Chemical aging of atmospheric mineral dust during transatlantic transport

Reply to Anonymous Referee #1 (doi:10.5194/acp-2016-470-RC1)

http://editor.copernicus.org/index.php/acp-2016-470-RC1.pdf?_mdl=msover_md&_jrl=10&_lcm=oc108lcm109w&_acm=get_comm_file&_ms=51800&c=111158&salt=895046841231028569

by Mohamed Abdelkader and Swen Metzger, et al.,

December 19, 2016

We thank the anonymous referee for the comments on this manuscript. The comments and questions raised are addressed below by our point-by-point reply (black) and the revised MS.

The authors used EMAC (The ECHAM5/MESSy2 atmospheric chemistry General Circulation Model) to evaluate transport and loadings of mineral dust particles during transatlantic transport. The study carefully considered aging mineral dust in the model and compared the results with non-aging mineral dust particles. They found some interesting results such as the removable efficiency and optical properties. These results will be potential useful for the future study on the ground base. On the other hand, the study carefully used the satellite data (AOD and CALIPSO) to calibrate the modeling results. They obtained the consistent results. The developed method is significant to improve the current model.

We thank the referee for this general comment.

The mineral dust particles are important for climate change, biogeochemical cycle, and heterogeneous atmospheric chemistry in global. Many studies found how the mineral dust changes in air. However, the modeling work is rare. The modeling work is useful to evaluate effects of mineral dust in the air. Although the modeling parameters are not based on measurements, the results and comparison is interesting.

We also appreciate this comment.

I would like to recommend accepting this paper after one minor revision. In the introduction section, the authors should add some findings in field campaigns which have revealed the nitrate coatings on alkaline mineral dust particles in the worlds. For example, Tobo, Li, Sullivan et al., found mineral dust aging process in the air. Although the authors consider the mineral dust particles absorbing acidic gases transformed from SO2, NOx, or HCl. However, these field study all pointed out the nitrate coating determine particle hygroscopic properties ("Asian dust particles converted into aqueous droplets under remote marine atmospheric conditions." P Natl Acad Sci USA 107(42): 17905-17910./ "Observation of nitrate coatings on atmospheric mineral dust particles." Atmos. Chem. Phys. 9(6): 1863-1871/"Direct observations of the atmospheric processing of Asian mineral dust." Atmos. Chem. Phys. 7: 1213-1236.).

We do agree that nitrate coating can determine hygroscopicity of mineral dust particles, which is especially the case in an polluted atmosphere (Bauer et al., 2007). Moreover, our EMAC set-up accounts for this effect, since the nitric acid (e.g., as oxidation end product of combustion NO_x) may react in our set-up with the calcium fraction of the mineral dust particles to form calcium nitrate, which takes up water vapour from the atmosphere at ambient conditions where the humidity is just about 50% (the RHD of $Ca(NO_3)_2$ is 48% at T=298 K). In strong contrast, dust coating by sulphuric acid does not lead to hygroscopic particles since the RHD of $CaSO_4$ is close to 100% (at any T).

The authors should mention the aged mineral dust particles become hydrophilic and can act as CCN during the transport (Mixing state and hygroscopicity of dust and haze particles before leaving Asian continent. J. Geophys. Res. 119 (2), 1044-1059.)

This sentence and the reference has been added to the introduction of the revised MS.

Page 1 line 16 miss blank after comma.

We have added this blank in the revised MS.

Figure 3 should be marked where is the Cribbean.

We have marked the Caribbean in Figure 3 of the revised MS.

Why did not the authors consider the mineral dust as ice nucleation? It could be one removable pathway for mineral dust in air.

