modelled dust mass in diameters between 2-6  $\mu\text{m}$ .” The data shown in Fig. 6 show the opposite behaviour. Please clarify. [Done](#).

2. The results presented graphically in Figs. 5 -7 are described in the text only qualitatively as “robust correlation” (page 4, line 13), “slight underestimation” (page 7, line 22) or “clear correlation” (page 8, line 13). In all cases the results from a linear correlation analysis should be presented and discussed in the text.

[Page 4, line 13, refers to Figure 2. The correlation coefficient between the 11 independent samples is 0.98. The text has been rewritten to include this.](#)

[Page 7, line 22 refers to Fig. 5. The correlation coefficient is 0.80 \(for those comparisons for which AERONET values are available\). This is now incorporated, in both the caption and the text.](#)

[Page 8, line 13 refers to Fig. 7, for which the correlation coefficient is 0.63. This value is now included within the text, where it is also discussed .](#)

3. The presentation and discussion of dust particle sizes lacks clarity. Size distributions were measured by a cascade impactor which sorts particles according to their Stokes number and thus aerodynamic equivalent diameter which assumes unit density ( $1 \text{ g cm}^{-3}$ ). The models use spheroidal particles to describe dust particles with given densities. All values are listed in Table 1, but there is a conversion missing from aerodynamic equivalent diameter with density  $1 \text{ g cm}^{-3}$  volume equivalent diameter with densities 2.5 to  $2.65 \text{ g cm}^{-3}$ . However, this conversion is essential for the comparison of the results and needs to be included. Furthermore, the title of column 4 should be modified from “Miami” to, e.g., “Miami impactor cut-off diameter”.

[Thank you for pointing this out, as did Reviewer 1. We have expanded Table 1 to also include the geometric diameters estimated from the aerodynamic diameters and have redone Fig. 9, as shown on the first page of this response.](#)

Figure 9 showing the results from the size distribution comparison need to be adjusted to the respective size classes after the diameters have been converted to one type (either volume equivalent or aerodynamic equivalent). Besides adjusting the diameters of one data set to the other, no matter which serves as reference, the size distributions need to be presented as  $dM/d\ln D$  or  $dM/d\log D$ , or  $dM/dD$ , whatever is preferred. Otherwise the size distributions cannot be compared.

[We have redone Fig. 9 \(see page 1 of this response\).](#)

Finally, Fig. 12 lacks explanation. To my understanding, it shows the vertical profiles of mass mixing ratios for the 5 size classes of GEOS-5 FP, and thus the vertical distribution of the total dust mass mixing ratio in the model would be the sum of the five classes. If this is correct, what is then the “Average”? Here, some explanation is requested.

[Thank you for catching this, the “average” line refers to a line from a different plot that is not referenced within the manuscript. We have removed this line.](#)

#### MINOR ISSUES

Page 2, line 32: blank space between “a” and “MPL”. [Done](#).

Page 4, line 31: It might be easier to read if the equations are presented on a separate line. [Done](#).

Page 6, line 25: The Chapter title “Overview” is misleading. The title should contain an indication that here the presentation of results starts, e.g., “Overview of dust seasons from 2014 to 2016”.

[Thank you for mentioning. We have retitled this section as “Overview of 2014-2016 summer dust seasons”.](#)

Section 4.4: Figure 10 seems to be introduced before Figure 9, please check. [We were missing a reference to Fig. 9, now included.](#)

Page 9, line 35: use lower –case letter after comma. [Fixed](#).



Page 11, line 8: the term " $\tau_{aer}^s$ " is confusing, since it also may refer to the product of aerosol optical depth  $\tau_{aer}$  and a property  $s$ . Please rephrase. I have removed the use of the plural throughout ( and simplified the subscript to just be an 'a').

Page 22, Figure 2: please remove the points from the plot where no data are available. The current plot is heavily misleading. Done.

Page 28, Figure 8: it would be of high relevance to the variability range of the average frequencies connected to the respective mass concentration bins. Please show  $\pm 1\tau_a$  as error bars. These aren't average frequencies, they are the actual frequencies of the days with total dust mass concentrations falling within the specified thresholds. There's no standard deviation to plot here.

# Apparent dust size discrepancy in aerosol reanalysis in north African dust after long-range transport

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**Abstract.** North African dust reaches the southeast United States every summer. Measurements taken in Miami, Florida indicate that more than one-half of the surface dust mass concentrations reside in particles with diameters less than  $2.1\ \mu\text{m}$ , while vertical profiles of micropulse lidar depolarization ratios show dust reaching above four km during pronounced events. These observations are compared to the representation of dust in the MERRA-2 aerosol reanalysis and closely-related GEOS-5 Forward Processing (FP) aerosol product, both of which assimilate satellite-derived aerosol optical depths using a similar protocol and inputs. These capture the day-to-day variability in aerosol optical depth well, in a comparison to an independent sun-photometer-derived aerosol optical depth dataset. ~~Measured near-surface dust mass concentrations slightly exceed model values, with~~ Most of the modeled dust mass resides in diameters between  $2\text{--}6\ \mu\text{m}$ , while most of the ~~modeled dust mass~~ in diameters between  $2\text{--}6\ \mu\text{m}$  size-resolved measured dust mass is in diameters less than  $2\ \mu\text{m}$ . Modeled-specified mass extinction efficiencies equate light extinction with approximately three times as much aerosol mass, in this size range, compared to the measured dust sizes. GEOS-5 FP surface-layer sea salt mass concentrations greatly exceed observed values, despite realistic winds and relative humidities. In combination, these observations help explain, why, despite realistic total aerosol optical depths, 1) free-tropospheric model volume extinction coefficients are lower than those retrieved from the micro-pulse lidar, suggesting too low model dust loadings, and 2) model dust mass concentrations near the surface are higher than those measured. The modeled vertical distribution of dust, when captured, is reasonable. Large, aspherical particles exceeding the modeled dust sizes are also occasionally present, but dust particles with diameters exceeding ten  $\mu\text{m}$  contribute little to the measured total dust mass concentrations after such long-range transport. A further integrated assessment is needed to confirm this study's interpretations.

*Copyright statement.* TEXT

## 1 Introduction

Africa contributes almost 50% of the world's dust emissions (Huneeus et al., 2011), with easterly winds transporting the dust over the Atlantic subtropical belt throughout the full seasonal cycle. The impacts of dust span from climate-relevant time scales,

which primarily focus on dust's radiative effects, to the shorter time scales important for human endeavours. The latter include dust storms capable of closing airports, of disrupting military exercises and agriculture, and of impacting human health. The amount of dust mass in diameters less than  $2.5\ \mu\text{m}$  in particular is important for air quality, as these can penetrate human lungs (Wilson and Spengler, 1996), setting the diameter threshold used by the United States Environmental Protection Agency in meeting the conditions of the Clean Air Act. Dust mass concentrations in this size range frequently exceed established standards in the Eastern Caribbean (Garrison et al., 2014), if rarely in the southeastern United States (Prospero, 1999b).

The desire for short-term information on dust emissions and their transport has encouraged the dissemination of forecasts through, for example, the World Meteorological Organization's Sand and Dust Storm Warning Advisory and Assessment System. More accurate shorter-term predictions have resulted from a further approach, in which the aerosol forecasts are adjusted by assimilating observed aerosol optical depths ( $\tau_{aer}\tau_a$ ), primarily derived from satellite. The data assimilation allows global aerosol models to improve the spatial distribution of aerosol plumes and is particularly valuable further away from aerosol source regions, where models tend to underestimate  $\tau_{aer}\tau_a$  (Kim et al., 2014; Evan et al., 2014).

A leading global aerosol model also used for operational forecasts is the Goddard Earth Observing System Model Version 5 (GEOS-5), which uses the Goddard Chemistry Aerosol Radiation and Transport (GOCART; Ginoux et al., 2001) scheme. The operational ~~aerosol-forecast~~ GEOS-5 Forward Processing (FP) model relies on a slightly more mature version of the GEOS-5 model used to produce the ~~arguably-better-documented-global~~ Modern-Era Retrospective analysis for Research and Application-Version 2 (MERRA-2; Randles et al., 2017). ~~The-assimilated- $\tau_{aer}$~~  For both, the assimilated  $\tau_a$ , with which the simulated aerosol loadings are adjusted, primarily come from the Moderate Resolution Imaging Spectroradiometer (MODIS) satellite sensor. ~~The- $\tau_{aer}$ -biases-produced-Any  $\tau_a$  biases introduced~~ by clouds and ~~inhomogeneous-observations-is-other optical inhomogeneities are~~ corrected using a neural network based on ~~reference~~-surface-based Aerosol Robotic Network (AERONET; Holben et al., 1998)  ~~$\tau_{aer}$ s. The- $\tau_a$  as a reference. The better-documented~~ MERRA-2 Reanalysis shows no global bias relative to the surface-derived AERONET  $\tau_{aer}\tau_a$  at 550 nm (Gelaro et al., 2017). The assimilation reduces the presence of spurious trends in MERRA-2 (Gelaro et al., 2017) and clearly improves depicted atmospheric aerosol loadings compared to those in non-aerosol-assimilating simulations (Buchard et al., 2017). Importantly for dust transport, the assimilation corrects for a tendency of GEOS-5 to remove dust too quickly from the atmosphere (Buchard et al., 2017), presumably through either excessive sedimentation of the larger particles, or excessive scavenging by clouds. ~~The-GEOS-5-FP/MERRA-2-products-are similar enough that they are used interchangeably within this study, reflecting their incorporation at different times of the study.~~

The assimilated  $\tau_{aer}\tau_a$  must be redistributed into a vertical structure of the aerosol mass mixing ratio, differentiated between diverse aerosol sources, of which two (dust and sea salt) are further distributed ~~into-five-different-over-five~~ size ranges. Mass extinction efficiencies are developed using radiative transfer theory for each of the aerosols and parameterized. These allow the second moment of the size distribution, ~~namely~~-the vertically-resolved volume extinction coefficient, to be interpreted as the third moment of the size distribution, ~~or-namely~~ the aerosol mass mixing ratio fundamental to the model. The assimilation ~~corrects-allows~~ the model-specified aerosol mass mixing ratios to approximately match the observed  $\tau_{aer}\tau_a$ , so that the spatial distribution of the aerosol is improved, with the aerosol source and vertical distribution primarily set by the aerosol parameter-

izations and model physics. The model physics driving the convective transport, cloud microphysics and mixing influence how much of the aerosol is lofted further, how much is captured by precipitation, how much mixes into the boundary layer, and how much is deposited back to the surface. The particle size and vertical structure distributions are not necessarily independent, as smaller particles will be lofted higher and larger particles will settle out of the atmosphere more quickly. The aerosol size parameterizations therefore have ramifications for the subsequent transport, the resulting direct radiative effect, and the model aerosol deposition rates. In one comparison of dust vertical profiles, an Aerosol Comparison between Observations and Models (AeroCom) intercomparison of five emission-based models found significant diversity in the modeled vertical dust profiles, with large discrepancies compared to space-based lidar observations (Koffi et al., 2016).

