2	
3	on the manuscript acp-2014-540: "Meridional distribution of aerosol optical thickness over
4	the tropical Atlantic Ocean"
5	by P. Kishcha, A.M. da Silva, B. Starobinets, C.N. Long, O. Kalashnikova, and P. Alpert.
6	
7	We would like to thank Reviewer #2 for his helpful remarks. These remarks have been taken
8	into account in the revised manuscript, as listed below.
9	
10	"Anonymous Referee #2
11	Received and published: 24 September 2014 Provide of Maridianal distribution of general antical thickness over the transical Atlantic Ocean by P. Visheha et
13	al.
14	Line numbers refer to printer friendly version of the PDF posted in doi:10.5194/acpd-14-23309-2014
15	
16	Summary This paper seeks to characterize the distribution of aerosols (chiefly smoke and dust aerosols) and
17 18	their coincidence with cloud coverage in the Atlantic. The author carries out a statistical study spanning roughly 10 years of satellite observations (MODIS_MISP_TPMM_datastors) and acrossl modeling (CEOS_5). The study
19	focuses on comparing aerosol optical depths, cloud fractions and total rain amounts in the northern (0 to 30N)
20	and southern (0 to _30S) hemisphere sectors and points out the difference.
21	While I see the subject in general of interest, the work presented is very limited and it does no contribute
22	anything new.
23	
24	Our main point is that, over the tropical Atlantic, not only is Saharan dust responsible for the
25	hemispheric aerosol asymmetry, but it also contributes to significant cloud fraction along the
26	Saharan Air Layer. This significant cloud fraction in the area of SAL together with clouds
27	over the Atlantic Inter-tropical Convergence Zone contributes to hemispheric asymmetry in
28	cloud cover over the tropical Atlantic. This could lead to the hemispheric imbalance in strong
29	solar radiation reaching the sea surface over the tropical Atlantic. The phenomenon of
30	significant CF along SAL is important to the community and has not been reported so far, to
31	our knowledge.
32 22	In accordance with the Deviewer's noncentre in the new sections (112) and
33 24	In accordance with the Reviewer's remarks, in the revised version, new Sections 4.4.2 and 4.5.2 have been added (Dages 11, 12), where we discuss revised wersion for the
24 25	4.5.5 have been added (Pages 11 - 15), where we discuss physical mechanisms for the
33 26	$(D_{\text{pages }1}, 2)$
30 27	(rages 1 - 2).
38	The paper shows interesting coincidences between the presence of genosals and cloud properties but there is no
39	discussion or hypothesis presented on why this happens or whether they are just covariant parameters with an
40	underlying driving factor (for example, synoptic).
41	
42	In accordance with the Reviewer's remarks, in the revised version, a new section 4.5.2 has
43	been added (Pages 11 - 13), where we discuss possible physical mechanisms for the effect of
44	dust loading on cloud fraction in the area of the Saharan Air Layer.
45	We consider that the most likely physical mechanism for the formation of significant cloud
46	cover along SAL is as follows: The observed temperature inversion over zones 1 - 4 prevents
47	deep cloud formation; this explains limited precipitation in these zones. On the other hand,
48	meteorological conditions below the temperature inversion at the SAL base include
49	significant atmospheric humidity and the presence of large amounts of settling dust particles
50	together with marine aerosols.
51	As known, aerosol species often combine to form mixed particles, with properties different
52	from those of their components (Andreae et al., 2009). Mineral dust particles are known to be

REPLY TO REVIEWER #2'S REMARKS

not very efficient cloud condensation nuclei (CCN), unless they are coated with soluble 1 2 materials (Andreae et al., 2009). Using airplane measurements, Levin et al. (2005) showed 3 that dust transport over the sea could lead to sea-salt coating on dust particles. Coating settling 4 dust particles with sea-salt could modify them into efficient CCN. Being below the 5 temperature inversion and acting as efficient CCN, Saharan dust particles coated with soluble 6 material contribute to the formation of shallow stratocumulus clouds. This physical 7 mechanism, based on the indirect effect of Saharan dust on stratocumulus clouds below the 8 temperature inversion, could explain the observed significant cloud cover (CF up to 0.8 - 0.9) 9 along the Saharan Air Layer. The significant cloud fraction along SAL contributes to 10 hemispheric CF asymmetry over the tropical Atlantic. This could lead to hemispheric 11 imbalance in strong solar radiation reaching the sea surface in the tropical Atlantic Ocean.

