1	REPLY TO REVIEWER #1'S REMARKS
2 3	on the manuscript acp-2014-540: "Meridional distribution of aerosol optical thickness over
4	the tropical Atlantic Ocean"
5 6	by P. Kishcha, A.M. da Silva, B. Starobinets, C.N. Long, O. Kalashnikova, and P. Alpert.
7	We would like to thank Reviewer #1 for his valuable remarks. These remarks have been taken
8 9	into account in the revised manuscript, as listed below.
10	"Anonymous Referee #1
11	Received and published: 17 September 2014
12	
13	This paper analyzes satellite products and assimilated data sets over the tropical Atlantic to explore
14 15	the meridional distribution of aerosol, cloud and rainfall products. The paper is well-written. Previous literature is well-referenced. By using MERRAero data for their aerosol, which is satellite-assisted
16	model results, the authors can distinguish between different aerosol types, including dust and OC/BC
17	that is associated with biomass burning smoke in their area of study. This is a nice capability not
18	directly available from satellite products. The MERRAero total aerosol optical depth is compared with
19	MISR products and looks really, really good. The cloud fraction data are taken directly from MODIS
20	aerosol products, as are the rainfall products taken directly from TRMM.
21	
22	The authors use their data set to identify both temporal and spatial associations between total aerosol,
23 24	aerosol types, cloud fraction and precipitation. They show that some of the asymmetry in cloud fraction between northern and southern hemispheres is linked to the migration of the ITCZ and its
24	hemispherical asymmetry. However, they note a spatial association between the location of the dust
26	and maximums in the cloud fraction. The authors imply, but do not state in their conclusions, that
27	increasing cloud fraction is due, in part, to increased aerosol loading.
28	
29	From my perspective, these results are interesting without being important. Without a conclusion that
30	discusses the possible reason(s) for these observed associations, the paper adds little value for the
31	community.
32	
33	Our main point is that, over the tropical Atlantic, not only is Saharan dust responsible for the
34	hemispheric aerosol asymmetry, but it also contributes to significant cloud fraction along the
35	Saharan Air Layer. This significant cloud fraction in the area of SAL together with clouds
36	over the Atlantic Inter-tropical Convergence Zone contributes to hemispheric asymmetry in
37	cloud cover over the tropical Atlantic. This could lead to the hemispheric imbalance in strong
38	solar radiation reaching the sea surface over the tropical Atlantic. The phenomenon of
39 40	significant CF along SAL is important to the community and has not been reported so far, to
40	our knowledge.
41 42	In accordance with the Reviewer's remarks, in the revised version, new Sections 4.4.2 and
42 43	4.5.3 have been added (Pages $11 - 13$), where we discuss physical mechanisms for the
43 44	formation of significant CF along SAL. Both the Abstract and the title have been updated
45	(Pages 1 - 2).
45 46	(1 ag co 1 - 2).
40 47	"Discussing possible reasons for these associations is not easy. My first thought is that the MODIS
48	Cloud Fraction is contaminated by heavy aerosol loading and that is why the associations are so
49	strong. There are several different cloud fraction products available from MODIS. I believe the
50	authors are using the cloud fraction that is derived from the standard cloud mask (MOD35). The

authors are using the cloud fraction that is derived from the standard cloud mask (MOD35). The purpose of this mask is to identify clouds, but to err on the side of preventing "leakage". It may very well call heavy aerosol "cloudy." 51 52

2 We appreciate this Reviewer's comment which led us to clarify this point in the revised 3 version (the new Section 4.5.3, Page 13).

Indeed, Collection 5 of MODIS-Terra monthly daytime cloud fraction data used are derived 4

5 from the standard cloud mask product based on the cloud mask algorithm MOD35 (Ackerman 6 et al., 1998, Frey et al., 2008). In heavy dust loading situations, such as dust storms over 7 deserts, MOD35 may flag the aerosol-laden atmosphere as cloudy (Ackerman et al., 1998).

8

During dust storms over deserts, observed AOT values range from 2 to 5 (e.g. Alam et al., 2014). However, over the tropical North Atlantic, strong AOT exceeding even 1 is a very rare 9 10 phenomenon. To demonstrate that AOT exceeding 1 is a rare phenomenon over the tropical North Atlantic, Fig. 12a represents a histogram of AOT observed over the tropical North 11 Atlantic in July, 2010, based on MODIS Level 3 AOT daily data. July 2010 was chosen 12 13 because AOT, averaged over the tropical North Atlantic, was maximal compared to AOT in 14 other July months, during the 10-year study period. One can see that AOT hardly exceed 1. A 15 similar situation can be seen over the latitudes with SAL presence $(12^{\circ}N - 24^{\circ}N)$ (Fig. 12b). 16 Therefore, the effect of MODIS cloud fraction contamination by heavy dust loading cannot essentially contribute to averaging CF over the tropical North Atlantic. Consequently, given 17 the large amount of available MODIS CF daily data over the 10-year study period, cloud 18 19 fraction contamination does not account for the obtained hemispheric CF asymmetry.

20

21 "There are other possible reasons for the observed associations, including some that are physical 22 mechanisms. However, it would surprise me if cloud fraction is increasing through the Albrecht 23 (1989) mechanism. Here in this study the authors find the strongest relationship between aerosol and 24 cloud fraction when the aerosol is very thick. We know to expect the strongest relationships when 25 clouds are starved for CCN, when aerosol goes from pristine to moderately loaded (Koren et al., 26 2008, 2014). A physical possibility may be a pathway that involves ice nuclei and ice processes. All 27 we see here is cloud fraction, so we don't know if the association between aerosol and clouds affects 28 water clouds, ice clouds or both. "

29

30 We agree with the Reviewer that the Albrecht (1989) mechanism is not suitable for the SAL 31 area with heavy dust loading.

32 As discussed in the revised version (Section 4.5.3, Pages 11 - 13), we consider that the most

33 likely physical mechanism for the formation of significant cloud cover along SAL is as 34 follows:

35 The observed temperature inversion over zones 1 - 4 prevents deep cloud formation; this explains limited precipitation in these zones. On the other hand, meteorological conditions 36

37 below the temperature inversion at the SAL base include significant atmospheric humidity 38 and the presence of large amounts of settling dust particles together with marine aerosols.

39 As known, aerosol species often combine to form mixed particles, with properties different

40 from those of their components (Andreae et al., 2009). Mineral dust particles are known to be 41 not very efficient cloud condensation nuclei (CCN), unless they are coated with soluble 42 materials (Andreae et al., 2009). Using airplane measurements, Levin et al. (2005) showed 43 that dust transport over the sea could lead to sea-salt coating on dust particles. Coating settling 44 dust particles with sea-salt could modify them into efficient CCN. Being below the temperature inversion and acting as efficient CCN, Saharan dust particles coated with soluble 45 46 material contribute to the formation of shallow stratocumulus clouds. This physical 47 mechanism, based on the indirect effect of Saharan dust on stratocumulus clouds below the 48 temperature inversion, could explain the observed significant cloud cover (CF up to 0.8 - 0.9) 49 along the Saharan Air Layer. The significant cloud fraction along SAL contributes to

hemispheric CF asymmetry over the tropical Atlantic. This could lead to hemispheric
 imbalance in strong solar radiation reaching the sea surface in the tropical Atlantic Ocean.

3 To examine the properties of clouds in the area of SAL, we analyzed available data on the

4 effective radius of cloud droplets. Fig. 11 represents histograms of the effective radius (Reff)

5 of cloud droplets for liquid water clouds in the specified zones 1 - 4 along SAL, based on

6 MODIS L3 gridded monthly data $(1^{\circ} x 1^{\circ})$ during the 10-year study period in July. The data

7 were supplied by the Giovanni data base. It is obvious that the cloud droplet effective radius

8 increases from zone 1 to zone 4 (Fig. 11). One can see a systematic shift in the whole 9 histogram to higher values of Reff from zone 1 to zone 4. This can be explained by the

10 decrease in CCN numbers associated with the decreasing numbers of settling Saharan dust

11 particles with distance from the Sahara, in accordance with the decrease in dust AOT shown

12 in Fig. 9a.

Thus, the cloud droplet effective radius in zone 4 was larger than in zones 1 - 3. This could lead to some increase in precipitation in zone 4. Indeed, as shown in Fig. 9b, TRMM accumulated rainfall in zone 4 was more intensive than over zones 1 - 3. This supports the above-mentioned physical mechanism of cloud formation below the temperature inversion at the SAL base.