Further understanding of global aerosol model forecasts can be gained from their assessment using *in-situ* and space-based observations. This study assesses the depiction of dust properties after long-range transport within the GEOS-5 FP forecasts and MERRA-2 reanalysis at a location slightly offshore of Miami, Florida, or ~~over 20,000~~ ~ 6000 km away from the source region. Dust is the dominant aerosol present in the Miami, Florida atmosphere during the summer months (Fig. 1), with sea salt a distant second, even at a coastal site with a consistent sea-salt presence throughout the year (Prospero, 1999a, replotted as the inset in Fig. 1)). Anthropogenic (nitrate, non-sea-salt sulfate) and biogenic (potassium, calcium) mass concentrations are low (Fig. 1). As detailed in many previous studies and easily verified with satellite imagery and the GEOS-5 forecasts, most of the dust originates from northern Africa, with the July maximum in the dust mass concentrations at Miami reflecting a combination of the easterly winds over the remote tropical Atlantic and more southerly winds over the Caribbean (~~?)~~ ~~, with less~~ (Kramer et al., 2020), and a reduced aerosol removal by precipitation in July ~~than in August compared to August~~ (e.g., v.d. Does et al., 2020). Variability in sea salt mass concentrations can be more directly related to local surface wind speeds. Overall, the relatively simple summer aerosol environment of Miami, consisting of primarily dust and sea salt, simplifies this study.

## 2 Background and Datasets

Primary observations include ground-based micropulse lidar (MPL) depictions of the dust vertical structure, surface measurements of bulk and size-resolved dust mass concentrations and sea salt mass concentrations. The daily resolved measurements span three consecutive Miami dust seasons, namely June through September of 2014, 2015 and 2016. The filter and lidar data are collected at the Rosenstiel School of Marine and Atmospheric Science campus of the University of Miami, located on Virginia Key at 25.73°N, 80.16°W, approximately four km east of the Florida mainland. Filter measurements of the bulk mass concentration are made daily from May-September, with more details on the protocol available within Zuidema et al. (2019); these basically follow Prospero (1999a). Further analysis, in support of this study, included analysis of extracts containing the major soluble inorganic ions using the samples from July and August of 2014, primarily to ascertain the sodium mass concentrations. This used flame atomic absorption to determine sodium ( $Na^+$ ) and suppressed ion chromatography to determine chloride ( $Cl^-$ ) ~~(Savoie et al., 1989)~~ along with nitrate ( $NO_3^-$ ) and sulfate ( $SO_4^{=}$ ) (Savoie et al., 1989). The sea salt concentration is computed from the sodium concentration multiplied by 3.256, following Prospero (1999a). Filter-based dust mass measure-

ments discriminated by size are also collected for 17 select days in 2016 encompassing ~~a strong synoptic event~~ strong synoptic events. Scanning electron microscopy imaging is additionally performed on select filters from this time period. Throughout, the daily dust bulk mass measurements at the surface are matched to the higher time-resolution of lidar measurements. The dust layers and their vertical extent are identified using the ~~polarization capability of a MPL~~ MPL polarization capability, and extinction profiles are retrieved from the lidar backscattered intensities for select cases following Delgadillo et al. (2018), with details relevant to this study provided below.

## 2.1 Size-resolved dust mass concentrations

Dust size distributions ~~were~~ are measured on 17 days in 2016 using a Series 230 High Volume Cascade Impactor: Multi-Stage Particulate Size Fractionator manufactured by Tisch Environmental, Inc, following Li-Jones and Prospero (1998). The instrument ~~was~~ is located directly below the bulk filters, using a secondary calibrated airflow. Air flow through three aluminum slots ~~diserminated~~ discriminates for particle aerodynamic diameters of ~~2.1, 10.2, 4.2, and 10.2 microns~~ and 2.1  $\mu\text{m}$  (see also Table 1) with one final plate capturing the remaining smaller sizes. The asphericity of dust may introduce an error in the interpretation of each size range, since the impactor selects for the particle size based on only the two horizontal dimensions. The aerodynamic diameter is related to the smaller geometric diameter through a factor of 0.75 (Kok et al., 2014).

The size-resolved dust masses are thereafter extracted identically to those on the bulk filters.

The total dust masses summed from the sized impactor samples agree well with the dust masses from the bulk samples (Fig. 2). Li-Jones and Prospero (1998) report dust masses summed from cascade impactors that are 25.4% to 53.5% less than those from bulk samples. A bias towards smaller summed dust masses from impactor samplers is also documented in a more comprehensive assessment of sizing techniques (Reid et al., 2003b). This suggests the good agreement documented here (correlation coefficient of 0.98) may be fortuitous, for example, through a difference between the true and assumed air flow. Nevertheless, the ~~robust correlation~~ clear correspondence between the two independent measures provides confidence in the size-resolved dust mass concentrations.

## 2.2 Scanning electron microscopy imaging

Scanning electron microscopy imaging ~~was~~ is done on filters from August 1 and 2 of 2016, to confirm the size of the largest dust particles and provide ~~some insight into dust geometry and composition~~ insight into the dust geometry. The imaging ~~was~~ is conducted on a 1 cm by 1 cm area of the original filter. The samples ~~were~~ are coated with palladium in a Cressington-108 Sputter Coater and ~~then~~ imaged with a Phillips XL-30/ESEM-FEG at 20kV at the University of Miami's Center for Advanced Microscopy.

## 2.3 Micropulse lidar

The MPL depolarization capability relies on a toggling between two modes using a polarizing beam splitter, with the ratio of the perpendicularly-polarized to the parallel-polarized radiation at 532 nm wavelength forming the volume linear depolar-

ization ratio ( $\delta_v$ ).  $\delta_v$  can be used to distinguish round particles, such as liquid drops and marine aerosols, from particles with distinctive aspherical shapes such as dust. In the free troposphere when dust is the dominant aerosol, the measured  $\delta_v$  is almost entirely due to dust. For pure near-source dust, Freudenthaler et al. (2009) report a particle linear depolarization ratio ( $\delta_p$ ; this includes a correction for the measured molecular contribution) ratio of  $0.31 \pm 0.03$  at 532 nm.  $\delta_p$  values for north African dust transported to the Caribbean can be slightly lower ( $0.30$  within Burton et al. (2015) and  $0.28 \pm 0.02$  within Haarig et al. (2017)), although not significantly so. A relatively consistent  $\delta_p$  throughout transport to the eastern Caribbean can reflect a lack of atmospheric processing (Denjean et al., 2015), in which the externally-mixed dust particles may even increase in asphericity (Huang et al., 2020), although less is known of the chemical composition and shape of dust arriving at Miami via the Gulf of Mexico and subsequent westward transport over southeastern United States (Kramer et al., 2020). The most likely cause is a shift to smaller sizes with a lower depolarization ratio, although some reduction in aerosol asphericity may also occur from chemical aging resulting in the accumulation of soluble material.

$\delta_v$  is a volume measurement, and when dust is mixed with other aerosols such as sea salt or sulfate, such as will occur in the boundary layer, the measured total  $\delta_v$  values will be reduced. This can be understood through  $\delta_v \sim \frac{d_{dust}X}{1-dX}$  where  $d_{dust} = \frac{\delta_v}{1+\delta_v}$

$$\delta_v \sim \frac{d_{dust}X}{1-dX} \text{ where } d_{dust} = \frac{\delta_v}{1+\delta_v} \quad (1)$$

and  $X$  is the dust's contribution to the backscattered radiation when no other depolarizing aerosols present (Sugimoto and Lee, 2006). The  $\delta_p$  of marine salt is approximately  $0.05$  (Burton et al., 2012), or much lower than that of dust.  $\delta_v$  values for north African dust after long-range transport range from  $0.20$  to  $0.30$  (Kanitz et al., 2014), attributed to mixing with other aerosols. Cloud particles, both liquid and ice, can be distinguished from dust by their higher volume backscatter intensities, with visual imagery from collocated cameras used to discriminate cloudy from cloud-free conditions (Delgadillo et al., 2018).

The Miami MPL was calibrated by the vendor, Sigma Space, prior to its delivery to Miami in 2013, and its calibration and overlap function were checked in 2016 using a mirror to horizontally send and receive the lidar laser beam through the atmosphere ~~horizontally~~, on a day chosen for ~~its~~ clear, horizontally-homogeneous conditions. The correction for the near-field overlap occurring within the laser beam is most significant below one km (Delgadillo et al., 2018), and has less influence on measurements of free tropospheric aerosol. The depolarization ratio is not sensitive to the overlap correction. The lidar is kept in a temperature-controlled environment, which reduces fluctuations in the lidar backscatter-to-extinction ratio, and is located approximately 100 m away from where the filter measurements are made. Increases in the volume depolarization ratio from 2015 to 2016 coincide with the incorporation of a new laser diode pump in May, 2016. This will increase the laser beam intensity, increasing the beam's ability to penetrate into the atmosphere, increasing the signal to noise of the signal in both channels (cross- and co-polarization), and ultimately increasing the  $\delta_v$  value. As such, the volume depolarization ratios reported here serve more as a qualitative indicator for the presence of the dust, than a quantitative measure, although more intense dust events are typically associated with higher volume depolarization ratios (at the same laser intensity), consistent with a higher dust fraction.