To examine the properties of clouds in the area of SAL, we analyzed available data on the 12 13 effective radius of cloud droplets. Fig. 11 represents histograms of the effective radius (Reff) of cloud droplets for liquid water clouds in the specified zones 1 - 4 along SAL, based on 14 15 MODIS L3 gridded monthly data (1° x 1°) during the 10-year study period in July. The data 16 were supplied by the Giovanni data base. It is obvious that the cloud droplet effective radius increases from zone 1 to zone 4 (Fig. 11). One can see a systematic shift in the whole 17 histogram to higher values of Reff from zone 1 to zone 4. This can be explained by the 18 19 decrease in CCN numbers associated with the decreasing numbers of settling Saharan dust 20 particles with distance from the Sahara, in accordance with the decrease in dust AOT shown 21 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.

27

28 Methodologically, this study is weak in that a large source of the datasets use is the Giovanni NASA interface.

29 This is not a science quality data base and should not be used for the kind of analysis presented here. In

30 addition, there are some procedural deficiencies in the data analysis that need to be considered and have not

31 been made clear in the text. I recommend to reject the paper in the present form. I suggest the author to 32 resubmit after addressing the structural concerns about the topic and methodologies used."

32 33

With respect to the Reviewer's statement that the Giovanni NASA interface is not a science quality data base, we quote Dr James Acker from NASA Goddard Earth Sciences (GES) Data and Information Services Center (DISC) (personal communication on October 16, 2014): "Most of the data products in Giovanni are Level 3 data products provided by the processing of the science data provider. This means that the algorithms that produce them are vetted by scientists and science teams. We receive the data from the science and mission teams; we do

40 not produce any data products ourselves."

41 Note that there are almost a thousand peer-reviewed publications based on satellite data
42 provided by the NASA Giovanni interface
43 (http://disc.sci.gsfc.nasa.gov/giovanni/additional/publications).

44

In the current research, we did not study aerosol – cloud interaction. We consider that MODIS
data from the NASA Giovanni interface are suitable for averaging AOT and CF over

- 47 significant territories in the tropical Atlantic or for determining their meridional distribution.
- 48 We used MODIS Level 3 CF and AOT data to study meridional distribution of aerosols and
- 49 cloud fraction. In our previous study (Kishcha et al., JGR, 2009), we used MODIS CF and
- 50 AOT data from the Giovanni NASA interface. Our results have not been disproved. The

current study is a continuation of the above-mentioned previous research. In order to compare 1

2 the results obtained over the tropical Atlantic with those from the previous study, the same

3 data from the Giovanni had to be used.

4 "Major comments

5 6 7 8 9 There are two main concerns with this analysis: One is methodological. There are datasets used in this analysis which are of borderline scientific quality. Specifically, the use of data downloaded from the Giovanni NASA

- portal should be used with extreme care, particularly in the subject of aerosol-cloud interactions. While
- Giovanni provides a handy way to obtain and plot data, it does not deliver all the information needed to judge
- 10 the quality of data in use (see more details below). So, while Giovanni may provide images acceptable for use in
- 11 a publication (such a general view of AOD or Cloud Fraction distribution in the Atlantic basin), the L3 data
- 12 provided by the same portal is not suitable for the application proposed here."
- 13

14 As mentioned in our reply to the previous Reviewer's remark, in the current research, we did 15 not study aerosol - cloud interaction. Collection 5 of MODIS Level 3 data is suitable for 16 averaging AOT and CF over significant territories or for determining their meridional 17 distribution.