We agree that the consideration of mineral dust can be regionally important for ice nucleation. However, the effect will be less pronounced for our global modeling. The main reason is simply that the cloud micro-physical processes needs to be parameterized for the still relatively coarse model grid box (here approx 110 km). On these (model grid) scales many (partly unknown) micro-physical processes are implicitly parameterized, if the model results more or less agree with e.g., AOD observations. Changes in the micro-physical assumptions will therefore not alter the overall picture much. We have learned that from several additional sensitivity studies. Thus, for the current scope of this paper, we omit a more explicit aerosol-cloud coupling that includes feedback of mineral dust particles on ice nucleation. Aerosol-cloud coupling of dust is implicitly accounted for by changes in solubility of the aged dust particles due water uptake, which feeds back with scavenging, cloud water content, remaining aerosol loadings and radiation. A more detailed analysis of the current assumptions on aerosol-cloud coupling will be presented elsewhere.

I recommend revising the current title. Because the study focused on the evaluation of mineral dust during transatlantic transport using model and other methods, it didn?t study chemical aging of mineral dust. The current title seems that the study understand the chemical aging mechanism of mineral dust in the air.

We have revised the title to:
"Sensitivity of transatlantic dust transport to chemical aging and related atmospheric processes".

References

Bauer, S. E., Mishchenko, M. I., Lacis, A. A., Zhang, S., Perlwitz, J., and Metzger, S. M.: Do sulfate and nitrate coatings on mineral dust have important effects on radiative properties and climate modeling?, Journal of Geophysical Research: Atmospheres, 112, D06 307, doi: 10.1029/2005JD006977, URL `http://dx.doi.org/10.1029/2005JD006977`, 2007.

Chemical aging of atmospheric mineral dust during transatlantic transport

Reply to Anonymous Referee #2 (doi:10.5194/acp-2016-470-RC2)

http://editor.copernicus.org/index.php/acp-2016-470-RC2.pdf?_mdl=msover_md&_jrl=10&_lcm=oc108lcm109w&_acm=get_comm_file&_ms=51800&c=111819&salt=102505675193170096

by Mohamed Abdelkader and Swen Metzger, et al.,

December 19, 2016

We thank the anonymous referee for comments on this manuscript. The comments and questions raised are addressed below by our point-by-point reply (black) and the revised MS.

General comments

This work describes the effects of chemical aging, emissions and convection parameterizations in the transport of desert dust over the Atlantic Ocean with the use of the atmospheric chemistry general circulation model EMAC. The authors have published the concept of dust chemical aging in a recent paper and in this new publication they deal with the transatlantic transport and how it can be affected by various model parameterizations related to the dust cycle. Modeling the desert dust cycle is a complicated topic given the necessity to parameterize physical processes that produce and cycle dust particles throughout the atmosphere and a better understanding of how to improve these processes is significant.

We thank the referee for this general comment.

I found the paper difficult to read, in terms of the flow, especially because there is a continuous description of the figures instead of using them to support a conclusion or remark. The main review comments are related to clarifications in the methodology and discussion of the results. I am in favor of publishing this paper with Atmospheric Chemistry and Physics with Major Revisions. The specific comments that follow will help improve the discussion of the methodology and significance of the findings so that the overall quality of the manuscript is enhanced.

We also appreciate the specific comments.

Specific comments/suggestions.

1. Please refer to aging of dust as "chemical aging" in all parts of the manuscript.

Changed "aging" to "chemical aging" throughout the manuscript.

2. Introduction, page 2, line 34: in the sentence "mean normalized bias of the AOD model varies", the word "model" should be omitted.

The word "model" is omitted.

3. Please provide the specific modules used in the EMAC configuration so that the results from this work can be reproducible.

Table 1 is included which shows the EMAC submodels used in the study.

4. Are indirect aerosol-cloud interactions included in the model configuration, besides the radiative feedback effect? How different the results might have been if these interactions were included?

Yes, through changes in the scavenging efficiency, but not through changes in CCN activity. The impact of the latter does not alter our results, since we have focused on the chemical aging of a major dust outflow between 2000 and 2013 (i.e., July 2009). For such a case, the chemical aging as represented here (various effects of changes in the wet radius) dominates the aerosol-cloud-radiation coupling. Nevertheless, the topic deserves further investigations and will be subject of a follow-up study, which then will focus on the chemical aging of weaker dust-outflow events.