The MPL measures a backscattered intensity, which can only be related to the more physically-relevant aerosol volume extinction coefficient through a retrieval that is under-constrained. A sun-photometer-derived aerosol optical depth provides a vertically-integrated constraint on the extinction retrieval (Delgadillo et al., 2018). The two sun photometers, located near the lidar on a rooftop of a three-story building, are part of the Aerosol Robotic Network (AERONET; Holben et al., 1998). The Miami sun photometer version 2 data were calibrated and cloud-filtered (level 2) for 2015 ~~and solely cloud-filtered during~~  
5 ~~2014 and 2016 (level 1.5).~~ 2016. The retrieval also produces a column-average ratio of the backscattered intensity to extinction, known as the lidar ratio. This will also vary with particle size. Because the vertical column above the lidar also contains sea salt in the lower atmosphere, the column-average lidar ratio is not that of the dust in the free troposphere. Lidar ratios range from 40-60 sr for dust and 15-25 sr for sea salt (Burton et al., 2012); a column-average lidar ratio less than that appropriate for dust is consistent with an overestimate in the lidar-retrieved extinction. ~~The optical~~ Optical properties of dust ~~are not thought~~  
10 ~~to do not~~ vary with relative humidity (Maring et al., 2003a) (Maring et al., 2003a; Denjean et al., 2015), but those for sea salt, a ~~hydrophillic~~ hygroscopic aerosol, will. We estimate the overall uncertainty in the retrieved lidar extinction conservatively with a factor of three. The lidar vertical resolution is 30 m and the time resolution is 15 seconds (done to facilitate detection of the small cumulus clouds common to Miami).

## 2.4 GEOS-5 FP/MERRA-2

15 The GEOS-5 FP/MERRA-2 products are similar enough that they are used interchangeably within this study, reflecting their incorporation at different times of the study. Aerosol products from the operational GEOS-5 FP model are available at a 25 km grid spacing, slightly finer than the 50 km grid spacing of the global MERRA-2 reanalysis. The GEOS-5 model possesses 72 vertical layers, of which the mid-level of the lowest layer is at approximately 69 meters. ~~Data from the lowest model layer are compared to the measured surface aerosol concentrations.~~ The aerosol assimilation occurs eight times per day, and the output  
20 frequency of the full three-dimensional aerosol field is also every three hours. We primarily consider the dust and sea salt contributions. The GOCART dust emission parameterization depends on a source function, the near-surface wind speeds, and soil moisture (Ginoux et al., 2001). The dust sizes, prescribed at emission, can evolve thereafter during transport, with sedimentation the primary process capable of altering the dust size distribution (the dust is non-hygroscopic). The parameterization of the dust mass distribution by size approximately follows that of Tegen and Lacis (1996) and contains five size categories,  
25 extending to a maximum diameter of 20  $\mu\text{m}$ . These are indicated in Table 1 along with the density and mass extinction efficiency (ratio of extinction to mass) corresponding to each size category. Spheroidal particles are assumed, with more details on the dust parameterization available in Colarco et al. (2010). ~~The dust is assumed to not be hygroscopic.~~

The flux of sea salt off of the ocean surface is also parameterized into five size bins, but the size is allowed to vary with the surface wind speeds and sea surface temperature, and the sea salt can undergo hygroscopic growth as a function of the relative  
30 humidity. The sea salt parameterization is adapted from Gong (2003) and described further in Chin et al. (2002) and Bian et al. (2019).

~~Mass extinction efficiencies—the ratio of extinction to mass, specific for each diameter size range (see Table 1 for the dust mass extinction efficiencies)—connect the model aerosol mass mixing ratios to the assimilated aerosol optical depth. The~~

clear-sky  $\tau_{aer} T_g$  values at a wavelength of 550 nm are primarily assimilated from the Moderate Resolution Imaging Spectroradiometer (MODIS) and secondarily the Multi-Angle Imaging SpectroRadiometer (MISR) satellite instruments (Randles et al., 2017). Data from the surface-based AERONET sites are no longer assimilated (Randles et al., 2017), allowing the Miami sun photometer data to independently validate the GEOS-5 FP  $\tau_{aer} S T_g$ . The lowest model level is assumed to represent surface values. [Inspection of the MERRA-2 and GEOS-5 FP dust and sea salt profiles for the cases shown in Fig. 11 revealed little difference between the two products.](#)

## 2.5 HYSPLIT back trajectories

Daily average back-trajectories explore the differences in the integrated air flow for days with high and low dust mass concentrations. The calculations rely on the NOAA Air Resources Laboratory HYSPLIT model (Stein et al., 2015). These extend for 300 hours (12.5 days) and are driven by the NCEP Reanalysis Climate Diagnostics Center-1 product. Each day's trajectory is initialized at 00Z and at an altitude of 2000 m. This is a robust height for the presence of dust above Miami, as indicated later. There is some evidence that the NCEP Reanalysis lower tropospheric winds may be weaker than observed (e.g., Adebiyi et al., 2015), which would increase the transport time, but this is not investigated further for this study.

## 3 Overview of 2014-2016 summer dust seasons

A time series of the primary measurements displays the daily surface-based filter-derived dust mass concentrations and the corresponding vertical structure as captured by the lidar volume depolarization ratio, for each of the three dust seasons (Fig. 3). The lidar measurements indicate that dust is often prevalent in the boundary layer, typically extends up to 2-3 km, and occasionally rises above 4 km but not above 5 km. The depolarization ratios indicate that when dust is present, it is typically also present within the boundary layer. This is corroborated by the filter measurements; only during mid-to-late August 2015 is no dust detected within the atmospheric column by either the filters or the lidar. The synoptic and year-to-year variability is clear, with the highest dust mass concentrations occurring on just a few select days of each summer.

The back-trajectories indicate the crucial role of the north Atlantic subtropical high in guiding the atmospheric flow towards Miami (Fig. 4(a-c)), explored further in [Kramer et al. \(2020\)](#). Many back-trajectories intersect the coastal region off of North Africa, with few continuing further eastward over the African continent. This may indicate a strong influence from the northerly coastal flow on the eastward side of the north Atlantic subtropical high. The HYSPLIT back-trajectories disregard explicit mixing with environmental air, but a back-trajectory with a large meridional component would imply more mixing of the dusty air with air emanating from the northeast Atlantic and Europe. ~~That this occurs is consistent with the co-existence of anthropogenic aerosols with dust (Fig. 1).~~ The separate influences of African versus Atlantic air on Miami and Caribbean thermodynamic profiles is also noted in Dunion (2011).

After twelve days, the integration confidence in the trajectories leading to the original location of air parcel is debatable, therefore we focus more on the trajectory characteristics over the Atlantic. Back-trajectories associated with days with dust mass concentrations exceeding  $20 \mu\text{g m}^{-3}$  are typically more zonal, suggesting less mixing with air originating from the

northeastern Atlantic. Back trajectories associated with days with high dust mass concentrations also have a slight preference  
 5 for a more northerly location. This may indicate less aerosol removal by precipitation en route (the northern boundary of the  
 JJA intertropical convergence zone is at approximately  $10^\circ$  N). A notable feature of the back-trajectories on days with low  
 ( $< 1 \mu\text{g m}^{-3}$ ) and moderate ( $1 - 20 \mu\text{g m}^{-3}$ ) dust mass concentrations in Miami is a less direct transport of dust, with the  
 back-trajectories passing over the Gulf of Mexico and then recirculating back to Miami via the lower southeast United States.  
 This is most evident in 2014 and 2015. This transport pathway is effective at dispersing dust over large areas of southeastern  
 10 United States, and increase the likelihood of dust interacting with other terrestrial and anthropogenic aerosols en route.

## 4 Comparison of GEOS-5 FP/MERRA-2 aerosol distributions to the Miami observations

### 4.1 Aerosol optical depth

The GEOS-5 FP  ~~$\tau_{aer}$  values~~  $\tau_a$  values (at 550 nm wavelength) slightly exceed those from the sun photometers ~~at (at 500 nm)~~  
 at optical depths below 0.15, and are slightly less at  ~~$\tau_{aer}$~~   $\tau_a$   $> 0.15$  (Fig. 5). The slight underestimation at higher  ~~$\tau_{aer}$~~   $\tau_a$ s is  
 15 also shown in Shi et al. (2019), in which it is attributed to missing emissions (Buchard et al., 2017), ~~and~~ It is also apparent in  
 a comparison of MERRA-2  ~~$\tau_{aer}$ s to shipborne~~  $\tau_{aer}$   $\tau_a$  to shipborne  $\tau_a$  observations (Fig. 9 of Randles et al., 2017). Surface  
 albedo inhomogeneities at this coastal location could perhaps contribute to the overestimate at the lowest  ~~$\tau_{aer}$ s~~  $\tau_a$ , although  
 another interpretation, shown next, is that an overestimate from the sea salt contribution becomes most apparent at the lowest  
 ~~$\tau_{aer}$ s~~  $\tau_a$ . An implication of the positive correlation (coefficient of 0.80), overall, is that the AERONET cloud screening is  
 20 effective. Although not shown, the day-to-day variations in  ~~$\tau_{aer}$~~   $\tau_a$  match well, as would be expected given the assimilation  
 and availability of satellite-derived  ~~$\tau_{aer}$ s~~  $\tau_a$ .