- In our other previous publication (Kishcha et al., JGR, 2012) on AOT trends over the Bay of 18
- 19 Bengal during the 10-year period (2000 - 2009), we compared MODIS Level 2 AOT trends
- with those based on MODIS Level 3 AOT data. We showed that, there was no noticeable 20
- difference between MODIS Level 3 AOT trends and those of MODIS Level 2. Therefore, 21
- 22 based on our experience, we consider that MODIS Level 3 data are suitable for averaging
- 23 AOT and CF over significant territories or for determining their meridional distribution.
- 24
- 25 " Specifically, causal relationships between aerosol and clouds need to be studied in the context of
- 26 meteorological influence that can cause covariance of aerosol and clouds parameters. This work does not
- 27 appear to make any consideration of such influence. A number of studies have already pointed out biases that
- 28 can confound the observation of aerosol-clouds relationship (Mauger and Norris, 2007). Furthermore recently,
- 29 Goren and Rosenfled (2014) listed in section 2 a number of reasons (with references) why cloud Fraction and
- 30 aerosol optical depth may or may not correlate. None of the effects can be discerned with the data set used and 31 original MODIS level 3 and level 2 data sets are needed for such analysis."
- 32

33 In the revised version (Section 4.5.2, see pages 11 - 13), we discuss physical mechanisms of 34 the influence of dust loading on cloud fraction along the Saharan Air Layer in July. See our 35 reply to the previous Reviewer's remark on "coincidences between the presence of aerosols 36 and cloud properties".

- In the current study, we focus on the phenomenon of significant cloud cover along SAL in the 37
- 38 summer months. As discussed in the revised version, the formation of shallow stratocumulus
- 39 clouds below the temperature inversion at the SAL base with the assistance of settling
- Saharan dust particles could explain the significant cloud cover along SAL. Neither Goren & 40
- 41 Rosenfeld (2014) nor Mauger & Norris (2007) discussed this specific case.
- 42
- 43 Second is conceptual. : What is the main science question addressed here? While the introduction implies that
- 44 this work is important for aerosol-cloud interaction studies, there is no case being made for the importance of an
- 45 N and S hemispheric asymmetry in cloud fraction and aerosol concentrations. How this asymmetry relates to the
- 46 indirect effects listed in the same introduction? In addition, the lack of discussion or explanations throughout the
- 47 text of features observed, reduces this whole work to a correlation study. Paper points out coincidences but no elaboration.
- 48 49
- 50 The main science question addressed here is the phenomenon of significant cloud cover (CF 51 up to 0.8 - 0.9) along SAL. This significant cloud cover, together with cloudiness over the
- 52 Atlantic Intertropical Convergence zone, contributes to hemispheric CF asymmetry, in the
- 53 presence of strong hemispheric aerosol asymmetry. This could lead to the hemispheric

- imbalance in strong solar radiation reaching the sea surface in the tropical Atlantic. This result sessential for understanding climate formation and its changes in the region in question. In the revised version (Section 4.5.2, pages 11 - 13), we discuss physical mechanisms responsible for the formation of significant cloud fraction along the Saharan Air Layer in the summer months.
- *Line by Line Comments Abstract: The opening paragraph should state why this study is important. The fact that there is an asymmetry observed needs to be accompanied of a hypothesis or a physical reason of why it is of interest to study this phenomenon. For clarity it would be useful to list in one sentence the data and tools use*
- 10 from MODIS, MISR and TRMM and MERRAAerol.
- 11
- In accordance with the Reviewer's remark, in the updated version, both the Abstract and the title have been updated in order to clarify the main point and why this study is important (Pages 1 - 3). Over a limited area such as the tropical Atlantic, we found that strong hemispheric asymmetry in dust aerosols was accompanied by hemispheric CF asymmetry, by contrast to the global ocean. This could lead to the hemispheric imbalance in strong surface solar radiation over the tropical Atlantic.
- The use of data from MODIS, MISR, and MERRAero was mentioned in the Abstract. We do
 not mention TRMM data as the abbreviation expansion takes too much space. Section 3
 (Method) is a suitable place to mentioned TRMM data together with all other data sets used.
- 21
 22 Page 23311. Line 20-26. This sentence is not clear. What does it mean that the "Southern Hemisphere contributed to the formation of noticeable meridional aerosol asymmetry"?
- 24
- In the manuscript, the sentence mentioned by the Reviewer is clarified by the subsequentsentence as follows:
- Kishcha et al. (2009) mentioned that not only the Northern Hemisphere but also the Southern
 Hemisphere contributed to the formation of noticeable meridional aerosol asymmetry. During
 the season of pronounced hemispheric aerosol asymmetry, an increase in AOT was observed
- 30 over the Northern Hemisphere, while a decrease in AOT was observed over the Southern31 Hemisphere.
- 32
- Pages 23311. Line 26 to P 233112 line 4. This is statement is not quite correct and mostly applicable to land
 satellite observations. Over the ocean, both MODIS and MISR are able to provide qualitative aerosol
- 35 identification that well suffice for a general study presented here.
- 36
- We agree with the Reviewer's comment. Indeed, over the ocean, MODIS and MISR satellites provide indication of three aerosol species such as dust, burning smoke and sea-salt aerosols (Guo et al., 2013, Kim et al., 2014). Although, sea-salt aerosol AOT is not a satellite retrieval, it is estimated from the empirical formula as a function of surface wind speed (Kaufman et al., 2005). No information about industrial anthropogenic aerosols is included in the abovementioned products of MODIS and MISR. The statement mentioned by the Reviewer has been removed from the revised version.
- 44
- 45 P 233112 line 17 to 25. It is not clear why the asymmetry pointed out is important. While the previous paragraph
 46 lists several possible effects, it is not clear what effect or what mechanism this study will address. The mere
- study of aerosol distribution with respect to cloud fraction does not necessarily can help to understand the
 phenomena listed in the previous paragraph.
- 48 49
- 50 As mentioned in the revised version (Introduction), hemispheric asymmetry in cloud fraction 51 (CF) and aerosols lead to hemispheric imbalance in solar radiation reaching the Earth's