5. Page 3, line 23: what is the meaning of "increases the level of dust aging"? Is there a specification of levels of chemical aging that the authors consider? I am assuming that inorganic acids uptake by the dust particles is what differentiates freshly emitted dust with dust being transported in the atmosphere, which eventually leads to "chemical aging" since the original dust particle has an altered chemical signature. Unless water uptake is considered the primary aging process. Please clarify.

We have changed this sentence to:
"This increases the dust particle mass, particle size and the removal rates, which tends to decrease the lifetime of chemically aged dust."

6. Following the same notion as in comment #4, Figure 1 indicates that insoluble emitted dust turns into aged-dust, followed by acid condensation. I would expect the acid condensation first and then the dust characterized as aged. Based on this schematic, there is no clear distinction about when dust is termed aged or non-aged.

Indeed. Figure 1 has been revised accordingly.

7. Page 3, line 27: "the mineral cations are used as reactivity proxy for natural aerosols, such as [. . .] mineral dust". Knowing how difficult it is to include chemical speciation of the emitted dust particles in the model, my question is how the authors apportioned the dust emitted mass to mineral cations. Is it a fixed percentage for calcium, magnesium and potassium? This information must be made clear in the text.

Yes, we follow Abdelkader et al. (2015) and use a fixed percentage for this study. This percentage has been determined in order to best match the observations of various mineral cations from EMEP and CASTNET observations. A more comprehensive treatment is under development.

8. Sections 3 and 4: as mentioned in the beginning, in a lot of parts of the discussion there is a description of the figure instead of a narrative about the main findings, followed and supported by the figures. I strongly encourage the authors to revise parts of the text accordingly, which will greatly benefit the quality of the manuscript.

Both parts have been revised.

9. What is the basis for the selection of the six specific stations that were included in the sensitivity tests, out of the ones shown in Fig. 4? It seems from fig.4 that more stations were available inside the specific zones.

Figure 4 includes the stations that have data for a longterm evaluation (2000-2012), while only the selected stations have observations for the selected period (July 2009).

10. Page 6, line 9: is the 600ug/m3 an observed or simulated value for dust concentration?.
The values refers to the model. We have added "modeled surface concentration" for clarification.

11. Page 7, lines 2-3: the aging of dust particles throughout the transatlantic transport depends also on the availability of inorganic acids in this region. The EMAC model outputs corroborate with the assumption that inorganic acids can be found in the DTA and/or DIZ zone?

Yes. The inorganic aerosol precursor gases (HCl, HNO_3, H_2SO_4) are ubiquitous, as we consider in our EMAC study various processes and anthropogenic (e.g., ships and flight traffic) and natural sources (e.g., lighting, chlorine activation of sea spray due condensation of e.g., HNO_3, H_2SO_4).

12. Table 1: I believe rm and ro are supposed to be standard deviations sigmam and sigmao. Please revise accordingly.

r_m and r_o are the geometric mean of the model and observations, respectively. We have added a description of the statistical parameters in the Appendix A.

13. Table 1: what is GFE, PF2 and PF10? They are not included in the appendix and never mentioned in the text.

GFE denotes the Growth Factorial Error, while PF2 the Fractions of points within a factor of two from the observations; accordingly, PF10, the points within a fraction of 10 from the observations. The definitions have been added to the description in Appendix A.

14. Figure 10, caption: please include the time period that the plots cover. Also, remind the reader which plots correspond to the ECMWF and TIEDTKE schemes.

The time period is now included in the figure captions.

15. Figure 11: is the standard deviation of the TRMM product calculated over the meridional mean to show the variation/dispersion of the precipitation at each longitude? Why not show the stdev for the model outputs as well?

Yes. The standard deviation of the model results has been included in Figure 11.