18

19 "When the aerosol is very thick it should create a temperature signal in the vertical temperature 20 profiles because both dust and OC/BC are absorbing aerosols that absorb sunlight and heat the 21 column (Alpert et al., 1998; Ackerman et al., 2000; Koren et al. 2008). The more aerosol, the greater 22 the amount of heating. Most studies associate this heating with a suppression of cloudiness, so that 23 there should be a negative correlation between aerosol loading and cloud fraction. I know of one 24 paper by Johnson et al. (2004), that finds a physical mechanism that can explain increased cloudiness, 25 due to the heating, in this region. There may be others."

26

Saharan dust particles can influence the vertical temperature profile in the atmosphere directly by absorbing solar radiation. Aerosol absorption by Saharan dust may decrease cloud cover by heating the air and reducing relative humidity (the semi-direct aerosol effect) (Johnson et al., 2004, Kaufman et al., 2005b). So that there should be a negative correlation between dust loading and cloud fraction along SAL. However, this is not the case: both dust AOT and CF decrease with the distance from the Sahara (Figs. 8b and 9b).

33

Anyway there are several possible reasons why we see these associations, some because of artifacts or co-varying meteorology, and some perhaps because aerosols are influencing the clouds through microphysics or by changing the environment in which the clouds evolve. Simply discussing all the possibilities, as I have done here, still only lifts the paper to the mediocre level. Some attempt should be made to PROVE which possibility is the most likely."

39 There is nothing WRONG with the analysis presented in this paper, but I question whether simply

40 presenting the results of the analysis with no conclusion is worth publication. I do not recommend 41 publishing this paper in this form.

42

We do not agree that significant CF along SAL is an artifact. As mentioned in our reply to the previous Reviewer's comment on this topic, the effect of MODIS cloud contamination by heavy dust loading does not account for meridional CF asymmetry over the tropical Atlantic (see a new section 4.5.3, Page 13). Therefore, the hemispheric 20% CF asymmetry found in the summer months is a real phenomenon. The hemispheric CF asymmetry could lead to the

48 hemispheric imbalance in surface solar radiation over the tropical Atlantic. We feel that 49 reporting important findings is worthy of publication. In the revised version (a new section

4.5.2, Pages 11 - 13) we discuss the most likely physical mechanism for the formation of 2 significant cloud cover along SAL in July. 3

4 Minor comments:

My comments are linked to the web-based (non printer friendly) version of the paper.

5 6 7 p. 23312 lines 17-23. Why does the meridional distribution of aerosol and cloud fraction matter? 8 Nowhere in the introduction do the authors explain their motivation for doing this study. 9

10 Hemispheric asymmetry in cloud fraction (CF) and aerosols could lead to hemispheric imbalance in surface solar radiation. Consequently, analyzing hemispheric asymmetry in CF 11 12 and aerosols is essential for understanding climate formation and its changes. Previous studies 13 showed that, over the global ocean, there is hemispheric asymmetry in aerosols and no 14 noticeable asymmetry in cloud fraction (CF). This contributes to the hemispheric balance in 15 solar radiation reaching the sea surface. We chose the tropical Atlantic because it is 16 characterized by significant amounts of Saharan dust dominating other aerosol species over 17 the North Atlantic. We wished to find out if the meridional CF distribution remains 18 symmetrical in the presence of such strong hemispheric aerosol asymmetry. This explains our 19 motivation for doing the current study (Page 3, lines 22 - 25).

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21 P23313 line22 and throughout the manuscript. The authors use ten yr to denote ten-year or 10-year 22 without explaining what yr stands for. The journal style book may disagree with me, but I feel that ten-23 year or 10-year is better.

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25 This was determined by the production editor, in accordance with the ACP journal style. In 26 our original version, we used the same expressions as suggested by the Reviewer.

28 P23314 lines 23-25. Reference Frey et al. (2008) or Ackerman et al. (1998) so that we 29 know that this is derived from MOD35.

31 Done. (Page 5, line 21)

33 *P23316 line 15. Here we have 10yr, which I really prefer to be 10-year.*

- 35 See our reply to the previous similar remark.
- 37 P23316 line 16. "much more" should be "many more"
- 39 Done. (Page 7, line 2)

40

41 P23321 lines 15-16. Do the authors know for sure that it is gravitational settling? Could it not be wet 42 deposition? All that implied aerosol-cloud interaction and I would expect that a lot of aerosol is 43 leaving the atmosphere because of washing out in the rain.

44

45 Indeed, rainfall always removes aerosols. However, as shown in Fig. 9, a strong decrease in 46 dust AOT from zone 1 to zone 3 was not accompanied by any changes in TRMM 47 accumulated rainfall. Therefore, the washing out of aerosols by rainfall does not account for 48 the aerosol spatial decrease with distance from the Sahara. It proves that gravitational settling 49 of Saharan dust particles accounts for this dust spatial decrease with distance from the Sahara.

(Page 11, lines 9 – 14). 50

1 2 "Figures 9 and 10, and associated discussion in the text. You lose the inversion as you travel from 3 zone 1 to zone 6. Meanwhile the convection intensifies. The authors see this as cause and effect, which 4 I'm willing to accept. However, the cloudiness decreases. More convection, but less cloud cover. 5 Earlier in the paper the authors noted that in the north-south direction the peak in cloudiness 6 corresponded to the peak in convection, but here in the east-west direction the opposite is true. This 7 only points out that the full story is still hidden.

8

9 The Reviewer should note that, in the current study, we focus on the relationship between 10 temperature inversion and convection over the area of SAL, and not over the Atlantic Inter-11 tropical convergence zone.

Let us start with the east-west direction. We lose inversion over zone 5 and 6. Significant precipitation in zones 5 and 6 (up to 110 mm month⁻¹) indicates the presence of developed clouds which are characterized by increased cloud thickness. These clouds over zones 5 and 6

15 differ from shallow stratocumulus clouds created under the temperature inversion in zones 1 - 4. The thick developed clouds over zones 5 and 6 may create CF lower than that created by

17 the shallow stratocumulus clouds over zones 1 - 4.

18 In the north-south direction, the first peak in CF corresponds to the peak in convection in the 19 Atlantic Inter-tropical convergence zone centered at approximately 8° N in July (Fig. 7).

20 However, the second peak in CF is seen over the SAL zone centered at approximately 18°N

21 (Fig. 7). The first CF peak (centered at 8° N) is created over the area with maximal convection,

while the second CF peak (centered at 18°N) is created over the area with minimal convection because of the presence of temperature inversion. Contrary to the Reviewer's opinion, we

found that, in the north – south direction over the SAL area, there is no direct relationship

- 25 between CF and convection.
- 26

27 "How are cloud types and morphology changing? How do these changes associate with the different
28 types of aerosol?"

29

There are different cloud types over zones 1 - 4 on the one hand, and over zones 5 - 6 on the other hand. Over zones 1 - 4, we consider the presence of shallow stratocumulus clouds below the temperature inversion at the SAL base. These shallow stratocumulus clouds are characterized by limited precipitation. Over zones 5 - 6, we consider the presence of developed clouds capable of producing strong precipitation up to 110 mm month⁻¹.

35

36 "Is there a possibility that the aerosol absorption is contributing to the strength of the inversion? Lots
37 and lots of questions that should be answered or at least partly answered."

38

The answer is in the affirmative. As mentioned in the manuscript, Saharan dust travels across the Atlantic Ocean within the hot and dry Saharan Air Layer with temperature inversion at the SAL base. In addition, the direct radiation effect of dust aerosols (i.e. absorption of solar radiation) could contribute to the strength of inversion. As seen in Fig. 10, the temperature inversion over zone 1 (in the presence of a large amount of dust) is stronger than over zone 4

- 44 where the amount of dust is lower than over zone 1.
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Hemispheric asymmetry in aerosol optical thickness and cloud fraction over the tropical Atlantic Ocean