### 4.2 Surface sea salt mass concentrations

A first step within the assimilation scheme is the aerosol speciation, and how much of the aerosol is partitioned into sea salt  
 will also affect the amount available for dust. The GEOS-5 FP aerosol product overestimates the near-surface salt mass con-  
 25 centration by an order of magnitude, with a model mean value of  $61 \mu\text{g m}^{-3}$  versus a measured mean salt mass concentration  
 of  $7.7 \mu\text{g m}^{-3}$  estimated from the sodium measurements (Fig. 6). The latter is consistent with previous measurements (Savoie  
 and Prospero, 1982; Prospero, 1999a). Further assessment of the GEOS-5 wind speeds and relative humidity using surface  
 meteorological data indicate that these are realistic (not shown), suggesting that the underlying size and/or light extinction pa-  
 rameterizations may be the issue. This is consistent with the findings of Bian et al. (2019), in which an inconsistency between  
 30 an overpredicted salt mass concentration and underpredicted  $\tau_{seasalt}$  in GOES-5 simulations is reconciled through the model  
 adoption of too-large sea salt sizes. As noted by Buchard et al. (2017), the assimilation of  ~~$\tau_{aer}$ s that exceed the forecast~~  $\tau_{aer}$ s  
aerosol optical depths that exceed those forecast can further exacerbate the discrepancy.

### 4.3 Surface dust mass concentrations

All else equal, an overestimate of the GEOS-5 FP sea salt mass concentrations might point to GEOS-5 FP dust mass concentrations that are too low, for the same assimilated aerosol optical depth. Instead, the GEOS-5 FP near-surface dust mass concentrations also exceed observed values on almost two-thirds of the days, although the overestimation is not as pronounced as that for sea salt, with a mean bias of  $3.46 \mu\text{g m}^{-3}$  (Fig. 7). A clear correlation (value of 0.63) is apparent, matching that reported in Buchard et al. (2017) for MERRA-2 based on Barbados measurements (~~which~~ Buchard et al. (2017) did not find a systematic bias). GEOS-5 FP approximately captures the monthly evolution from July to September, although the GEOS-5 FP overestimate is most pronounced for June, and is enough to alter the perception of the monthly evolution (i.e. the GEOS-5 FP maximum in 2014 and 2016 occurs in June, in contrast to a July (August) maximum in the observations). The cause for this is unclear.

The model overestimate in the surface dust mass concentrations has consequences for inferences of the frequency of high and low dust loading days (Fig. 8). Measured dust mass concentrations exceeding  $40 \mu\text{g m}^{-3}$  or less than  $1 \mu\text{g m}^{-3}$  were infrequent, with dust present in the Miami boundary layer at concentrations between 1 to  $10 \mu\text{g m}^{-3}$  over one-half of the time. In contrast, GEOS-5 FP include more days with concentrations exceeding  $10 \mu\text{g m}^{-3}$  than are observed, and correspondingly fewer days with concentrations between  $1\text{--}10 \mu\text{g m}^{-3}$ . GEOS-5 FP overall places too much dust in the boundary layer. ~~That dust is consistently present~~ The consistent presence of dust in the boundary layer is not controversial (e.g., Reid et al., 2002).

### 4.4 Does dust size explain the difference between the GEOS-5 FP/MERRA-2 and measured dust mass concentrations?

The use of a size distribution parameterization that permits a larger mass for the same visible extinction (as is thought to occur for sea salt; Bian et al. (2019)) is one explanation for why the MERRA-2 dust mass concentrations may exceed those observed, despite realistic assimilation-constrained aerosol optical depths. This idea is assessed using size-resolved dust mass concentrations from 17 days in 2016, collected in addition to the bulk filter samples. The 28 July to 9 August time period included one of the heaviest and longest dust mass concentration events from 2016, and data from another dust event spanning 1-2 September are also included. The trajectories for these days indicate ~~the dust transport occurred directly over the Atlantic to Miami without passing over the Gulf of Mexico and recirculating back~~ direct dust transport from northern Africa. By happenstance, the subtropical high was strongest and located furthest north in 2016 of the three years considered, based on the 1023 hPa sea level pressure contour (Fig. 4). This may have decreased dust transport that year (~~?~~) (Kramer et al., 2020), resulting in relatively fewer days with dust exceeding  $20 \mu\text{g m}^{-3}$  for that year. The episodic-maximum dust mass concentration of  $28 \mu\text{g m}^{-3}$  occurred on August 4, 2016 (Fig. 2). The days with the largest size particles (7/30, 8/2, and 8/4) correspond to high lidar volume depolarization ratios and total bulk surface dust mass concentrations (Fig. 3).

~~Overall, the GEOS-5 FP size distribution is broader than that measured, with GEOS-5 FP placing 16% of the dust mass concentration in diameters  $< 2$~~  The comparison is eased by first estimating the geometric diameter corresponding to the measured aerodynamic particle diameter using a factor of 0.75 (Kok et al., 2014), and thereafter normalizing the measured

and modeled size-resolved dust mass concentrations into diameter bins with a  $1\text{ }\mu\text{m}$ , with 46%, 32%, 5.4% and 0.05% in the subsequent larger size bins. In contrast, the measurements place 56% width (Fig. 9). The measurements place 74% of the total dust mass concentrations in particles with geometric diameters  $< 2.1\text{--}2\text{ }\mu\text{m}$ , with 18%, 21%, and 3.6% in the larger size bins. We note that the model the presence of some larger particles resulting in a mass-weighted mean measured diameter of  $3.0\text{ }\mu\text{m}$ . The GEOS-5 FP size distribution places equal mass in the three size bins with diameters exceeding most of the dust in diameters between  $2\text{--}3\text{ }\mu\text{m}$  (almost 30%), with 40% within the  $3\text{--}6\text{ }\mu\text{m}$  diameter range and almost no mass in larger diameters. Less than 25% is placed into diameters  $< 2\text{ }\mu\text{m}$ , or approximately one-third of the amount measured in this size range. The GEOS-5 FP mass-weighted mean diameter is thus larger than observed, at  $3.9\text{ }\mu\text{m}$ .

Particles with diameters  $< 2\text{ }\mu\text{m}$ , and that thereafter, the only mechanism that can alter the dust sizes is differential settling. This effect is evident in the possess larger mass extinction efficiencies than larger particles (Table 1), by at least a factor of three, implying that more mass is required to produce the same extinction for larger particles than for smaller particles. Fig. 9 indicates that one cause for the overestimated near-surface dust mass concentrations in GEOS-5 dust size distribution after long range transport, with FP above southern Florida may be a dust size parameterization that distributes most of the mass contained in the  $(2.0\text{--}3.6\text{ }\mu\text{m})$  diameter range, and the mass distribution within the three larger size bins no longer equal. As has been previously noted by others (e.g., Kok, 2011) dust mass into a larger size, for which the mass extinction efficiency is lower.

Though difficult to see in Fig. 9, more of the measured mass is contained in particles with diameters  $> 6\text{ }\mu\text{m}$ , than in the model (10% versus 4%). This type of model behavior has been previously noted (e.g., Adebisi and Kok, 2020, and references therein). Filters from 1 and 2 August indicate mass from particles with diameters  $> 10\text{ }\mu\text{m}$ , and these were interrogated further with scanning electron microscopy to verify the particle size (Fig. 10). The dust particle images independently corroborate the presence of larger sizes. Four of the five examples have a dimension exceeding  $20\text{ }\mu\text{m}$  (see Table 2 for dimensions of each identified particle), with the largest particle measuring  $36.65$  by  $26.86\text{ }\mu\text{m}$  (Fig. 10b). The particles are highly aspherical, with aspect ratios ranging from 0.38 to 0.73. This could contribute to their survival 1.36 to 2.6. Although these are only five samples, their mean aspect ratio of 1.9 is equal to the median calculated from almost 78000 samples gathered in Puerto Rico (Reid et al., 2003a, b; Huang et al., 2020). This asphericity will contribute to the survival of the particles, as aspherical particles will fall at slower terminal speeds than spheres of equivalent mass (e.g., Yang et al., 2013). These (Yang et al., 2013; Huang et al., 2020). These observed dust particles exceed the GEOS-5 upper limit of a  $20\text{ }\mu\text{m}$  diameter. However, only  $1\text{--}0.45\text{ }\mu\text{g m}^{-3}$  of the total measured dust mass concentration (approximately 3%) is contained in particles with diameters exceeding  $10\text{ }\mu\text{m}$ , implying that the neglect of particles with diameters  $> 20\text{ }\mu\text{m}$  by GEOS-5 FP only results in a small error in the total dust mass concentration after such long-range transport.

The more striking result is that the measurements place more than half of the mass in particles with diameters  $< 2\text{ }\mu\text{m}$ , or more than three times as much as does GEOS-5 FP. Particles with diameters  $< 2\text{ }\mu\text{m}$  possess larger mass extinction efficiencies than larger particles (Table 1), by at least a factor of three, implying that more mass is required to produce the same extinction for larger particles than for smaller particles. Fig. 9 indicates that one cause for the overestimated near-surface dust mass concentrations in GEOS-5 FP above southern Florida may be a dust size parameterization that distributes most of the dust mass into a larger size, for which the mass extinction efficiency is lower.

We are not aware of other dust size distribution measurements gathered over coastal Florida against which to compare this study's measurements. Measurements made close to the African coast emphasize the presence of larger particles, with Ryder et al. (2013) reporting most of their measured dust mass in diameters exceeding  $5\text{ }\mu\text{m}$ , consistent with Haywood et al. (2003). Kok et al. (2017) show that most of the dust mass at emission is contained in particles with diameters reaching  $10\text{ }\mu\text{m}$ . The largest particles sediment out during transport, and in-situ measurements made during the Puerto Rico Dust Experiment most of the mass is in particles with diameters between  $5\text{--}7\text{ }\mu\text{m}$  (Maring et al., 2003b; Reid et al., 2003e), with similar sizes between the boundary layer and the free troposphere (Maring et al., 2003a). More of the dust mass is reported in larger sizes than is reported here for south Florida. In contrast, inferences of dust size deduced from multi-wavelength lidar measurements at Barbados (Haarig et al., 2017) estimate most dust particles are  $1.6\text{--}2.0\text{ }\mu\text{m}$  in diameter, which is more broadly consistent with the Miami impactor measurements. This is at first glance consistent with the in-situ dust mass concentrations reported in Jung et al. (2013), but these do not extend beyond  $2.5\text{ }\mu\text{m}$  in diameter (most aircraft aerosol intake inlets cut off at  $3\text{ }\mu\text{m}$ ), thus do not resolve the larger sizes.