16. Are Figures 10 and 11 for the same time period, July 2009? If so, the meridional means are confusing. They show that B1T5 is closer to the observations but Figure 10 indicates that maybe EMAC base case is closer to TRMM.

Yes, both figures show monthly averages for July 2009. But, comparing Figure 10 and 11 is somewhat deceptive, since Figure 10 represents a qualitative comparison of the spatial distribution of precipitation and the extent of the dust plume, while Figure 11 represents a quantitate comparison, which generally is more accurate. And from Figure 11 the simulation B1T5 is closer to the observations, at least for 90-50W, while the opposite is only true for the region of 20-10W.

17. Figures 13 and 14 show monthly means for July 2009?

Yes, this is now noted in the figure caption.

18. The paper title in the supplement is not correct. Please revise accordingly.

Both changed, according to the comment of reviewer one to:
"Sensitivity of transatlantic dust transport to chemical aging and related atmospheric processes".

19. In the conclusions section, there is discussion on the findings from the sensitivity tests and model evaluation. A general conclusion about the new and significant findings from this work is necessary and, perhaps, a recommendation to the model users about the choices that would produce more reliable mineral dust simulations.

A general conclusion and a recommendation has been added to the conclusions section.

References

Abdelkader, M., Metzger, S., Mamouri, R. E., Astitha, M., Barrie, L., Levin, Z., and Lelieveld, J.: Dust–air pollution dynamics over the eastern Mediterranean, Atmospheric Chemistry and Physics, 15, 9173–9189, doi:10.5194/acp-15-9173-2015, URL http://www.atmos-chem-phys.net/15/9173/2015/, 2015.

Chemical aging of atmospheric mineral dust during transatlantic transport

Reply to Anonymous Referee #3 (doi:10.5194/acp-2016-470-RC3)

http://editor.copernicus.org/index.php/acp-2016-470-RC3.pdf?_mdl=msover_md&_jrl=10&_lcm=oc108lcm109w&_acm=get_comm_file&_ms=51800&c=112035&salt=1532899541089805197

by Mohamed Abdelkader and Swen Metzger, et al.,

December 19, 2016

We thank the anonymous referee for the in-depth comments on this manuscript. The comments and questions raised are addressed below by our point-by-point reply (black) and the revised MS.

General comments

The abstract and the body of the text are not consistent, and the text does not efficiently support the conclusions in the abstract. In fact, the abstract and the text look like parts of different papers.
There are two major results in the abstract. One result is on the pattern of dust transport over the Atlantic, which is characterized by (1) a steep and linear westward gradient due to the dust sedimentation (dry deposition) in the DTA zone and (2) an efficient removal dominated by cloud interaction and wet deposition in the DIZ zone. Another result is on the aging process of dust particles and on the effect of the aging on dust AOD in addition to the removal of the dust. About the later result, authors give the details as (1) aging of dust particles by absorbing inorganic acids changes the particles into soluble modes, enhances the absorption of water vapor, and consequently causes the increase of AOD, which the authors name as "direct effect of dust aging", and (2) aging of dust particles causes more efficient removal of particles in comparison with non-aged dust particles, and consequently results in a decrease of dust AOD, which the authors name as "indirect effect of dust aging". However, the text of results and discussion in the manuscript does not focus on the above two results.

The abstract and the discussion in the manuscript have been revised accordingly.

Here are my understandings on the text. Section 3: In the first part (Figure 2, and also Figure 3, which is somewhat a repeat of Figure 2), the simulated result (first result mentioned above) and the possible reasons for the result are simply introduced and described. As a major result of this study described in the abstract, more details and a deep discussion are necessary. My major concern on this part is the lack of a discussion on the uncertainties in the result. Another concern is that this part is not consistent with the purpose of this Section, which is to evaluate the performance of the model (the first line of Section 3). The remaining parts of this section are the evaluation of the model performance with the comparison to AERONET observations.