surface. Consequently, analyzing hemispheric asymmetry in CF and aerosols is essential for 1 2 our understanding of climate formation and its changes. Previous studies showed that, over 3 the global ocean, there is hemispheric asymmetry in aerosols and no noticeable asymmetry in 4 cloud fraction (CF). We chose the tropical Atlantic because it is characterized by significant 5 amounts of Saharan dust dominating other aerosol species over the North Atlantic. We wished 6 to find out if the meridional CF distribution remains symmetrical in the presence of such 7 strong hemispheric aerosol asymmetry. This explains our motivation for doing the current 8 study. 9

10 We found a phenomenon that, in the summer months, along the Saharan Air Layer, 11 significant cloud cover (up to 0.8 - 0.9) was observed, based on MODIS CF data. This cloud 12 fraction along SAL together with clouds over the Atlantic Inter-tropical Convergence Zone 13 contributes to the hemispheric CF asymmetry

14 15

19

P 23313, line 25. State the MISR algorithm version and data level used in the analysis.

17 In accordance with the Reviewer's remark, in the revised version, it has been mentioned that 18 Version 3.1 of MISR Level 3 AOT data was used (Page 5, lines 12 - 13).

Line 23315. line 1-5. 1) This is the first time that TRMM data is mentioned. Since it is a dataset well used in this
 paper, it should be mentioned in the abstract

As mentioned in our reply to the previous Reviewer's comment on TRMM data, we do not
mention TRMM data in the Abstract as the abbreviation expansion takes too much space.
Section 3 (Method) is a suitable place to mention TRMM data together with all other data sets
used (Page 5, lines 22 – 23).

27

2) Giovanni is NOT a reliable source of scientific quality data. While the Giovanni interface is a handy tool for
the creation of quicklooks and general assessment of data sets, it does not have datasets of research quality
level. There are multiple reasons: a) data versions are not updated regularly b) downloaded data from Giovanni
does not include a number of quality flags and checks that are available in the original Level 3 data. Issues such
as formulae used for averages, weights used on the average, time of the day when the observation was made,
propagation of the quality flag from the original L2 to the L3 are not properly explained or not included in the
data displayed in the interface.

35

As mentioned in our reply to the previous Reviewer's remarks on Giovanny NASA data, the current study is a continuation of our previous research (Kishcha et al., JGR, 2009). In that paper we used MODIS Level 3 CF and AOT data from the NASA Giovanni, and we found that over the global ocean, there is hemispheric asymmetry in aerosols and no noticeable asymmetry in cloud fraction. Our results have not been disproved. In order to compare the results obtained over the tropical Atlantic with those from the previous study, the same MODIS Level 3 CF data are used.