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13

14 Abstract

15 Previous studies showed that, over the global ocean, there is no noticeable hemispheric 16 asymmetry in cloud fraction (CF). This contributes to the balance in solar radiation reaching 17 the sea surface in the Northern and Southern hemispheres. In the current study, we focus on 18 the tropical Atlantic $(30^{\circ}N - 30^{\circ}S)$ which is characterized by significant amounts of Saharan 19 dust dominating other aerosol species over the North Atlantic. Our main point is that, over the 20 tropical Atlantic, not only is Saharan dust responsible for the pronounced hemispheric aerosol 21 asymmetry, but it also contributes to significant cloud cover along the Saharan Air Layer. 22 This could lead to the hemispheric imbalance in strong solar radiation reaching the sea surface 23 in the tropical Atlantic. During the 10-year study period (July 2002 – June 2012), NASA 24 Aerosol Reanalysis (aka MERRAero) showed that, when the hemispheric asymmetry in dust 25 aerosol optical thickness (AOT) was the most pronounced (particularly in July), dust AOT averaged separately over the tropical North Atlantic was one order of magnitude higher than 26 27 that averaged over the tropical South Atlantic. In the presence of such strong hemispheric asymmetry in dust AOT in July, CF averaged separately over the tropical North Atlantic 28

exceeded that over the tropical South Atlantic by 20%. In July, along the Saharan Air Layer, 1 2 Moderate Resolution Imaging Spectroradiometer (MODIS) CF data showed significant cloud cover (up to 0.8 - 0.9). This significant cloud fraction along SAL together with clouds over 3 4 the Atlantic Inter-tropical Convergence Zone contributes to the above-mentioned hemispheric 5 CF asymmetry. Both Multi-Angle Imaging SpectroRadiometer (MISR) measurements and MERRAero data were in agreement on seasonal variations in hemispheric aerosol asymmetry. 6 7 Hemispheric asymmetry in total AOT over the Atlantic was the most pronounced between 8 March and July, when dust presence over the North Atlantic was maximal. In September and 9 October, there was no noticeable hemispheric asymmetry in total AOT over the tropical 10 Atlantic.

11

12 **1** Introduction

13 Hemispheric asymmetry in cloud fraction (CF) and aerosols could lead to hemispheric 14 imbalance in solar radiation reaching the surface and, consequently, could affect the Earth's 15 climate. Satellite observations have been widely used in the study of aerosol optical thickness 16 and cloud cover because of their capability of providing global coverage on a regular basis. 17 Previous studies, using different space-borne aerosol sensors, discussed the idea that the 18 hemispheres are asymmetric in aerosol distribution (Remer et al., 2008; Kaufman et al., 19 2005a; Remer and Kaufman, 2006, Mishchenko and Geogdzhavev, 2007, Chou et al., 2002, 20 Zhang and Reid, 2010, Hsu et al., 2012, Kishcha et al, 2007, 2009). The Advanced Very High Resolution Radiometer (AVHRR) satellite data over the ocean were used by Mishchenko and 21 22 Geogdzhayev (2007) to compare monthly averaged aerosol optical thickness (AOT) over the 23 Northern and Southern Hemispheres. They found a difference in AOT averaged over the two 24 hemispheres. Chou et al. (2002) obtained meridional distribution of AOT over the ocean by using the Sea-viewing Wide Field-of-view Sensor (SeaWIFS) satellite data for the year 1998. 25 26 Hsu et al. (2012) displayed the asymmetric spatial distribution of seasonally-averaged 27 SeaWiFS AOT from 1997 to 2010. Several studies based on the Moderate Resolution 28 Imaging Spectroradiometer (MODIS) and Multi-Angle Imaging SpectroRadiometer (MISR) 29 data showed that aerosol parameters are distributed asymmetrically on the two hemispheres 30 (Remer et al., 2008; Kaufman et al., 2005a; Remer and Kaufman, 2006, Zhang and Reid, 2010, Kishcha et al., 2007, 2009). In our previous study (Kishcha et al., 2009), AOT data 31 32 from three satellite sensors (MISR, MODIS-Terra, and MODIS-Aqua) were used in order to

1 analyze seasonal variations of meridional AOT asymmetry over the global ocean. The 2 asymmetry was pronounced in the April-July months, while there was no noticeable asymmetry during the season from September to December. Kishcha et al. (2009) mentioned 3 that not only the Northern Hemisphere but also the Southern Hemisphere contributed to the 4 5 formation of noticeable hemispheric aerosol asymmetry. During the season of pronounced hemispheric aerosol asymmetry, an increase in AOT was observed over the Northern 6 7 Hemisphere, while a decrease in AOT was observed over the Southern Hemisphere. It was 8 found that, over the global ocean, there was no noticeable asymmetry in meridional 9 distribution of cloud fraction.

10 The Sahara desert emits dust in large quantities over the tropical Atlantic (Prospero and 11 Lamb, 2003). Previous studies have shown that desert dust particles can influence the Earth's 12 atmosphere in the following ways: directly by scattering and absorbing solar and thermal 13 radiation, and indirectly by acting as cloud and ice condensation nuclei (Choobari et al, 2013 14 and references therein, Pey et al., 2013). It was shown by Wilcox et al. (2010) that the 15 radiative effect of Saharan dust tends to draw the Atlantic Intertropical Convergence Zone 16 (ITCZ) northward toward the Saharan Air Layer (SAL). Alpert et al. (1998) discussed the 17 response of the atmospheric temperature field to the radiative forcing of Saharan dust over the 18 North Atlantic Ocean. Dust particles over the Atlantic Ocean may essentially influence 19 tropical cloud systems and precipitation (Kaufman et al., 2005b, Johnson et al., 2004, Min et 20 al., 2009, Ben-Ami et al., 2009, Feingold et al., 2009, Rosenfeld et al., 2001).

21 To our knowledge, over a limited ocean area, hemispheric asymmetry of aerosols and cloud 22 fraction relative to the equator has not been investigated so far. We chose the tropical Atlantic 23 $(30^{\circ}N - 30^{\circ}S)$ because it is characterized by significant amounts of Saharan dust. We wished 24 to find out if the meridional CF distribution remains symmetrical relative to the equator in the 25 presence of such strong hemispheric aerosol asymmetry. We determined and compared the contribution of desert dust and that of other aerosol species to aerosol asymmetry between the 26 27 tropical North and South Atlantic Oceans. Analyzing the meridional distribution of various aerosol species over the tropical Atlantic Ocean was carried out using the NASA Aerosol 28 Reanalysis (aka MERRAero). This reanalysis has been recently developed at NASA's Global 29 Modeling Assimilation Office (GMAO) using a version of the NASA Goddard Earth 30 31 Observing System-5 (GEOS-5) model radiatively coupled with Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) aerosols. An important property of GEOS-5 is data 32

assimilation inclusion of bias-corrected aerosol optical thickness from the MODIS sensor on
both Terra and Aqua satellites. Of course, AOT assimilation is effective only for two short
periods of MODIS's appearance over the study area. All other time (18 hours per day) the
GEOS-5 model works independently of MODIS (Kishcha et al., 2014).

5

6 2 GEOS-5 and the MERRA Aerosol Reanalysis (MERRAero)

7 GEOS-5 is the latest version of the NASA Global Modeling and Assimilation Office 8 (GMAO) Earth system model, which was used to extend the NASA Modern Era-9 Retrospective Analysis for Research and Applications (MERRA) with five atmospheric aerosol components (sulfates, organic carbon, black carbon, desert dust, and sea-salt). GEOS-10 11 5 includes aerosols based on a version of the Goddard Chemistry, Aerosol, Radiation, and 12 Transport (GOCART) model (Colarco et al., 2010, Chin et al., 2002). Both dust and sea salt 13 have wind-speed dependent emission functions (Colarco et al., 2010), while sulfate and carbonaceous species have emissions principally from fossil fuel combustion, biomass 14 15 burning, and bio-fuel consumption, with additional biogenic sources of organic carbon. 16 Sulfate has additional chemical production from oxidation of SO2 and dimethylsulfide 17 (DMS), as well as volcanic SO2 emissions. Aerosol emissions for sulfate and carbonaceous 18 species are based on the AeroCom version 2 hindcast inventories 19 [http://aerocom.met.no/emissions.html]. Daily biomass burning emissions are from the Quick 20 Fire Emission Dataset (QFED) and are derived from MODIS fire radiative power retrievals 21 (Darmenov and da Silva, 2013). GEOS-5 also includes assimilation of AOT observations 22 from the MODIS sensor on both Terra and Aqua satellites. The obtained ten-year (July 2002 -23 June 2012) MERRA-driven aerosol reanalysis (MERRAero) dataset was applied to the 24 analysis of hemispheric aerosol asymmetry in the current study. In order to verify the 25 obtained meridional aerosol distribution based on MERRAero, we used the Multi-angle Imaging SpectroRadiometer (MISR) monthly global 0.5° x 0.5° AOT dataset available over 26 27 the study period.