The text has been revised to be consistent with the purpose of this section and a note on the uncertainties of the result has been added. For a discussion on the uncertainties we refer, however, to Section 4 "Sensitivity studies", since this section is exactly about the modeling uncertainties.

Section 4: This part is an evaluation of model performance, too. First, the evaluation is conducted with a focus on the model sensitivity to emission flux and to removal mechanisms. Then the influence of different convection schemes and dust chemical aging on simulation results is examined. Although the major results described in the abstract are introduced in Section 3 and Section 4, the results are not described in a clear and compacted way. In addition, the explanations of the consistence and difference between the simulation results and the observational facts are very qualitative and the uncertainties are not quantitatively discussed.

The text has been revised such that this section 4 "Sensitivity studies", now clearly deals with modeling uncertainties (and not again of model performance evaluation).

The evaluation of the model performance is not bad and is acceptable. But the evaluation shows the quality of the model and has a weak relation with the conclusions described in the abstract.

The conclusions and the abstract have been revised accordingly.

So the contents of abstract are inconsistent with the contents of results and discussion (Section 3 and Section 4). Actually, many parts in the text of results and discussions are repeats of the paper of Abdelkader et al. (2015). The first result described in the abstract is original in this model study, but the second result contains less new information in comparison with Abdelkader et al. (2015).

The study of Abdelkader et al. (2015) presents the dust-air pollution interaction over the Easter Mediterranean, while this work focuses on a "Sensitivity of transatlantic dust transport to chemical aging and related atmospheric processes" – the new title (see our reply to referee #1). Since both studies focus on the chemical aging of dust, there is of course some overlap in the description. Otherwise this paper would not be able to stand alone. To our opinion, the overlap is small and important to have for the average reader to understand the main text flow without referring to Abdelkader et al. (2015), which an interested reader of course will/shall do.

Other major comments
The abstract is tedious and hardly followed.

The abstract has been revised.

Figure 1 is not necessary according to the abstract. The model has been described and evaluated in Abdelkader et al. (2015).

We prefer to have this paper a standalone (see our above) and, hence, we keep Figure 1.

Removal processes of dust particles by dry and wet deposition, including the subsequences of dust aging, are repeatedly applied to explain simulation results. In addition to that the repeats make the manuscript very tedious, almost all explanations lack of a discussion on the confidence of the explanations, i.e. to what a degree the explanations can account for the results. Discussions with quantitative evaluation are necessary to increase the quality of the explanations.

Redundancies have been removed and an extended discussion on a more quantitative evaluation has been included based on the statistical parameters shown in Table 1a,b of the Supplement.

The description on the wet deposition of dust particles associated with the aging of particles lacks of details and is not clear. The removal is simply described as the processes of the hygroscopic growth of aged particles (Section 4.3) and is discussed with comparisons associated with precipitation (convection) and dust emission (Section 4.2). Hygroscopic growth is a subsequence of particle aging (i.e. interaction with cloud), which is emphasized in this manuscript. However, precipitation is fundamentally governed by thermodynamic properties and the movement of air parcels (the convection: Aerosol particles are not included in the simulation of water vapor distributions by Tost et al. (2006b)). Precipitation removes dust particles via the adoption of dust particles by cloud droplets and raindrops in cloud and in below-cloud air (the effect of washout) and/or via the raincloud droplet formation on dust particles under saturate conditions in cloud or the adjacent air (the effect of nucleation scavenging). The two scavenging processes are closely dependent on the size of particles and droplets. Under saturate conditions (in cloud), dust-induced droplets (nucleation scavenging) may grow into a large droplets. But the size, rather than the composition, of a particle is the key factor for the nucleation at the size range of dust particles, usually larger than several hundred nano-meters (Dusek et al. 10.1126/science.1125261, Science, Vol. 312, Issue 5778, pp. 1375-1378). In below-cloud air under sub-saturate conditions, the growth of aged particles due to water vapor absorption is limited and the particles are not expected to frequently become considerable larger than the original particles. So the relative importance of the two processes in the dust removal needs to be clearly described and discussed in order to quantitatively show how important of the subsequence of dust aging is and how the aging enhances the removal of aged dust particles. It sounds that washout is not important for the removal of the dust particles in DIZ zone. Is this correct?.