Moreover, in our other previous publication (Kishcha et al., JGR, 2012) on AOT trends over
the Bay of Bengal during the 10-year period (2000 – 2009), we compared MODIS Level 2
AOT trends with those based on MODIS Level 3 AOT data. We showed that, there was no
noticeable difference between MODIS Level 3 AOT trends and those of MODIS Level 2.
Therefore, based on our experience, we consider that MODIS Level 3 data can be used for
averaging AOT and CF over significant territories or for determining their meridional

49 distribution.

" Specifically and by own experience of this reviewer, the MODIS CF reported by Giovanni is vey buggy with a

tendency to report high cloud fraction in dusty areas because the cloud algorithm interprets the high reflectance

of heavy dust as a cloud. This is a defect of the original MODIS algorithm cloud detection method and it is

1 2 3 4 5 propagated to all products downstream. Because this failure to properly detect clouds in heavy aerosol loading environments, the aerosol MODIS group choose to create their own cloud mask algorithm (and used in the 6 aerosol algorithm)."

7

8 In the revised version (Section 4.5.3, page 13), we discuss the effect of MODIS CF 9 contamination by heavy dust loading. Specifically,

10 Collection 5 of MODIS-Terra monthly daytime cloud fraction data used are derived from the

11 standard cloud mask product based on the cloud mask algorithm MOD35 (Ackerman et al.,

12 1998, Frey et al., 2008). In heavy dust loading situations, such as dust storms over deserts,

13 MOD35 may flag the aerosol-laden atmosphere as cloudy (Ackerman et al., 1998).

14 During dust storms over deserts, observed AOT values range from 2 to 5 (e.g. Alam et al.,

15 2014). However, over the tropical North Atlantic, strong AOT exceeding even 1 is a very rare 16 phenomenon. To demonstrate that AOT exceeding 1 is a rare phenomenon over the tropical

17 North Atlantic, Fig. 11a represents a histogram of AOT observed over the tropical North

Atlantic in July, 2010, based on MODIS Level 3 AOT daily data. July 2010 was chosen 18

19 because AOT, averaged over the tropical North Atlantic, was maximal compared to AOT in

20 other July months, during the 10-year study period. One can see that AOT hardly exceed 1. A

21 similar situation can be seen over the latitudes with SAL presence $(12^{\circ}N - 24^{\circ}N)$ (Fig. 12b).

22 Therefore, the effect of MODIS cloud fraction contamination by heavy dust loading cannot

23 essentially contribute to averaging CF over the tropical North Atlantic. Consequently, given

24 the large amount of available MODIS CF daily data over the 10-year study period, cloud

25 fraction contamination does not account for the obtained hemispheric CF asymmetry.

26

27 P23315 Line 10, replace "demonstrated" with "shows" Sections 4.2 and 4.3. Both sections are limited to 28 describe the listed figures but there is no explanations offered for the coincidences (or anti-coincidences) noted. 29 Is there an indirect effect or are these co-varying parameters? 30

31 In accordance with the Reviewer's comment, the word "demonstrated" has been replaced by 32 "showed" (Page 6, line 5). In the revised version, a new section 4.5.2 has been added, where 33 we discuss the effect of dust loading on cloud fraction in the area of the Saharan Air Layer. 34

35 "Section 4. There is description of Figure 4a but there is nothing said about figures 4b and 4c. Please add text 36 or remove the figures altogether. 37

38 This is the erroneous remark. There is only Fig. 4 in the manuscript, but there are no Figs. 4a, 39 4b or 4c. We discuss Fig. 4 in Sections 4.3 and 4.5.

40

41 "P 23317, line 22-25. This is a generalization that is not quite correct." 42

43 In the revised version, the statement has been updated as follows:

44 Therefore, aerosols over the tropical Atlantic can be divided into two groups with different 45 meridional distribution relative to the equator: dust and carbonaceous aerosols were 46 distributed asymmetrically, while other aerosol species were distributed more symmetrically 47 (Table 2) (Page 8, lines 2-5).

48

49 P23321, 115-16. What is the justification of this statement? Wet removal can be an important reason too and I 50 do not see why it should be ignored.