#### 4.5 Dust vertical structure

The GEOS-5 FP dust mass mixing profile combined with the mass extinction efficiencies specific to each size range (Table 1) can be assessed using extinctions retrieved from the lidar backscattered intensities (Delgadillo et al., 2018). This is similar to the strategy invoked within Liu et al. (2012) for comparison between space-based lidar extinctions and GEOS-5 dust mass loadings, using a mass extinction efficiency of  $0.7\text{ m}^2\text{ g}^{-1}$ , and Sauter et al. (2019) (who use a smaller value of  $0.4\text{ m}^2\text{ g}^{-1}$ , implying larger particles). Four examples, spanning the 28-30 July 2016 dust event and that of 5 August, 2016, are shown in Fig. 11. The AERONET-derived  $\tau_{aer}$  from the early morning, overlapping with 12 UTC, indicate  $\tau_{aer}$  of 0.1-0.2, with the highest aerosol loading occurring on 29 July, 2016.

The time series of the lidar-derived extinctions (Fig. 11c,f,i and l, all corresponding to nighttime, when the lidar signal is more robust) indicate dust extending up to 4 km at times, stratified into distinct layers supporting what appear to be gravity waves (Fig. 11). A peak at 2 km is specific to the 28-30 July dust event (Fig. 3), and a secondary peak at approximately 800 m represents the boundary layer top, where the maximum in relative humidity may induce the hygroscopic swelling of sea salt (or of other hygroscopic aerosols mixed with dust) spray aerosols, or indicate undetected optically-thin cloud (Delgadillo et al., 2018).

The GEOS-5 FP dust mass concentrations clearly indicate the presence of dust in the free troposphere on two of the days, though, perhaps surprisingly, not on the day with the highest AERONET-derived optical depth. When dust is present, its vertical distribution is similar to that inferred from the lidar. Within the boundary layer, the filter-based size-resolved dust mass concentrations measurements (thick red bars within Fig 11a,d,g and j) reach  $10\text{ }\mu\text{g m}^{-3}$  or more on all four days. GEOS-5 FP near-surface dust mass concentration values are higher on all four days than those measured (compare to the thick black bars).

Corresponding model aerosol extinction profiles, resolved over the 3-12 UTC time frame encapsulated within the average of the cloud-free lidar extinction profiles, indicate that the GEOS-5 FP product does not distribute enough of the assimilated  $\tau_{aer}$



$\tau_a$  above 1.5 km on any of the four days (Fig. 11b,e,h,k). Most of the model  $\tau_{aer}$  is confined to the boundary layer, where the  $\tau_{aer}$  also contains a substantial sea salt spray contribution given relative humidities capable of exceeding 80% (RH not shown). The lidar extinction values within the boundary layer are much lower than those from GEOS-5 FP, and, are lower than those in the free-troposphere, despite including a sea salt contribution. Overall this assessment suggests that GEOS-5 FP places too much dust within the boundary layer, and not enough in the free troposphere above the boundary layer, where the dust can be advected further more easily. Where GEOS-5 FP does place dust within the free troposphere, its vertical distribution is reasonable it is reasonably distributed by altitude, however.

Agreement between the model-derived and observed lidar extinctions, for the same dust mass concentrations, can in theory be produced through the application of higher mass extinction efficiencies. The model tendency to overestimate the mean particle size could also be one reason why the model may place too much of the dust at lower altitudes (Fig. 9). Since larger particles fall more quickly, they should be preferentially located at lower altitudes. Size-resolved model dust mass mixing ratios for 28 July 2016 indicate that larger particles do prefer lower altitudes (Fig. 12), and in particular much fewer particles with diameters  $> 6 \mu\text{m}$  occur above the boundary layer. In the two size ranges with the largest discrepancy between the model and measurements (2-3.6  $\mu\text{m}$  versus 0.2-2  $\mu\text{m}$ ), the altitude difference between the vertical distribution of the two size-resolved dust mass mixing ratios is only on the order of 100-300 m, however.

## 5 Conclusions and Discussion

Dust forecasts incorporating the assimilation of satellite-derived  $\tau_{aer}$  can circumvent a difficulty global aerosol models otherwise encounter, in which  $\tau_{aer}$  tend to be too low further away from a source region (e.g., Kim et al., 2014; Evan et al., 2014; Ansmann et al., 2017). As such, the assimilation of observed  $\tau_{aer}$  holds the promise of more accurate depictions of the global aerosol distribution. While the GEOS-5 FP aerosol forecasts and MERRA-2 capture independently-measured  $\tau_{aer}$  and their  $\tau_a$  and their spatial variability well for an aerosol environment dominated by sea salt and dust transported far from its source regions spray and long-range dust, the more challenging objectives of realistic aerosol vertical distributions and surface mass concentrations are less well met. A clear overestimate in the modeled sea salt loading, apparent in an *in situ* comparison (Fig. 6) and a comparison of model-derived extinctions to those from a lidar (Fig. 11), is corroborated by the independent findings of Bian et al. (2019). This will foster an underestimate in the dust loading, for the same assimilated  $\tau_{aer}$ .

A comparison to dust mass concentrations measured at the surface and to lidar profiles of retrieved extinction indicates that GEOS-5 FP often distributes dust too low in the atmosphere, with too much mass placed in particle sizes that are larger than observed (except at the largest sizes). An overestimate of the amount of dust in the boundary layer has implications for model-deduced ocean fertilization by the soluble iron (e.g. Colarco et al., 2003), and for cloud nucleation. Size-resolved measurements place most of the mass in diameters smaller than 2  $\mu\text{m}$ , while GEOS-5 FP/MERRA-2 places most of the dust mass in diameters between 2 to 3.6  $\mu\text{m}$  (Fig. 9). The prescribed dust mass extinction efficiency, by which the dust portion of the assimilated aerosol optical depth is cast as a dust mass mixing ratio, is more than a factor of 3 larger for the smaller size (Table 1). Thus, a model dust size distribution that places more of the dust in the smaller sizes, but otherwise using

the same prescribed size-resolved mass extinction efficiencies, will place less dust mass within the boundary layer. A relative increase in the amount of dust within the smaller sizes in the free troposphere may be important for the overall large-scale spatial distribution of the dust, although the vertical distribution of dust differs little for the smaller sizes (Fig. 12). slightly larger sizes, will overestimate the dust mass even if the light extinction by dust is correctly represented.

The overestimate in the model dust size after long-range transport is opposite to that documented for most global aerosol models, in which the number of small particles can be overestimated relative to the large particles (e.g., Kok, 2011). We recognize a recent emphasis on the presence of very large dust particles within the Saharan Air Layer (e.g., Ryder et al., 2019), and their ability to be transported for long distances. Large aspherical dust particles are also detected at Miami after a transport of ~~at least 20,000 ~ 6000~~ km, exceeding the amount modeled, but their contribution to the overall mass is negligible ~~at  $1 \mu\text{g m}^{-3}$~~ . Their presence is consistent with other observations that perceive little mixing of African dust with other air masses en route for select cases (Karyampudi et al., 1999), and find larger dust particles closer to the top of the Saharan air layer (Jung et al., 2013; Yang et al., 2013; Gasteiger et al., 2017). The existence of the large particles may be more typical of the dust events that advect more directly to Miami and undergo little precipitation. The neglect of particles exceeding the maximum GEOS-5-specified particle diameter of  $20 \mu\text{m}$  introduces little error in the total GEOS-5 dust mass concentrations, but may be more important for the direct aerosol radiative effect, which is not addressed here. ~~Overall global aerosol models with and without aerosol assimilation benefit from a more realistic modeling of dust particle size and its evolution with transport and age after emission (Adebisi et al., 2019).~~

~~Our dust size measurements, at a location further away from the dust source than Puerto Rico (Reid et al., 2002, 2003c; Maring et al., 2003) and Barbados (Jung et al., 2013; Weinzierl et al., 2016), are difficult to compare in a consistent manner to those reported in previous studies.~~ We are not aware of other dust size distribution measurements gathered over coastal Florida. Kok et al. (2017) show that most of the north African dust mass at emission is contained in particle diameters reaching  $10 \mu\text{m}$ . Ryder et al. (2013) report that most of their dust mass, in measurements made close to the African coast, is in diameters exceeding  $5 \mu\text{m}$ , consistent with Haywood et al. (2003). *In-situ* measurements made during the Puerto Rico Experiment suggest Dust Experiment place most of the ~~dust mass resides in~~ mass in particles with diameters between  $5\text{--}7 \mu\text{m}$ , while (Maring et al., 2003b; Reid et al., 2003c) , with similar sizes between the boundary layer and the free troposphere (Maring et al., 2003a). The Denjean et al. (2015) study, based on measurements taken in Puerto Rico, does not consider particle diameters  $< 1 \mu\text{m}$ . In contrast, inferences of dust size deduced from multi-wavelength lidar measurements at Barbados (Haarig et al., 2017) estimate most dust particles are  $1.6\text{--}2.0 \mu\text{m}$  in diameter, which is more broadly consistent with the Miami impactor measurements. Other recent lidar measurements ~~tend to also~~ perceive larger concentrations of aerosols with diameters  $< 2 \mu\text{m}$  (Haarig et al., 2019). ~~Reasons for such~~ This is at first glance consistent with the *in-situ* dust mass concentrations reported in Jung et al. (2013) near Barbados, but these do not extend beyond  $2.5 \mu\text{m}$  in diameter (most aircraft aerosol intake inlets cut off at  $3 \mu\text{m}$ ), and do not resolve the larger sizes.