28

29 **3 Method**

30 Over the tropical Atlantic Ocean $(30^{\circ}N - 30^{\circ}S)$, variations of zonal-averaged AOT as a 31 function of latitude were used to analyze meridional aerosol distribution, following our 32 previous study (Kishcha et al., 2009). This included total AOT and AOT of various aerosol species. To quantify hemispheric AOT asymmetry, the hemispheric ratio (R) of AOT averaged separately over the tropical North Atlantic (X_N) to that over the tropical South Atlantic (X_S) was estimated. The hemispheric ratio is equal to 1 in the case of the two parts of the tropical Atlantic holding approximately the same averaged AOT, while the ratio is greater (less) than 1 if the North (South) Atlantic dominates the other one. Standard deviation of the reported hemispheric ratio (Table 1) was estimated in accordance with the following formula by Ku [1966], NIST/SEMATECH (2006):

$$C_{R} = \frac{1}{\sqrt{N}} \cdot \frac{X_{N}}{X_{S}} \cdot \sqrt{\frac{C_{N}^{2}}{X_{N}^{2}}} + \frac{C_{S}^{2}}{X_{S}^{2}} - 2 \cdot \frac{C_{NS}^{2}}{X_{N} \cdot X_{S}}$$

8

9 where C_N , C_S are standard deviations of zonal averaged AOTs in the tropical North and 10 South Atlantic Oceans respectively, C_{NS} is their covariance, and N = 120 stands for the 11 number of months in the MISR/ MERRAero AOT monthly data set used.

Variations of meridional aerosol distributions were analyzed by using Version 3.1 of MISR 12 13 Level 3 AOT measurements and MERRAero data during the 10-year period, from July 2002 to June 2012. The MISR swath width is about 380 km and global coverage is obtained every 9 14 15 days. MISR AOT has been extensively validated against Aerosol Robotic Network (AERONET) Sun photometer measurements over different regions (Martonchik et al., 2004; 16 17 Christopher and Wang, 2004; Kalashnikova and Kahn, 2008; Liu et al., 2004). For the 18 purpose of comparing meridional distributions of cloud cover with those of AOT during the same 10-year period (July 2002 - June 2012), Collection 5.1 of MODIS-Terra Level 3 19 20 monthly daytime cloud fraction (CF) data, with horizontal resolution 1° x 1° was used (Ackerman et al., 1998, Frey et al., 2008, King et al., 2003). Furthermore, to analyze 21 22 meridional rainfall distribution, the Tropical Rainfall Measuring Mission (TRMM) monthly 0.25° x 0.25° Rainfall Data Product (3B43 Version 7) was used (Huffman et al., 2007). 23 24 MODIS CF data and TRMM data were acquired using the GES-DISC Interactive Online Visualization and Analysis Infrastructure (Giovanni) as part of NASA Goddard Earth 25 Sciences (GES) Data and Information Services Center (DISC) (Acker and Leptoukh, 2007). 26

1 4 Results

2 4.1 Ocean zone with the predominance of desert dust aerosols

3 MERRAero showed that the Sahara desert emits a significant amount of dust into the 4 atmosphere over the Atlantic Ocean (Fig. 1, a, c, and e). With respect to different oceans, 5 MERRAero showed that desert dust dominates all other aerosol species only over the Atlantic 6 Ocean. Fig. 1 (b, d, and f) represents spatial distribution of the ratio of dust AOT to AOT of 7 all other aerosol species. The red contour lines represent the boundary of the zone where dust 8 AOT is equal to AOT of all other aerosol species. One can see that, through the 10-year 9 period under consideration, over the Atlantic Ocean within the latitudinal zone between 7°N 10 and 30°N, Saharan dust dominates other aerosol species (Fig. 1b). The longitudinal dimension of this zone is subject to seasonal variability. During the dusty season from March to July, the 11 12 zone of dust predominance occupies a significant part of the tropical Atlantic between North Africa and Central America. Specifically, as shown in Fig. 1d, in July, the zone of dust 13 14 predominance is extremely extensive. By contrast, from October to February, this zone is observed only over some limited territory close to North Africa. 15

Desert dust can be seen not only over the Atlantic Ocean, but also over the Pacific and Indian Oceans (Fig. 1, a, c, and e). However, outside the Atlantic Ocean, one can see only limited zones of desert dust predominance over the Mediterranean Sea and over the Arabian Sea (Fig. 1, b, d, and f). Therefore, the tropical North Atlantic Ocean is the largest ocean area where dust particles determine the atmospheric aerosol content, based on MERRAero data.

21

4.2 Meridional distribution of total AOT over the tropical Atlantic Ocean

23 Figure 2a represents meridional distribution of ten-year mean AOT (July 2002 – June 2012), zonal averaged over the tropical Atlantic Ocean. One can see that MERRAero showed 24 25 similarity to the meridional AOT distribution, based on MISR data (Fig. 2a). Specifically, MERRAero was able to reproduce the hemispheric asymmetry in the AOT distribution, 26 27 including a monomodal maximum in the tropical North Atlantic and a minimum in the tropical South Atlantic. This monomodal AOT maximum was discussed in our previous study 28 29 (Kishcha et al., 2009). Both MISR and MERRAero showed that, in the minimum, the AOT values were three times lower than those in the maximum. We quantified meridional AOT 30 asymmetry relative to the equator in the tropical Atlantic Ocean $(30^{\circ}N - 30^{\circ}S)$ by obtaining 31 32 the hemispheric ratio (R_{AOT}) of AOT averaged separately over the tropical North Atlantic to AOT averaged over the tropical South Atlantic: R_{AOT} was estimated to be about 1.7 (Table 1).
 This means that, over the 10-year period under consideration, there were many more aerosol
 particles over the tropical North Atlantic than over the tropical South Atlantic.

4

5 4.3 Seasonal variations of meridional distribution of AOT

For each month of the year, we analyzed variations of meridional distribution of AOT over the tropical Atlantic Ocean (Fig. 3). It was found that the meridional AOT distribution is seasonal dependent. In particular, both MISR and MERRAero were in agreement that the monomodal AOT maximum, a characteristic feature of hemispheric asymmetry in AOT, exists but not in each month. In the months from September to October, two AOT maxima can be observed: one maximum in the North Atlantic, and another one in the South Atlantic.

Figure 4 represents month-to-month variations of the hemispheric ratio R_{AOT} over the tropical Atlantic for each month of the year. Both MISR and MERRAero showed that meridional AOT asymmetry was most pronounced during the season from March to July (Fig. 4). One can see that, from month to month during the year, R_{AOT} ranges from 1 to 2.4, while during the season of pronounced hemispheric aerosol asymmetry (March – July) R_{AOT} ranges from 2.0 – 2.4. In September and October, R_{AOT} was close to 1, indicating no noticeable asymmetry (Fig. 4).

19

20 4.4 Meridional distribution of AOT of various aerosol species

21 Fig. 2c represents meridional distribution of ten-year mean MERRAero AOT for total AOT 22 (Total), dust AOT (DU), organic and black carbon aerosol AOT (OC & BC), and AOT of 23 other aerosol species (Other), zonal averaged over the tropical Atlantic Ocean. One can see 24 that meridional dust distribution is much more asymmetric relative to the equator than 25 meridional distribution of OC & BC and other aerosol species. The hemispheric asymmetry of 26 DU, characterized by the hemispheric ratio (R_{DU}) of dust AOT was about 11 (Table 2). Such 27 strong asymmetry in meridional distribution of desert dust over the ocean can be explained by 28 its transport by winds from the Sahara desert to the ocean in the North Atlantic. Being the 29 major contributor to the AOT maximum in the North Atlantic, Saharan dust was responsible 30 for the pronounced meridional AOT asymmetry in total AOT over the tropical Atlantic 31 Ocean. Carbon aerosols also displayed some hemispheric asymmetry characterized by the 32 hemispheric ratio $R_{OC\&BC} = 0.7$, although this asymmetry was much less pronounced than that of desert dust (Fig. 2c and Table 2). Meridional distribution of AOT of other aerosol species was almost symmetrical (R_{Other} is 1.1) (Table 2). Therefore, aerosols over the tropical Atlantic can be divided into two groups with different meridional distribution relative to the equator: dust and carbonaceous aerosols were distributed asymmetrically, while other aerosol species were distributed more symmetrically.

6 MERRAero showed that seasonal variations of transatlantic Saharan dust transport 7 determined the seasonal variations of meridional dust asymmetry. In May - July, when 8 hemispheric asymmetry in dust AOT over the tropical North Atlantic was the most 9 pronounced, dust AOT averaged separately over the tropical North Atlantic was one order of 10 magnitude higher than dust AOT averaged over the tropical South Atlantic (Table 2). In July, 11 the most pronounced hemispheric asymmetry of dust AOT was characterized by the 12 hemispheric ratio R_{DU} of about 30 (Table 2).