No, the washout is of course also important for the removal of the dust particles in DIZ zone, but the chemical aging and scavenging of aged dust particles are according to our study more important in the DIZ–zone compared to DTA–zone. The text has been revised accordingly.

The definition of "direct effect" and "indirect effect" of dust aging needs to be carefully reconsidered. In this study, the effect is limited to that on AOD. However, there are many other effects associated with the aging, such as the absorption of acid gaseous species and the change of gas phase reactions. In addition, the definition may cause a confusion when readers think the "direct and indirect climate effects of aerosol particles".

We do agree that the "direct effect" and "indirect effect" of chemical aging of dust seems limited only by a definition of AOD, but it actually includes all other effects. Indeed, we try to limit the definition to the AOD, since only the net-effect AOD eventually drives the radiation. Of course, the total effect includes many other processes, such as heterogeneous reactions on dust particles, which can either increase of decrease the AOD. But, at the end of a computation step only the net-effect on AOD accounts. Therefore, we keep our definitions as introduced here.

References

Abdelkader, M., Metzger, S., Mamouri, R. E., Astitha, M., Barrie, L., Levin, Z., and Lelieveld, J.: Dust–air pollution dynamics over the eastern Mediterranean, Atmospheric Chemistry and Physics, 15, 9173–9189, doi:10.5194/acp-15-9173-2015, URL `http://www.atmos-chem-phys.net/15/9173/2015/`, 2015.

~~Chemical aging~~ Sensitivity of ~~atmospheric mineral dust during~~ transatlantic dust transport to chemical aging and related atmospheric processes

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Abstract. ~~Transatlantic dust transport~~We present a sensitivity study on transatlantic dust transport, a process which has many implications for the atmosphere, ocean and climate. We ~~present a modeling study on~~ investigate the impact of ~~the key processes (dust emissions flux, convection and dust aging parameterizations)~~ key processes that control the ~~transatlantic dust transport.Typically, the Inter-Tropical Convergence Zone (ITCZ) acts as a barrier for the meridional dust transport~~dust outflow, i.e., the emission flux, convection schemes and the chemical aging of mineral dust by using the EMAC model following Abdelkader et al. (2015) . To characterize the dust outflow over the Atlantic Ocean, we ~~address two regional phenomena~~distinguish two geographic zones: (i) dust interactions ~~with~~ within the ITCZ (DIZ) and (ii) the adjacent dust transport over the Atlantic Ocean (DTA). In the ~~DTA~~ latter zone, the dust loading shows a steep and linear gradient westward over the Atlantic Ocean~~where~~ , since particle sedimentation is the dominant removal process, whereas in the DIZ zone ~~cloud~~ aerosol–cloud interactions and wet deposition ~~predominate. To study the different impacts of aging, we present two case studies that exclude condensation and coagulation, and include dust aging at various levels of complexity. For dust aging, we consider the uptake of inorganic acids on the surface of mineral particles that form salt compounds. Calcium, used~~ / scavenging processes determines the extent of the dust outflow. Generally, the EMAC simulated dust compares well with CALIPSO observations, however, our reference model configuration tends to overestimate the dust extinction at lower elevation and underestimates it at higher elevation. The Aerosol Optical Depth (AOD) over the Caribbean responds to the dust emission flux, only when the emitted dust mass is significantly increased over the source region in Africa by a factor of ten. These findings point to the dominate role of dust removal (especially wet deposition) in transatlantic dust transport. Experiments with different convection schemes indeed revealed that the transatlantic dust transport is more sensitive to the convection scheme than to the dust emission flux parameterization.