Indeed, rainfall always removes aerosols. However, as shown in Fig. 9, a strong decline in dust AOT from zone 1 to zone 3 was not accompanied by any changes in TRMM accumulated rainfall. Therefore, rainfall does not account for the dust spatial decrease with distance from the Sahara. It proves that gravitational settling of dust particles accounts for the serosol spatial decrease with distance from the Sahara. (Page 11, lines 9 - 14).

6 7

8

P23321, lines 25-30. What data was used to generate the plots in figures 10? it is not indicated.

As mentioned in the revised version (in the caption of Fig. 10, Page 32) Fig. 10 represents
 vertical profiles of 10-year mean MERRA Reanalysis atmospheric temperature over the
 specified zones in July.

- P23322, lines 1-4. Why the increase in rainfall is attributed to heavy convection? Shallow Cu are abundant in
 this area too so it is not clear why they are not considered.
- 15

12

16 The increase in the 10-year mean TRMM accumulated rainfall over zones 5 and 6 (Fig. 10) is 17 attributed to the absence of temperature inversion. In the absence of temperature inversion

- there is no suppression of deep cloud formation. We consider that thick developed clouds
- mainly contribute to significant precipitation up to 110 mm month⁻¹ observed over zone 6
- 19 mainly contribute to significant precipitation up to 110 mm month observed over zone (20 (Fig. 0). Shallow everyly along a sense he responsible for such significant precipitation
- 20 (Fig. 9). Shallow cumulus clouds cannot be responsible for such significant precipitation.21
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- 10

Hemispheric asymmetry in aerosol optical thickness and cloud fraction over the tropical Atlantic Ocean

3

P. Kishcha¹, A.M. da Silva², B. Starobinets¹, C.N. Long³, O. Kalashnikova⁴, and P. Alpert1

6 [1]{Department of Geosciences, Tel-Aviv University, Tel-Aviv, Israel}

- 7 [2]{Global Modeling and Assimilation Office, NASA/GSFC, Greenbelt, Maryland USA}
- 8 [3]{Pacific Northwest National Laboratory, Richland, Washington, USA}
- 9 [4]{Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109, USA
- 10

}

11

12 Correspondence to: P. Kishcha (pavelk@post.tau.ac.il)

13

14 Abstract

Previous studies showed that, over the global ocean, there is no noticeable hemispheric 15 16 asymmetry in cloud fraction (CF). This contributes to the balance in solar radiation reaching the sea surface in the Northern and Southern hemispheres. In the current study, we focus on 17 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 in the tropical Atlantic. During the 10-year study period (July 2002 – June 2012), NASA 23 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 3 cover (up to 0.8 - 0.9). This significant cloud fraction along SAL together with clouds over 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 and cloud cover because of their capability of providing global coverage on a regular basis. 16 17 Previous studies, using different space-borne aerosol sensors, discussed the idea that the hemispheres are asymmetric in aerosol distribution (Remer et al., 2008; Kaufman et al., 18 19 2005a; Remer and Kaufman, 2006, Mishchenko and Geogdzhayev, 2007, Chou et al., 2002, Zhang and Reid, 2010, Hsu et al., 2012, Kishcha et al, 2007, 2009). The Advanced Very High 20 21 Resolution Radiometer (AVHRR) satellite data over the ocean were used by Mishchenko and 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 25 using the Sea-viewing Wide Field-of-view Sensor (SeaWIFS) satellite data for the year 1998. Hsu et al. (2012) displayed the asymmetric spatial distribution of seasonally-averaged 26 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, 31 2010, Kishcha et al., 2007, 2009). In our previous study (Kishcha et al., 2009), AOT data 32 from three satellite sensors (MISR, MODIS-Terra, and MODIS-Aqua) were used in order to

analyze seasonal variations of meridional AOT asymmetry over the global ocean. The 1 2 asymmetry was pronounced in the April-July months, while there was no noticeable 3 asymmetry during the season from September to December. Kishcha et al. (2009) mentioned 4 that not only the Northern Hemisphere but also the Southern Hemisphere contributed to the 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 Lamb, 2003). Previous studies have shown that desert dust particles can influence the Earth's 11 atmosphere in the following ways: directly by scattering and absorbing solar and thermal 12 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 (ITCZ) northward toward the Saharan Air Layer (SAL). Alpert et al. (1998) discussed the 16 17 response of the atmospheric temperature field to the radiative forcing of Saharan dust over the North Atlantic Ocean. Dust particles over the Atlantic Ocean may essentially influence 18 19 tropical cloud systems and precipitation (Kaufman et al., 2005b, Johnson et al., 2004, Min et al., 2009, Ben-Ami et al., 2009, Feingold et al., 2009, Rosenfeld et al., 2001). 20