13 When dust presence over the North Atlantic was minimal, the contribution of other aerosol 14 species to the meridional distribution of total AOT could be significant. In particular, in 15 December, the maximum in OC & BC at low-latitudes (due to the transport of bio-mass 16 burning smoke) contributed significantly to the maximum in total AOT in the tropical North 17 Atlantic (Fig. 5a). Note that the reason for the aforementioned transport of bio-mass burning 18 aerosols is the burning of agricultural waste in the Sahelian region of northern Africa. This burning activity is maximal during December – February (Haywood et al., 2008). MERRAero 19 20 showed that no noticeable hemispheric asymmetry of total AOT was observed in September 21 and October (Fig. 4). This is because the contribution of carbonaceous aerosols (OC & BC) to 22 total AOT over the South Atlantic is approximately equal to the contribution of Saharan dust 23 to total AOT in the North Atlantic (Fig. 5). The reason for the observed increase in OC & BC 24 over the South Atlantic in September and October is that these months fall within the burning 25 period in Central Africa, where slash-and-burn agriculture is prevalent (Tereszchuk et al., 2011). In September and October, AOT of carbonaceous aerosols over the tropical South 26 Atlantic was five times higher than that over the tropical North Atlantic ($R_{OC\&BC} = 0.2$) (Table 27 28 2).

- Meridional distribution of AOT of other aerosol species remains more symmetrical than dust and carbonaceous aerosols throughout all months (the hemispheric ratio R_{Other} ranged from 0.8 - 1.3) (Table 2). This group includes marine aerosols, such as sea-salt and dimethylsulfide
- 32 (DMS) aerosols, which are produced everywhere in the tropical Atlantic Ocean.

2 **4.5** Meridional distribution of cloud fraction

3 We analyzed meridional distribution of cloud cover over the tropical $(30^{\circ}N - 30^{\circ}S)$ Atlantic 4 Ocean, which includes the area of transatlantic Saharan dust transport within SAL. Fig. 2b represents the meridional distribution of 10-year mean cloud fraction, zonal averaged over the 5 6 Atlantic Ocean. One can see the local maximum near the equator due to clouds concentrated 7 over the Intertropical Convergence Zone: this maximum shifts to the north from the equator. 8 Despite this CF maximum, the hemispheric CF ratio (R_{CF}), characterized by the ratio of CF 9 averaged separately over the tropical North and over the South Atlantic, did not exceed 1.1 10 (Table 1).

11 As mentioned in Sect. 4.4, MERRAero showed that dust and carbonaceous aerosols were 12 distributed asymmetrically in relation to the equator, while other aerosol species were 13 distributed more symmetrically. During the period of pronounced meridional AOT 14 asymmetry over the tropical Atlantic from May - July, dust AOT averaged separately over the 15 tropical North Atlantic was about one order of magnitude higher than dust AOT averaged 16 over the tropical South Atlantic (Table 2). In July, the hemispheric ratio R_{DU} was roughly 30. 17 In the presence of such strong meridional dust asymmetry, in July, R_{CF} reached 1.2 (Table 2 18 and Fig. 4). As shown in previous study (Kishcha et al., 2009), over the global ocean, RAOT 19 was about 1.5, while R_{CF} was 1. Therefore, by contrast to the global ocean (where meridional 20 CF distribution was symmetrical over the two hemispheres), over the tropical Atlantic in July, 21 CF averaged separately over the tropical North Atlantic exceeded CF averaged over the 22 tropical South Atlantic by 20%. In September - October, when there was no hemispheric 23 asymmetry in total AOT over the tropical Atlantic (RAOT was close to 1), meridional CF 24 distribution was also almost symmetrical (R_{CF} was equal to 1, (Table 2 and Fig. 4)).

Fig. 6 represents meridional distribution of MODIS CF and TRMM accumulated rainfall, zonal averaged over the tropical Atlantic Ocean, for all months of the year. One can see some changes in CF from month to month on the high background level of approximately 0.6. This background level of CF is almost the same over the tropical North and South Atlantic Oceans.

In each month, the main CF maximum coincides with the Atlantic Ocean inter-tropical convergence zone, which is characterized by intensive rainfall (Fig. 6). In the summer months (when pronounced meridional dust asymmetry was observed), MODIS CF data showed significant CF to the north from the main CF maximum, over the latitudes of transatlantic dust

1 transport within the Saharan Air Layer (SAL) (Fig. 6, g to i). Saharan dust travels across the 2 Atlantic Ocean within the hot and dry Saharan Air Layer (Dunion and Velden, 2004). The SAL's base is at $\sim 900 - 1800$ m and the top is usually below 5500 m (Diaz et al., 1976). The 3 significant cloud fraction along SAL, together with the Atlantic Inter-tropical Convergence 4 5 Zone (centered over the tropical North Atlantic) contributed to the above-mentioned hemispheric CF asymmetry. Following is our analysis of cloud fraction in the area of the 6 7 Saharan Air Layer in July, when the most pronounced meridional dust asymmetry was 8 observed.

9

10 **4.5.1** Cloud fraction in the area of the Saharan Air Layer in July

Figure 7 represents meridional distribution of the 10-year mean of MERRAero dust AOT, 11 MODIS-Terra cloud fraction, and TRMM accumulated rainfall, zonal averaged over the 12 Atlantic Ocean ($60^{\circ}W - 0^{\circ}E$). The near-equatorial maximum in meridional distribution of 13 TRMM accumulated rainfall indicates the position of the North Atlantic Ocean inter-tropical 14 15 convergence zone (ITCZ) (Fig. 7). One can see that, in July, when dust presence over the 16 Atlantic is maximal, the meridional distribution of CF becomes essentially asymmetric with 17 respect to the center of ITCZ. In particular, significant CF up to 0.8 is seen to the North of ITCZ, over the latitudes with SAL presence $(12^{\circ}N - 24^{\circ}N)$ (Fig. 7). These values are higher 18 than the 10-year mean MODIS CF over the tropical North Atlantic (0.66) (Table 1). One can 19 20 consider that, in the North Atlantic, the wide maximum in the meridional distribution of CF consists of two different partly-overlapping maxima: one CF maximum located within ITCZ, 21 22 and the other CF maximum located to the north of ITCZ, over the ocean area where Saharan 23 dust is transported within the SAL across the Atlantic (Fig. 7).

24 More detailed information about the aforementioned two partly-overlapping maxima in the meridional distribution of CF in July can be obtained from a comparison between spatial 25 distribution of 10-year mean MERRAero dust AOT and MODIS CF over the tropical North 26 27 Atlantic (Fig. 8, a and b). It is clearly seen that the ocean area with Saharan dust transported 28 across the Atlantic is covered by cloudiness characterized by significant values of MODIS CF 29 up to 0.8 - 0.9. This CF is higher than the 10-year mean MODIS CF over the tropical North 30 Atlantic (0.66) (Table 1). Note that there is a strong difference between the two zones of significant CF in the North Atlantic. High values of CF within ITCZ are accompanied by 31

1 intensive rainfall (Fig. 8, b and c). By contrast, the area of SAL with significant CF $(12^{\circ}N - 24^{\circ}N)$ is characterized by essentially lower precipitation (Fig. 8, b and c).

3 To quantify changes in dust AOT, MODIS-based CF, and TRMM monthly-accumulated 4 rainfall with distance from the Sahara, we analyzed the 10-year mean (July 2002 – June 2012) of these parameters over six zones, each $6^{\circ} \times 6^{\circ}$, located along the Saharan Air Layer, in 5 6 accordance with the direction of dust transport (Fig. 8a). In July, there was a decrease of 7 approximately 300% in dust AOT from zone 1 to zone 6 (Fig. 9a). The reason for the 8 decrease in dust AOT with increasing distance from dust sources in the Sahara is gravitational 9 settling of dust particles (mainly coarse fraction). As shown in Figs. 9 a and b, the strong 10 decrease in dust AOT from zone 1 to zone 3 was not accompanied by any changes in TRMM 11 accumulated rainfall. Therefore, the washing out of aerosols by rainfall does not account for 12 the aerosol spatial decrease with distance from the Sahara. Consequently, it proves that 13 gravitational settling of dust particles accounts for the aerosol spatial decrease with distance 14 from the Sahara.

MODIS cloud fraction also decreased from zone 1 to zone 3, although less pronounced than dust (Fig. 9b). Over zones 1 - 3, there was significant cloud fraction in the presence of limited precipitation less than 20 mm month⁻¹. This indicates that clouds in zones 1 - 3 were not developed enough to produced intensive precipitation. This can be explained by the effect of temperature inversion below the SAL base on cloud formation (Prospero and Carlson, 1972).