Our dust size measurements, at a location further away from the dust source than Puerto Rico (Reid et al., 2002, 2003a, c; Maring et al., 2003) and Barbados (Jung et al., 2013; Weinzierl et al., 2016), are difficult to compare in a consistent manner to those reported in previous studies. Reasons for the size discrepancies are not entirely understood. Neither the impactor measurements nor the GOCART dust size parameterization discriminates for aerosol diameters  $< 2 \mu\text{m}$ , ~~and published dust size distributions are not~~

~~always directly comparable. Although critiques~~, Critiques can be made of each individual measurement presented within this study ~~, in their totality~~ (see, e.g., Reid et al. (2003b) for a full discussion of uncertainties with impactor measurements, while also affirming the value of this fundamental measurement). We cannot discount that particle break-up may contribute to more dust mass at smaller particle sizes for the cascade impactor, and have neglected removal of the larger sizes by cloud processing.

10 In their totality, however, a consistent interpretation emerges based on the retrieved lidar extinctions, and near-surface dust and sea salt mass concentrations, suggesting the model mean dust sizes, by mass, are too large, ~~with~~ leading to relatively too much dust mass placed in the boundary layer. ~~The~~ At the same time, the total dust loading is too low within GEOS-5 FP and MERRA-2, possibly because too much of the assimilated aerosol optical depth is speciated into sea salt. We consider the current analysis a pilot study, however, and recommend a further dedicated assessment with a multi-wavelength depolarization extinction lidar

15 as well as a more complete set of size-resolved in-situ measurements, both at the ground and in the free troposphere. This will better anchor ideas for ~~changes~~ future improvements to the GEOS-5 sea salt and dust size parameterizations.

*Data availability.* The datasets developed for this study (lidar extinction retrievals, size-resolved dust mass concentrations and measured sodium mass concentrations) are available from the University of Miami Scholarly Repository at <https://doi.org/10.17604/1b5v-h184>. These include the sun photometer aerosol optical depths, which are also publicly available through the AERONET website. The bulk dust mass

5 concentration dataset is available through the University of Miami Scholarly Repository at <https://doi.org/10.17604/q3vf-8m31>, for which Zuidema et al. (2019) is the appropriate reference. The HYSPLIT model can be accessed through <https://www.ready.noaa.gov/HYSPLIT.php>, a NOAA Air Resource Laboratory product.

*Author contributions.* PZ designed the study and led the writing of the manuscript. SK carried out the data collection and initial analysis and drafted the initial manuscript. The GEOS-5 FP and MERRA2 values were provided by RG. RD provided the lidar analysis. CA contributed

10 to the Miami data collection, with LC providing input on the methodology and additional mentorship of SK. AB provided the SEM analysis. PC provided substantial insight into the GEOS-5 and MERRA2 methodology. All authors commented on the manuscript.

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**Table 1.** Particle geometric diameter range applied within the GEOS-5 dust size parameterization, corresponding mass extinction efficiencies (MEE) at 550 nm wavelength ~~based on the~~ assuming non-spherical optics ~~model presented in Colarco et al. (2014)~~ (Colarco et al., 2014), and Miami impactor ~~size selection~~ thresholds, in both aerodynamic and geometric diameters ( $D_a$  and  $D_g$ ), where  $D_g=0.75D_a$  (Kok et al., 2014)

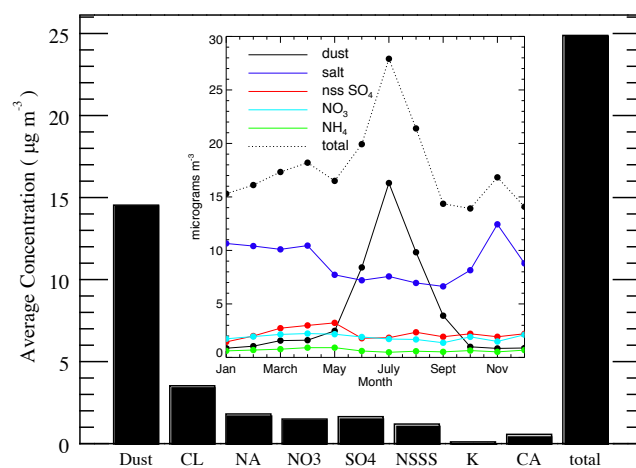
density kg m <sup>-3</sup>	GEOS-5 FP $\mu\text{m}$ ( <u><math>D_g</math></u> )	MEE m <sup>2</sup> g <sup>-1</sup>	Miami $\mu\text{m}$ ( <u><math>D_a</math></u> )	<u>Miami</u> $\mu\text{m}$ ( <u><math>D_g</math></u> )
2500	0.2-2.0	2.02	< 2.1	<u>&lt; 1.575</u>
2650	2.0-3.6	0.64	2.1-4.2	<u>1.575-3.15</u>
2650	3.6-6.0	0.33	4.2-10.2	<u>3.15-7.65</u>
2650	6.0-12.0	0.17	>10.2	<u><math>\geq 7.65</math></u>
2650	12.0-20.0	0.08	NA <sup>1</sup>	<u>NA<sup>1</sup></u>

<sup>1</sup>not available

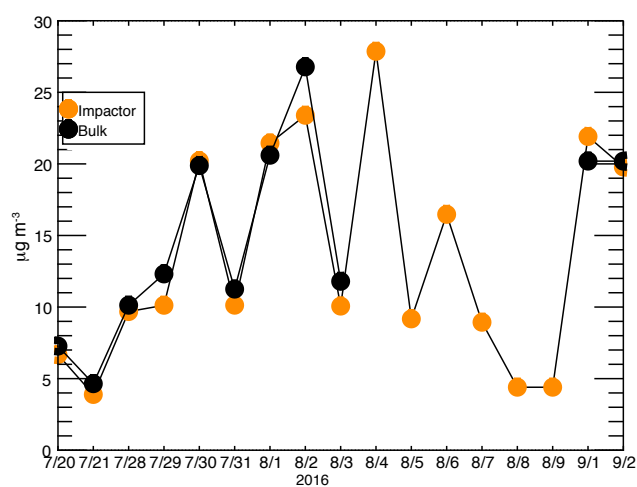
**Table 2.** Measured size of imaged particles in Fig. 910.

particle <sup>1</sup>	length $\mu\text{m}$	width $\mu\text{m}$	length/width
a 1	26.8	11.5	<del>0.43</del> <u>2.33</u>
a 2	17.0	12.0	<del>0.70</del> <u>1.42</u>
b	36.6	26.9	<del>0.73</del> <u>1.36</u>
c	27.8	14.6	<del>0.52</del> <u>1.9</u>
d	21.6	8.3	<del>0.38</del> <u>2.6</u>

<sup>1</sup> indicated by panel label and image number

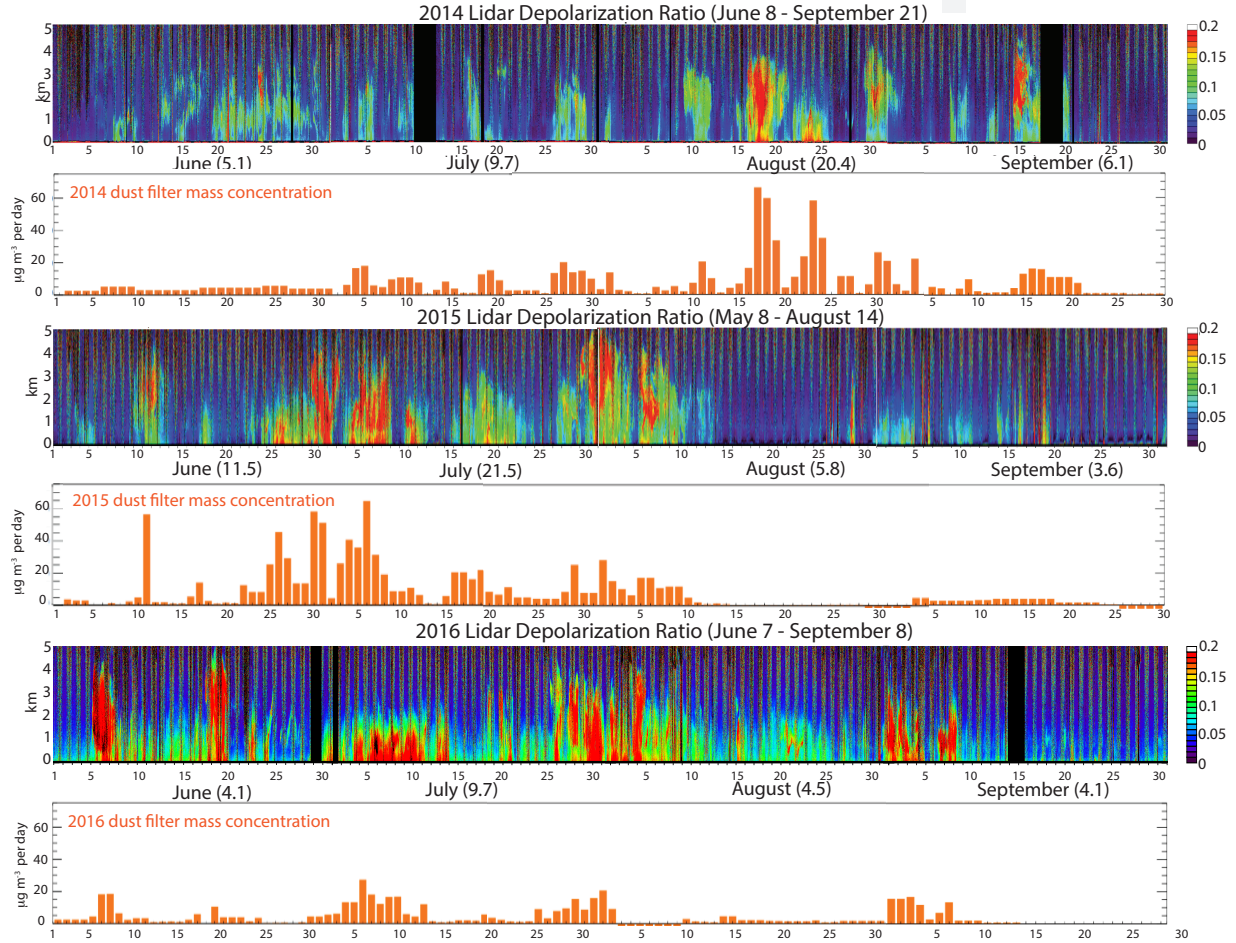


**Figure 1.** Average aerosol concentrations for July - August 2014 of the soluble ions chlorine (CL), sodium (NA), nitrate (NO<sub>3</sub>), sulfate (SO<sub>4</sub>), and non-sea-salt sulfate (NSSS), potassium (K), and calcium (CA). Inset plot is the Miami monthly-mean aerosol concentration by species from 1989 through 1996, replotted from Prospero (1999a).