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 $(30^{\circ}N - 30^{\circ}S)$ because it is characterized by significant amounts of Saharan dust. We wished 23 24 to find out if the meridional CF distribution remains symmetrical relative to the equator in the presence of such strong hemispheric aerosol asymmetry. We determined and compared the 25 26 contribution of desert dust and that of other aerosol species to aerosol asymmetry between the tropical North and South Atlantic Oceans. Analyzing the meridional distribution of various 27 28 aerosol species over the tropical Atlantic Ocean was carried out using the NASA Aerosol Reanalysis (aka MERRAero). This reanalysis has been recently developed at NASA's Global 29 30 Modeling Assimilation Office (GMAO) using a version of the NASA Goddard Earth 31 Observing System-5 (GEOS-5) model radiatively coupled with Goddard Chemistry, Aerosol, 32 Radiation, and Transport (GOCART) aerosols. An important property of GEOS-5 is data 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 10 aerosol components (sulfates, organic carbon, black carbon, desert dust, and sea-salt). GEOS-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 have wind-speed dependent emission functions (Colarco et al., 2010), while sulfate and 13 14 carbonaceous species have emissions principally from fossil fuel combustion, biomass burning, and bio-fuel consumption, with additional biogenic sources of organic carbon. 15 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 (Darmenov and da Silva, 2013). GEOS-5 also includes assimilation of AOT observations 21 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 obtained meridional aerosol distribution based on MERRAero, we used the Multi-angle 25 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.

12 Variations of meridional aerosol distributions were analyzed by using Version 3.1 of MISR 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 16 (AERONET) Sun photometer measurements over different regions (Martonchik et al., 2004; 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 monthly daytime cloud fraction (CF) data, with horizontal resolution 1° x 1° was used 20 21 (Ackerman et al., 1998, Frey et al., 2008, King et al., 2003). Furthermore, to analyze 22 meridional rainfall distribution, the Tropical Rainfall Measuring Mission (TRMM) monthly 23 0.25° x 0.25° Rainfall Data Product (3B43 Version 7) was used (Huffman et al., 2007). MODIS CF data and TRMM data were acquired using the GES-DISC Interactive Online 24 Visualization and Analysis Infrastructure (Giovanni) as part of NASA Goddard Earth 25 26 Sciences (GES) Data and Information Services Center (DISC) (Acker and Leptoukh, 2007).

1 4 Results

2 4.1 Ocean zone with the predominance of desert dust aerosols

MERRAero showed that the Sahara desert emits a significant amount of dust into the 3 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 and 30°N, Saharan dust dominates other aerosol species (Fig. 1b). The longitudinal dimension 10 11 of this zone is subject to seasonal variability. During the dusty season from March to July, the 12 zone of dust predominance occupies a significant part of the tropical Atlantic between North 13 Africa and Central America. Specifically, as shown in Fig. 1d, in July, the zone of dust predominance is extremely extensive. By contrast, from October to February, this zone is 14 15 observed only over some limited territory close to North Africa.

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

Figure 2a represents meridional distribution of ten-year mean AOT (July 2002 – June 2012), 23 zonal averaged over the tropical Atlantic Ocean. One can see that MERRAero showed 24 similarity to the meridional AOT distribution, based on MISR data (Fig. 2a). Specifically, 25 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 28 tropical South Atlantic. This monomodal AOT maximum was discussed in our previous study 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 19 burning activity is maximal during December - February (Haywood et al., 2008). MERRAero 20 showed that no noticeable hemispheric asymmetry of total AOT was observed in September and October (Fig. 4). This is because the contribution of carbonaceous aerosols (OC & BC) to 21 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 (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 5 represents the meridional distribution of 10-year mean cloud fraction, zonal averaged over the 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 distributed more symmetrically. During the period of pronounced meridional AOT 13 asymmetry over the tropical Atlantic from May - July, dust AOT averaged separately over the 14 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