20 To examine temperature inversion over the specified zones, we analyzed vertical profiles of 21 10-year mean MERRA Reanalysis atmospheric temperature in July, averaged over the 22 specified zones. As shown in Fig. 10, the temperature inversion existed over zones 1 - 4, and 23 it disappeared over zones 5 and 6. The observed temperature inversion over zones 1 to 4 (Fig. 10) prevented deep cloud formation, which explains the observed limited precipitation in 24 25 these zones (Fig. 9b). In the absence of temperature inversion over zones 5 and 6 (Fig. 10), one can consider the presence of developed clouds, which were capable of producing 26 27 intensive rainfall. Such developed clouds could explain the observed precipitation up to 110 mm month⁻¹ over zones 5 - 6 (Fig. 9b). 28

29

30 4.5.2 Influence of dust loading on CF in the area of SAL

The observed temperature inversion over zones 1 - 4 prevents deep cloud formation; this explains limited precipitation in these zones. On the other hand, meteorological conditions below the temperature inversion at the SAL base include significant atmospheric humidity
 and the presence of large amounts of settling dust particles together with marine aerosols.

3 As known, aerosol species often combine to form mixed particles, with properties different 4 from those of their components (Andreae et al., 2009). Mineral dust particles are known to be 5 not very efficient cloud condensation nuclei (CCN), unless they are coated with soluble 6 materials (Andreae et al., 2009). Using airplane measurements, Levin et al. (2005) showed 7 that dust transport over the sea could lead to sea-salt coating on dust particles. Coating settling 8 dust particles with sea-salt could modify them into efficient CCN. Being below the 9 temperature inversion and acting as efficient CCN. Saharan dust particles coated with soluble 10 material contribute to the formation of shallow stratocumulus clouds. This physical 11 mechanism, based on the indirect effect of Saharan dust on stratocumulus clouds below the 12 temperature inversion, could explain the observed significant cloud cover (CF up to 0.8 - 0.9) 13 along the Saharan Air Layer. The significant cloud fraction along SAL contributes to 14 hemispheric CF asymmetry over the tropical Atlantic. This could lead to hemispheric imbalance in strong solar radiation reaching the sea surface in the tropical Atlantic Ocean. 15

16 To examine the properties of clouds in the area of SAL, we analyzed available data on the 17 effective radius of cloud droplets. Fig. 11 represents histograms of the effective radius (Reff) 18 of cloud droplets for liquid water clouds in the specified zones 1 - 4 along SAL, based on MODIS L3 gridded monthly data $(1^{\circ} \times 1^{\circ})$ during the 10-year study period in July. The data 19 20 were supplied by the Giovanni data base. It is obvious that the cloud droplet effective radius 21 increases from zone 1 to zone 4 (Fig. 11). One can see a systematic shift in the whole 22 histogram to higher values of Reff from zone 1 to zone 4. This can be explained by the 23 decrease in CCN numbers associated with the decreasing numbers of settling Saharan dust 24 particles with distance from the Sahara, in accordance with the decrease in dust AOT shown 25 in Fig. 9a.

Thus, the cloud droplet effective radius in zone 4 was larger than in zones 1 - 3. This could lead to some increase in precipitation in zone 4. Indeed, as shown in Fig. 9b, TRMM accumulated rainfall in zone 4 was more intensive than over zones 1 - 3. This supports the above-mentioned physical mechanism of cloud formation below the temperature inversion at the SAL base.

In accordance with the above-mentioned mechanism of cloud formation along SAL, there are different cloud types over zones 1 - 4 on the one hand, and over zones 5 - 6 on the other 1 hand. Over zones 1 - 4, we consider the presence of shallow stratocumulus clouds below the 2 temperature inversion at the SAL base. These shallow stratocumulus clouds are characterized 3 by limited precipitation. Over zones 5 - 6, we consider the presence of developed clouds 4 capable of producing strong precipitation up to 110 mm month⁻¹.

5

6 4.5.3. Possible MODIS CF contamination by heavy dust loading

Collection 5 of MODIS-Terra monthly daytime cloud fraction data used in the current study
are derived from the standard cloud mask product based on the cloud mask algorithm MOD35
(Ackerman et al., 1998, Frey et al., 2008). In heavy dust loading situations, such as dust
storms over deserts, MOD35 may flag the aerosol-laden atmosphere as cloudy (Ackerman et al., 1998).

12 During dust storms over deserts, observed AOT values range from 2 to 5 (e.g. Alam et al., 13 2014). However, over the tropical North Atlantic in July, strong AOT exceeding even 1 is a 14 very rare phenomenon. To demonstrate that AOT exceeding 1 is a rare phenomenon over the 15 tropical North Atlantic, Fig. 12a represents a histogram of AOT observed over the tropical 16 North Atlantic in July, 2010, based on MODIS Level 3 AOT daily data. July 2010 was chosen 17 because AOT, averaged over the tropical North Atlantic, was maximal compared to AOT in 18 other July months, during the 10-year study period. One can see that AOT hardly exceeded 1. A similar situation can be seen over the latitudes with SAL presence $(12^{\circ}N - 24^{\circ}N)$ (Fig. 19 20 12b). Therefore, the effect of MODIS cloud fraction contamination by heavy dust loading 21 cannot essentially contribute to averaging CF over the tropical North Atlantic. Consequently, 22 given the large amount of available MODIS CF daily data over the 10-year study period, 23 cloud fraction contamination does not account for the obtained hemispheric CF asymmetry 24 over the tropical Atlantic Ocean.

25

26 **5** Conclusions

27 Meridional distribution of aerosol optical thickness and cloud fraction were analyzed using 28 10-year satellite measurements from MISR and MODIS, together with MERRAero data (July 29 2002 - June 2012). In the current study, we focus on the tropical Atlantic ($30^{\circ}N - 30^{\circ}S$) 30 which is characterized by significant amounts of Saharan dust dominating other aerosol 31 species over the North Atlantic. Our main point is that, over the tropical Atlantic, not only is Saharan dust responsible for the
 pronounced hemispheric aerosol asymmetry, but it also contributes to significant cloud cover
 along the Saharan Air Layer. This could lead to the hemispheric imbalance in strong solar
 radiation reaching the sea surface in the tropical Atlantic.

5 When hemispheric AOT asymmetry over the tropical North Atlantic was the most 6 pronounced, dust AOT averaged separately over the tropical North Atlantic was one order of 7 magnitude higher than that over the tropical South Atlantic. In July, the most pronounced 8 hemispheric asymmetry of dust AOT was characterized by the hemispheric ratio R_{DU} of 9 approximately 30. In the presence of such strong hemispheric asymmetry in dust AOT in the 10 summer months, CF averaged separately over the tropical North Atlantic exceeded CF 11 averaged over the tropical South Atlantic by 20%.

12 In July, along the Saharan Air Layer, MODIS CF data showed cloud cover up to 0.8 - 0.913 with limited precipitation ability. These CF values are higher than the 10-year mean MODIS 14 CF over the tropical North Atlantic (0.66) (Table 1). The observed significant cloud fraction 15 along SAL could be explained by the formation of shallow stratocumulus clouds below the 16 temperature inversion at the SAL base with the assistance of settling Saharan dust particles. 17 This cloud fraction along SAL together with clouds over the Atlantic Inter-tropical 18 Convergence Zone contributes to the above-mentioned hemispheric CF asymmetry between 19 the tropical North and South Atlantic.

With respect to different oceans, only over the Atlantic Ocean did MERRAero demonstrate that desert dust dominated all other aerosol species and was responsible for hemispheric aerosol asymmetry there. MERRAero showed that, over the tropical Atlantic, dust and carbonaceous aerosols were distributed asymmetrically relative to the equator, while other aerosol species were distributed more symmetrically.

Both MISR measurements and MERRAero data were in agreement on seasonal variations in hemispheric aerosol asymmetry. Hemispheric asymmetry in total AOT over the Atlantic was the most pronounced between March and July, when dust presence over the North Atlantic was maximal. In September and October, there was no noticeable hemispheric aerosol asymmetry in total AOT (R_{AOT} was close to 1). During these two months, the contribution of carbonaceous aerosols to total AOT in the South Atlantic was comparable to the contribution of dust aerosols to total AOT in the North Atlantic. Our study showed that, in September and 1 October, meridional CF distribution over the tropical Atlantic was almost symmetrical (R_{CF}

- 2 was close to 1).
- 3

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- 1 Table 1. Average AOT and CF over the tropical North (X_N) and South (X_S) Atlantic and their
- 2 hemispheric ratio (R)^a . 10-year MERRAero AOT, MISR AOT, and MODIS CF data were
- 3 used.