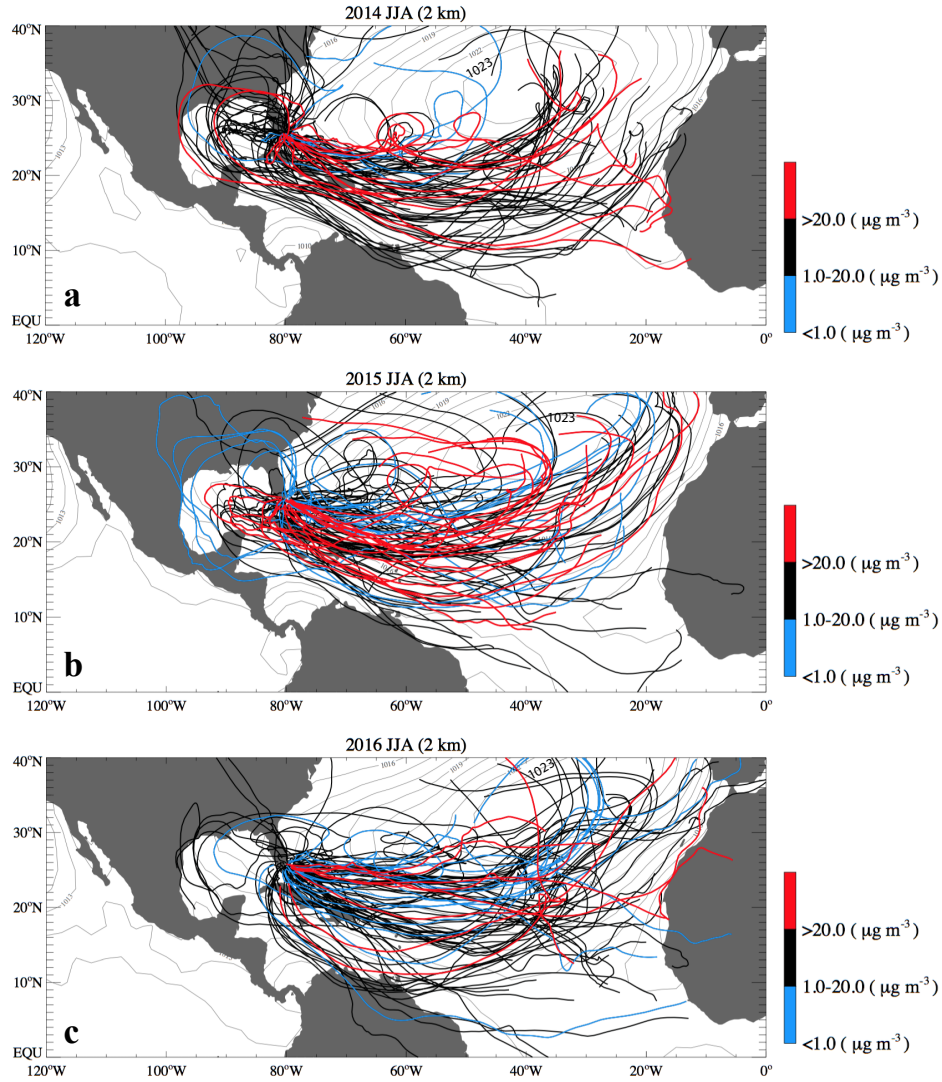


**Figure 2.** Daily bulk filter dust mass concentrations (black filled circles) and the sum of the impactor ~~filter-dust~~ mass concentrations (orange filled circles) for 20 July - 2 September, ~~2016-2016~~ (17 days total). No bulk mass concentration data are available for 4-9 August. Correlation coefficient of 0.98.

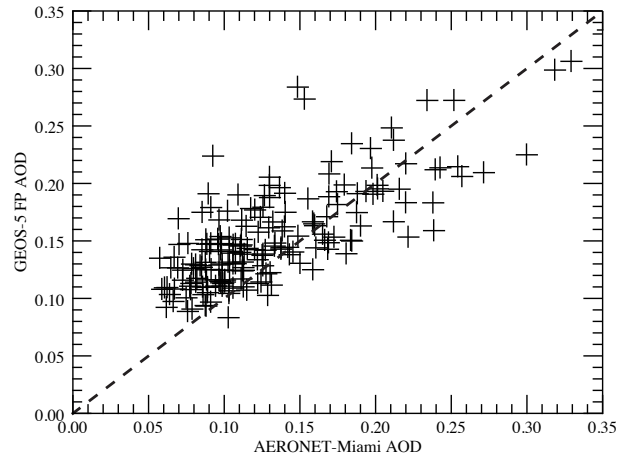




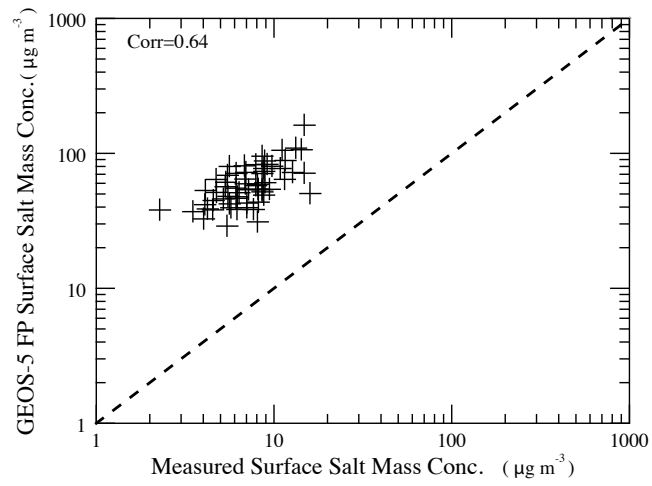
**Figure 3.** Time series of the MPL volume linear depolarization ratios for June-September, 2014, 2015, 2016, interspersed with the corresponding time series of the daily filter-retrieved bulk dust mass concentrations (units of  $\mu\text{g m}^{-3} \text{ day}^{-1}$ ). Subjectively-determined dates of the first and last days with significant dust is-are indicated within the lidar panel titles. Monthly-mean surface dust mass concentrations (units of  $\mu\text{g m}^{-3}$ ) are indicated for each month.



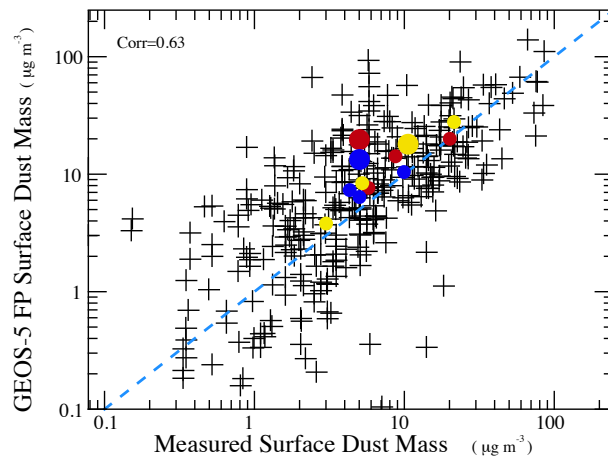
**Figure 4.** Daily HYSPLIT back trajectories for June-August of a) 2014, b) 2015, and c) 2016, extending for 300 hours and initialized at a height of 2 km above sea level, with the average seasonal NCEP Reanalysis sea level pressure (grey contours). Back trajectories are color-coded by the dust mass concentrations measured at Miami: initially exceeding  $> 20 \text{ } \mu\text{g m}^{-3}$  (red),  $1 - 20 \text{ } \mu\text{g m}^{-3}$  (dark blue), and  $< 1 \text{ } \mu\text{g m}^{-3}$  (light blue).



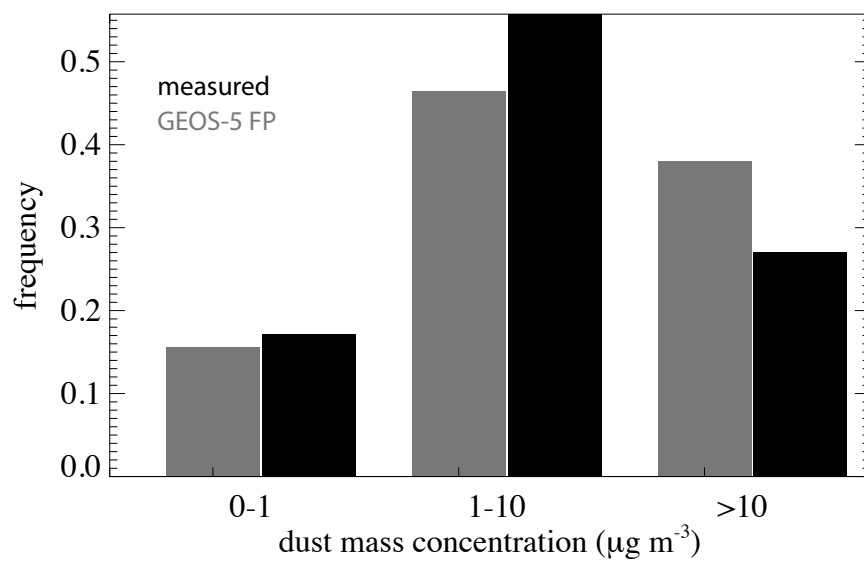
**Figure 5.** Daily-mean GEOS-5 FP aerosol optical depths  $\tau_{550}$  versus AERONET-derived values  $\tau_{500}$  (Level 1.5 version 2) at 500 nm wavelength, for June-September 2015 and 2016. Correlation coefficient of 0.80.