transport within the Saharan Air Layer (SAL) (Fig. 6, g to i). Saharan dust travels across the 1 2 Atlantic Ocean within the hot and dry Saharan Air Layer (Dunion and Velden, 2004). The 3 SAL's base is at $\sim 900 - 1800$ m and the top is usually below 5500 m (Diaz et al., 1976). The 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 14 TRMM accumulated rainfall indicates the position of the North Atlantic Ocean inter-tropical convergence zone (ITCZ) (Fig. 7). One can see that, in July, when dust presence over the 15 16 Atlantic is maximal, the meridional distribution of CF becomes essentially asymmetric with respect to the center of ITCZ. In particular, significant CF up to 0.8 is seen to the North of 17 ITCZ, over the latitudes with SAL presence $(12^{\circ}N - 24^{\circ}N)$ (Fig. 7). These values are higher 18 19 than the 10-year mean MODIS CF over the tropical North Atlantic (0.66) (Table 1). One can 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).

To quantify changes in dust AOT, MODIS-based CF, and TRMM monthly-accumulated 3 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 accordance with the direction of dust transport (Fig. 8a). In July, there was a decrease of 6 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 the aerosol spatial decrease with distance from the Sahara. Consequently, it proves that 12 13 gravitational settling of dust particles accounts for the aerosol spatial decrease with distance 14 from the Sahara.

15 MODIS cloud fraction also decreased from zone 1 to zone 3, although less pronounced than 16 dust (Fig. 9b). Over zones 1 - 3, there was significant cloud fraction in the presence of limited 17 precipitation less than 20 mm month⁻¹. This indicates that clouds in zones 1 - 3 were not 18 developed enough to produced intensive precipitation. This can be explained by the effect of 19 temperature inversion below the SAL base on cloud formation (Prospero and Carlson, 1972).

To examine temperature inversion over the specified zones, we analyzed vertical profiles of 20 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. 24 10) prevented deep cloud formation, which explains the observed limited precipitation in 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.

As known, aerosol species often combine to form mixed particles, with properties different 3 4 from those of their components (Andreae et al., 2009). Mineral dust particles are known to be not very efficient cloud condensation nuclei (CCN), unless they are coated with soluble 5 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 temperature inversion and acting as efficient CCN, Saharan dust particles coated with soluble 9 material contribute to the formation of shallow stratocumulus clouds. This physical 10 mechanism, based on the indirect effect of Saharan dust on stratocumulus clouds below the 11 temperature inversion, could explain the observed significant cloud cover (CF up to 0.8 - 0.9) 12 13 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 14 15 imbalance in strong solar radiation reaching the sea surface in the tropical Atlantic Ocean.

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

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., 2014). However, over the tropical North Atlantic in July, strong AOT exceeding even 1 is a 13 very rare phenomenon. To demonstrate that AOT exceeding 1 is a rare phenomenon over the 14 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. 19 A similar situation can be seen over the latitudes with SAL presence $(12^{\circ}N - 24^{\circ}N)$ (Fig. 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 - 0.9with limited precipitation ability. These CF values are higher than the 10-year mean MODIS 13 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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- 20

- 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

6



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



3 Figure 5. Meridional distribution of dust AOT (DU), organic and black carbon aerosol AOT 4 5 6 7 (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 6 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 Ocean ($60^{\circ}W - 0^{\circ}E$), in July. The near-equatorial maximum in meridional distribution of 5 6 7 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 ($12^{\circ}N - 18^{\circ}N$; $65^{\circ}W - 59^{\circ}W$).



3 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.



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.

- 4 The error bars show the standard deviation of temperature.



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

5 Fig. 12. The histogram of AOT observed over the tropical North Atlantic in July 2010, based 6 on Collection 5 MODIS-Terra Level 3 AOT daily data with resolution $1^{\circ} \times 1^{\circ}$: (a) over the 7 tropical North Atlantic ($30^{\circ}N - 0^{\circ}N$; $60^{\circ}W - 0^{\circ}E$); (b) – over the latitudes with the SAL

8 presence $(12^{\circ}N - 24^{\circ}N; 60^{\circ}W - 0^{\circ}E)$.