Data set	$X_N\pm\sigma_N$	$X_S\pm\sigma_S$	$R\pm\sigma_R$
MISR AOT	0.25 ± 0.06	0.15 ± 0.05	1.70 ± 0.06
MERRAero AOT	0.19 ± 0.05	0.12 ± 0.05	1.61 ± 0.06
MODIS CF	0.66 ± 0.09	0.61 ± 0.06	1.08 ± 0.01

4 ^aStandard deviations of X_N , X_S , and R are designated by σ_N , σ_S , σ_R respectively.

- 1 Table 2. The hemispheric ratio (± standard deviation) of dust AOT (DU), organic and black
- 2 carbon AOT (OC & BC), other aerosol species AOT (Other), and MODIS CF over the

3 tropical Atlantic Ocean ($30^{\circ}N - 30^{\circ}S$). 10-year MERRAero data and MODIS CF data were

4 used.

Month	DU	OC & BC	Other	MODIS CF
All months	11.50 ± 1.20	0.70 ± 0.10	1.10 ± 0.10	1.08 ± 0.01
January	6.10 ± 2.30	1.30 ± 0.50	1.10 ± 0.10	1.10 ± 0.07
February	4.20 ± 1.80	1.20 ± 0.40	1.20 ± 0.10	1.15 ± 0.09
March	6.90 ± 3.20	2.00 ± 0.40	1.20 ± 0.10	1.14 ± 0.10
April	8.80 ± 4.10	2.70 ± 0.40	1.20 ± 0.10	1.07 ± 0.09
May	21.00 ± 10.10	1.70 ± 0.30	1.20 ± 0.10	1.14 ± 0.07
June	23.50 ± 10.80	0.90 ± 0.30	1.30 ± 0.10	1.20 ± 0.09
July	29.30 ± 10.30	0.70 ± 0.30	1.30 ± 0.20	1.21 ± 0.08
August	25.00 ± 8.50	0.40 ± 0.10	1.10 ± 0.10	1.04 ± 0.07
September	23.80 ± 6.70	0.20 ± 0.10	0.90 ± 0.10	0.98 ± 0.05
October	17.00 ± 4.30	0.20 ± 0.10	0.80 ± 0.10	0.97 ± 0.05
November	9.70 ± 2.30	0.70 ± 0.20	0.80 ± 0.10	0.98 ± 0.05
December	6.80 ± 1.90	1.00 ± 0.30	0.90 ± 0.10	1.05 ± 0.05

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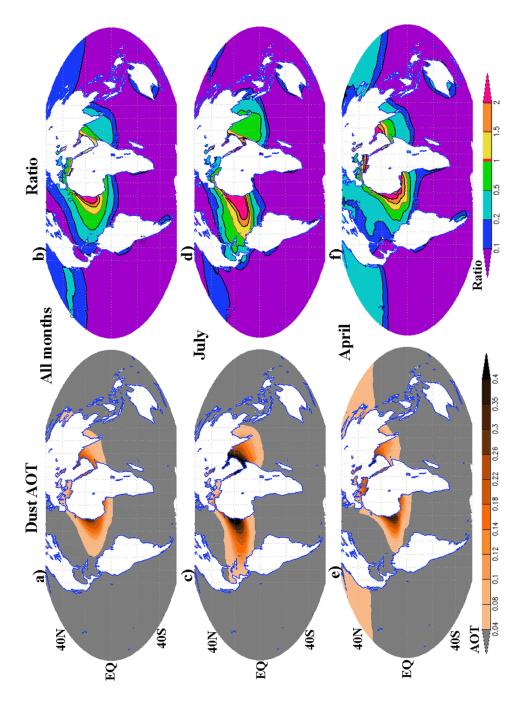


Figure 1. Spatial distributions of (a, c, and e) dust AOT (DU) and (b, d, and f) the ratio of DU AOT of all other aerosol species, based on the 10-y MERRAero data. In the right panel, the red contour line represents the boundary of the zone where dust AOT is equal to AOT of all other aerosol species.

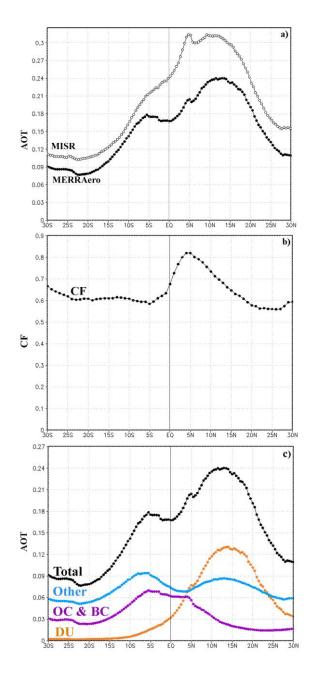


Figure 2. The meridional distribution of 10-year mean AOT/ CF, zonal averaged over the Atlantic Ocean ($60^{\circ}W - 0^{\circ}E$): a –total AOT based on MERRAero and MISR data; b – MODIS CF, c - MERRAero total AOT, dust AOT (DU), organic and black carbon AOT (OC & BC), and other aerosol species AOT (Other). The vertical lines designate the position of the equator.

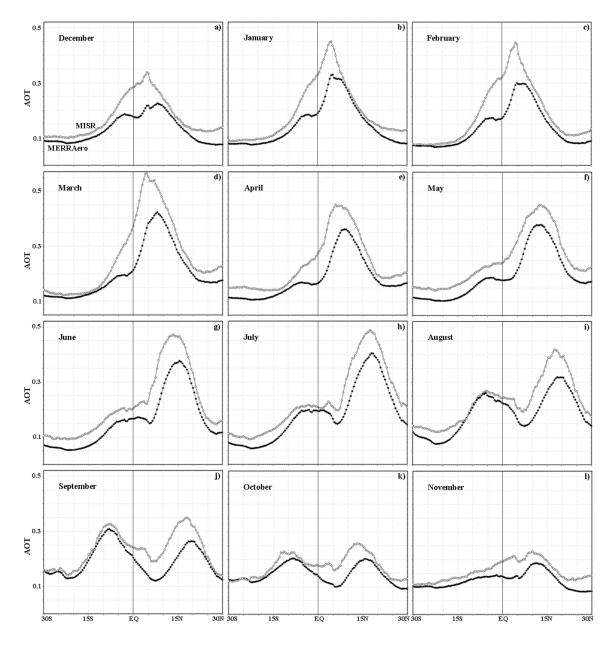
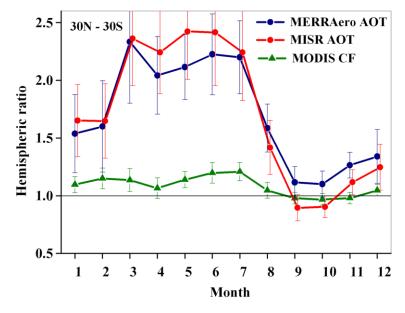


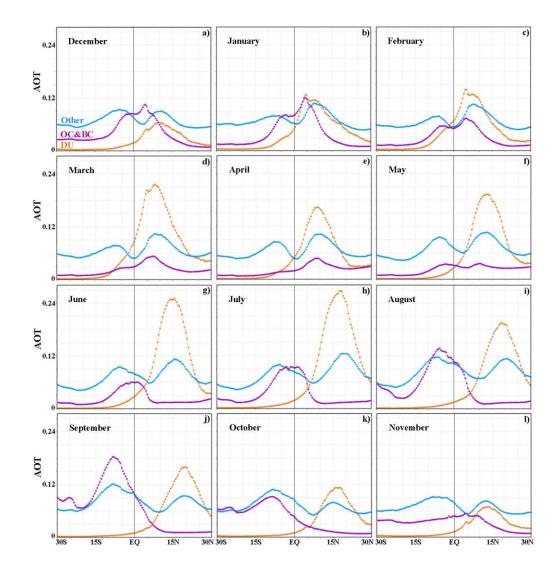
Figure 3. Meridional distribution of MISR and MERRAero total AOT, zonal averaged over
the Atlantic Ocean, for all months of the year. The vertical lines designate the position of the
equator.