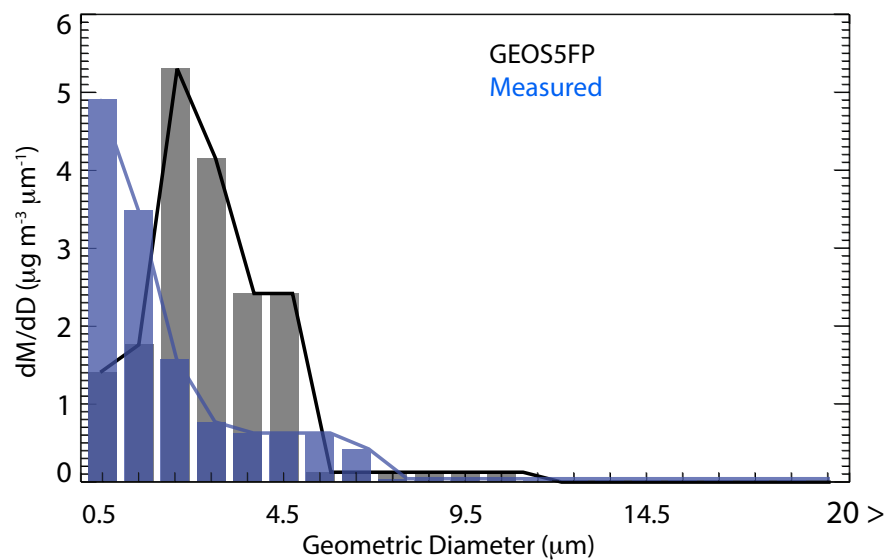
**Figure 6.** Daily-mean GEOS-5 FP versus measured near-surface salt mass concentrations for July-August, 2014 (32 days). The measured sea salt concentration is computed from the sodium concentration multiplied by 3.256, following Prospero (1999a).



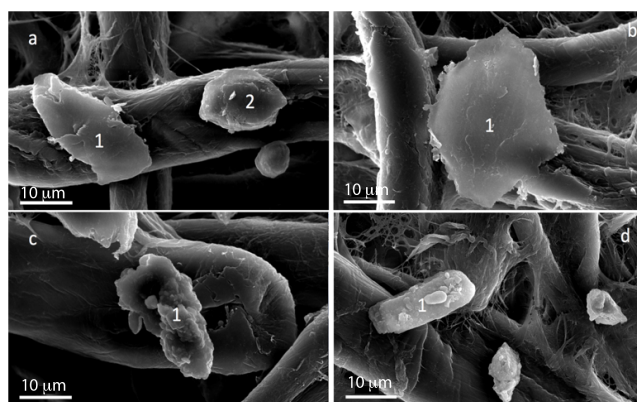
**Figure 7.** GEOS-5 FP daily versus measured near-surface dust mass concentrations for June-September from 2014-2016 (366 days), with monthly-mean values superimposed for 2014 (dark red), 2015 (dark yellow) and 2016 (blue); a larger filled circle highlights June.  $y=x$  line indicated. Correlation [\(in logarithmic space\)](#) of 0.63.



**Figure 8.** Frequency of days with measured (black) and model (grey) daily-mean dust mass concentrations in three ~~diameter~~diameter bins, based on 366 days from June-September, 2014-2016.

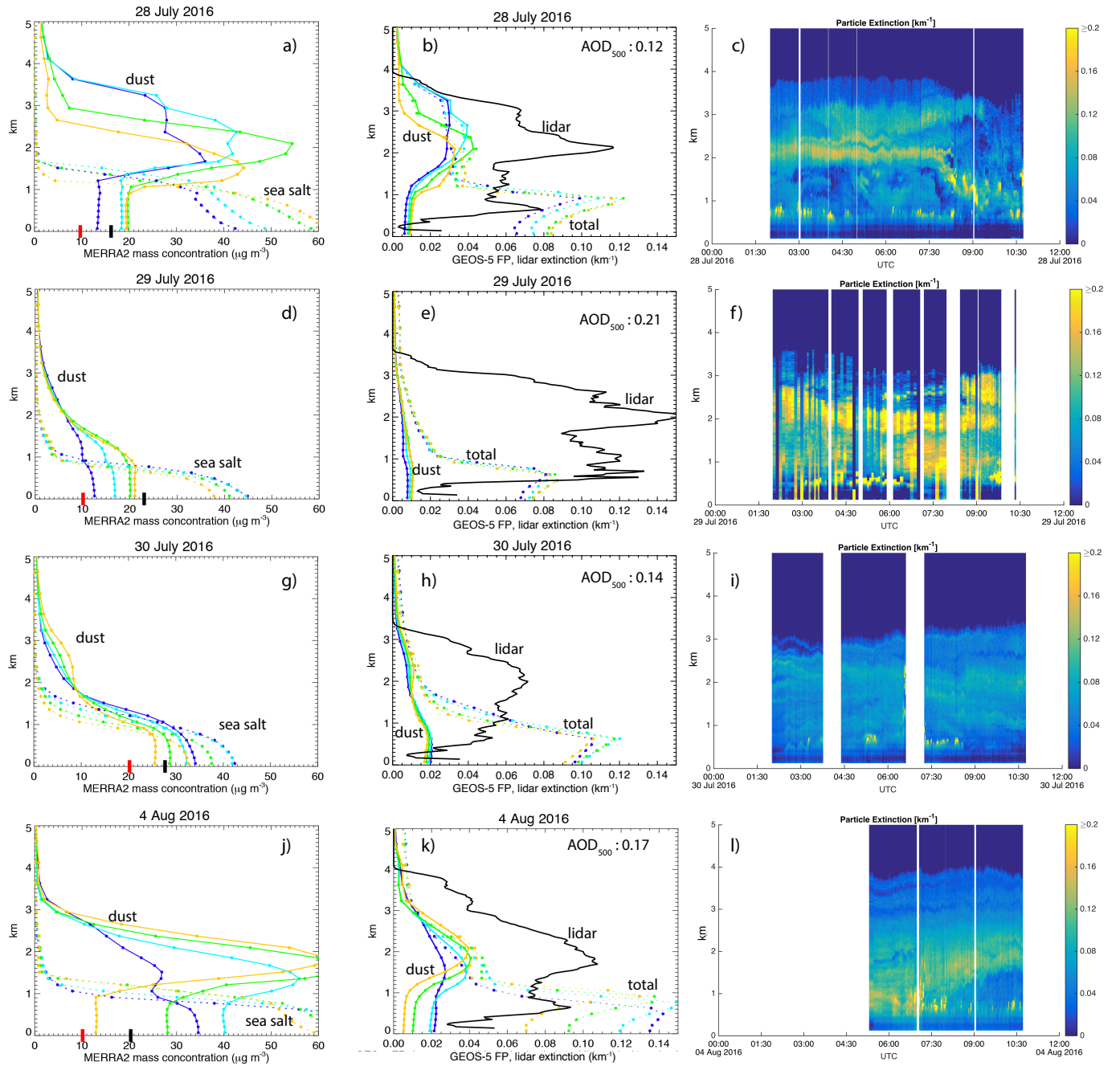


**Figure 9.** a) Daily-mean-17-day-average dust mass concentration over each diameter bin for each of the 17 sample days in measurements and b) GEOS-5 FP per (black one  $\mu\text{m}$ ); c) 17-day-average dust mass concentration for each  $1\text{-}\mu\text{m}$  diameter bin from measurements (blue) and GEOS-5 FP (black). Note that GEOS-5 FP and the measurements fall within separate diameter bins (see Table 1).

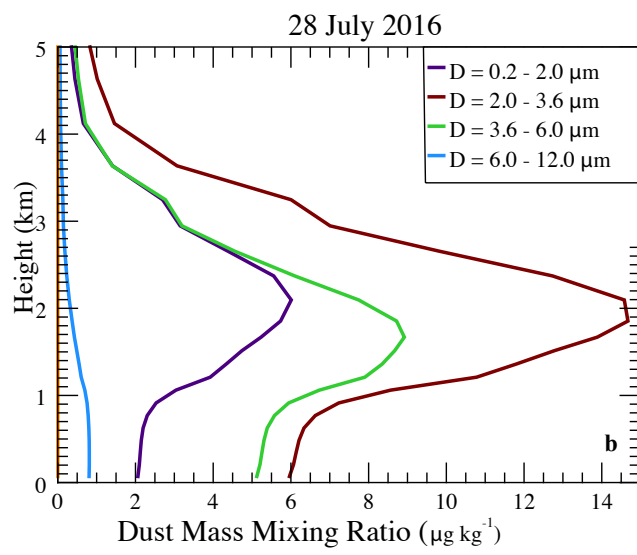


**Figure 10.** Scanning electron microscopy images of filter samples from a) and b) August 1, 2016, and c) and d) August 2, 2016. Individual particle dimensions are shown in Table 2.





**Figure 11.** a) MERRA-2 dust (solid) and sea salt (dashed) mass concentrations for 28 July 2016 3-12 UTC (dark blue, light blue, green and yellow lines indicate 3, 6, 9, and 12 UTC respectively), with surface model (black) and impactor (red) total dust mass concentrations included as thick vertical lines. b) GEOS-5 FP dust (solid) and total (dashed) volume extinction coefficients for 28 July 2016 3-12 UTC (same labeling scheme as in a)) and the lidar-derived volume extinction coefficient profile averaged over cloud-free portions (determined visually from camera imagery) of 28 July 2-11 UTC (black). The average 11-13 UTC AERONET-derived aerosol optical depth at 500 nm is indicated. c) 28 July 2011 UTC lidar-derived volume extinction coefficient time series. d)-f) same as a)-c) but for 29 July 2016. g)-i) same as a)-c) but for 30 July 2016. j)-l) same as a)-c) but for 5 August 2016. 01:12 UTC corresponds to 19-7 local time.



**Figure 12.** Size-resolved GEOS-5 FP dust mass mixing ratio vertical profiles for 28 July, 2016.