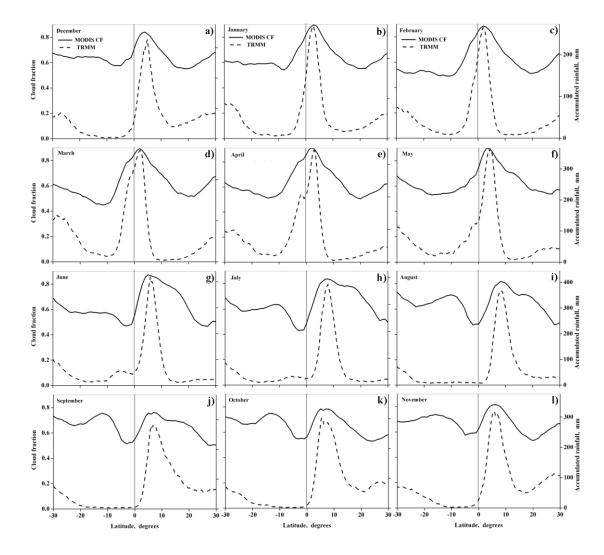
2 Figure 4. Month-to-month variations of the hemispheric ratio (R) of MISR AOT, MERRAero

3 AOT and MODIS cloud fraction (CF) over the tropical Atlantic Ocean $(30^{\circ}N - 30^{\circ}S)$. The

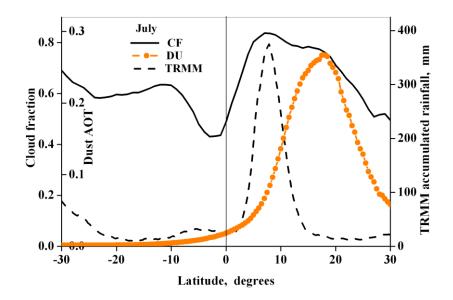
- 4 error bars show the standard deviation of R.
- 5



2 3 4 5 6 7 Figure 5. Meridional distribution of dust AOT (DU), organic and black carbon aerosol AOT (OC & BC), and other aerosol species AOT (Other), zonal averaged over the Atlantic Ocean, for all months of the year, based on 10-year MERRAero data. The vertical lines designate the position of the equator.



3 Figure 6. Meridional distribution of MODIS-Terra CF and TRMM accumulated rainfall, zonal averaged over the tropical Atlantic Ocean, for all months of the year. The vertical lines designate the position of the equator.





2 3 Figure 7. Meridional distribution of 10-year mean MODIS-Terra cloud fraction (CF), TRMM 4 accumulated rainfall and MERRAero dust AOT (DU), zonal averaged over the Atlantic 5 6 7 Ocean ($60^{\circ}W - 0^{\circ}E$), in July. The near-equatorial maximum in meridional distribution of TRMM accumulated rainfall indicates the position of the North Atlantic Ocean inter-tropical convergence zone (ITCZ). The vertical lines designate the position of the equator.

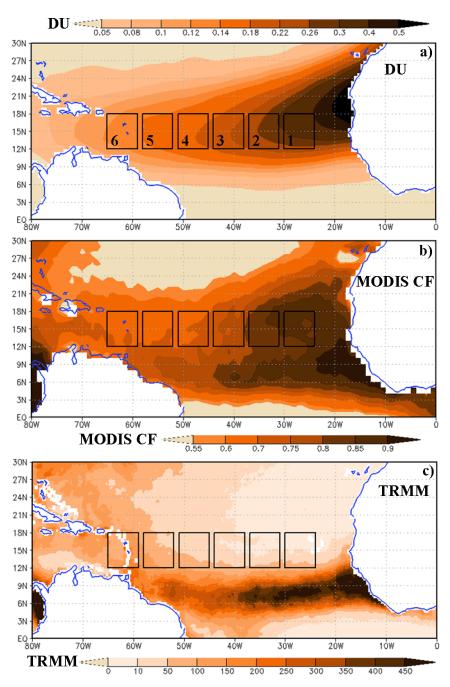


Figure 8. Spatial distributions of 10-year mean (a) MERRAero dust AOT (DU), (b) MODIS-Terra CF, and (c) TRMM accumulated rainfall over the North Atlantic in July. The geographic coordinates of the specified zones are as follows: zone 1 ($12^{\circ}N - 18^{\circ}N$; $30^{\circ}W 24^{\circ}W$), zone 2 ($12^{\circ}N - 18^{\circ}N$; $37^{\circ}W - 31^{\circ}W$), zone 3 ($12^{\circ}N - 18^{\circ}N$; $44^{\circ}W - 38^{\circ}W$), zone 4 ($12^{\circ}N - 18^{\circ}N$; $51^{\circ}W - 45^{\circ}W$), zone 5 ($12^{\circ}N - 18^{\circ}N$; $58^{\circ}W - 52^{\circ}W$), zone 6 ($12^{\circ}N - 18^{\circ}N$;

- $7 \quad 65^{\circ}W 59^{\circ}W).$
- 8

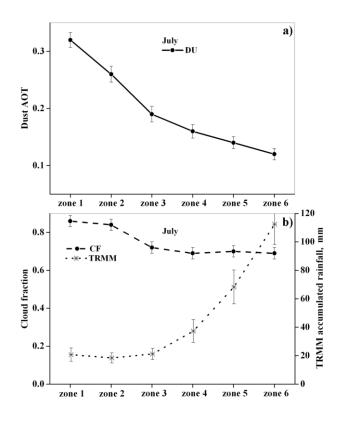
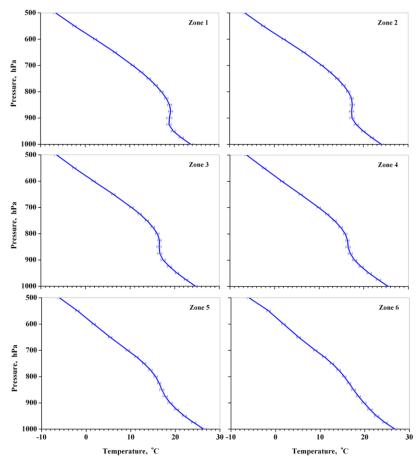


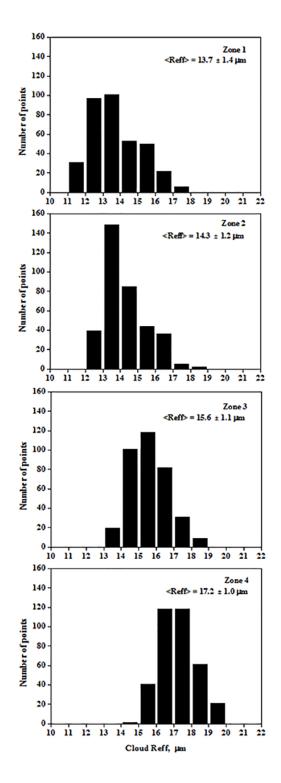
Figure 9. Zone-to-zone variations of (a) MERRAero dust AOT (DU); (b) MODIS-Terra CF

- and TRMM accumulated rainfall over the specified zones in July, averaged over the ten-year
- study period (2002 2012). The error bars show the standard error of mean.



2 3 4 Figure 10. Vertical profiles of 10-year mean MERRA Reanalysis atmospheric temperature (°C) in July, averaged over the specified zones along the route of transatlantic dust transport.

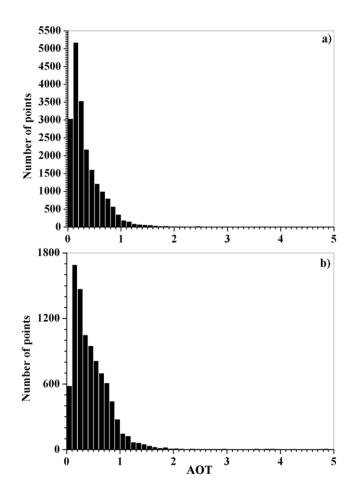
- The error bars show the standard deviation of temperature.
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1 2

Fig. 11. The histograms of effective particle radius (R_{eff}) for liquid water clouds in the specified zones 1 – 4 along SAL, based on MODIS Level 3 gridded monthly data with resolution 1° x 1° during the 10-year study period in July. The average R_{eff} (± its standard deviation) in each zone is shown.

3



4

Fig. 12. The histogram of AOT observed over the tropical North Atlantic in July 2010, based on Collection 5 MODIS-Terra Level 3 AOT daily data with resolution $1^{\circ} \times 1^{\circ}$: (a) over the tropical North Atlantic ($30^{\circ}N - 0^{\circ}N$; $60^{\circ}W - 0^{\circ}E$); (b) – over the latitudes with the SAL presence ($12^{\circ}N - 24^{\circ}N$; $60^{\circ}W - 0^{\circ}E$).