To study the impact of dust chemical aging, we focus on a major dust-outflow in July 2009. We use the calcium cation as a proxy for the overall ~~chemically~~ chemical reactive dust fraction ~~, drives the dust-related neutralization reactions leading to~~

~~higher dust aerosol optical depth (AOD) . The aged dust particles are transferred to the soluble aerosol modes in the model and are mixed with other species that originate from anthropogenic and natural sources. The neutralization products (salts) take up water vapor~~ and consider the uptake of major inorganic acids (i.e., H_2SO_4, HNO_3, HCl) and their anions, i.e., sulfate (SO_4^{2-}), bi-sulfate (HSO_4^-), nitrate (NO_3^-) and chloride (Cl^-)) on the surface of mineral particles. The subsequent neutralization reactions with the calcium cation forms various salt compounds that cause the uptake of water vapour from the atmosphere~~and increase the dust~~ , i.e., by chemical aging of dust particles leading to an increase of 0.15 in AOD under subsaturated conditions ~~. We define the *"direct effect of dust aging"* to refer to the increase in AOD as~~ (monthly mean, July 2009). As a result of ~~hygroscopic growth~~the radiative feedback on surface winds, dust emissions regionally increased. On the other hand, the aged dust ~~is~~ particles, compared to the "non-aged" case, are more efficiently removed ~~(~~by both wet and dry ~~) because of the increase in particle size and hygroscopicity. This more efficient removal~~ deposition, due to the increased hygroscopicity and particle size (mainly due to water uptake). The enhanced removal of aged particles decreases the dust burden and lifetime, which indirectly reduces the dust AOD ~~over the DIZ zone. We define this as the *"indirect effect of dust aging"*, complementary to the direct effect that is dominant in the DTA zone. Distinction of the two agingeffects helps develop insight into the regional importance of dust–air-pollution interactions~~by 0.05 (monthly mean). Both processes can be significant (major dust-outflow, July 2009), but the net effect depends on the region and level of dust chemical aging.

1 Introduction

In the past several decades, transatlantic dust transport has gained tremendous attention because of many important impacts on Earth's climate, human health and ecosystems. North African dust transport over the Atlantic Ocean has emerged as a major contributor to the soil nutrient input to many islands in the Caribbean, the Bahamas (Muhs et al., 2007), Bermuda (Muhs et al., 2012) and in the Amazon Basin (Bristow et al., 2010; Ben-Ami et al., 2012; Abouchami et al., 2013). Dust deposition influences the oceanic and terrestrial biogeochemistry by the transport of nutrients such as iron (Ussher et al., 2013; Baker et al., 2013, 2010; Jickells et al., 2005) and phosphorus (Nenes et al., 2011) that ~~efficiently~~ dissolve into the ocean water. The emission, transport, and deposition processes of the North African dust are strongly influenced by meteorology causing strong seasonal, inter-annual and decadal variability (Mahowald, 2007; Mahowald et al., 2010). Large fractions of the dust emissions are carried across the west coast of North Africa up to the Western Atlantic (Prospero et al., 2014) and significant correlations exist between the dust and climate variables, such as sea surface temperature, the North Atlantic Oscillation (NAO), and the Madden-Julian Oscillation (MJO) (Ginoux et al., 2004; Wong et al., 2008; Guo et al., 2013). In addition, the African dust in the Sahara air-layer region influences the rates of rainfall in the Inter-Topical Convergence Zone (ITCZ) (Huang et al., 2009, 2010), and its radiative impacts ~~shifted and widened~~ can shift and widen the ITCZ northward (Bangalath and Stenchikov, 2015).

African dust is transported in great quantities to the Caribbean basin throughout the year, although the strong seasonal cycle shows the maximum transport of ~~the~~ dust in boreal summer and the minimum in winter (Prospero et al., 2014; Yu et al., 2015). The seasonality is corroborated by satellite measurements of ~~aerosol optical depth~~ Aerosol Optical Depth(AOD), which show ~~huge~~ extensive plumes of high AOD in summer extending from the west coast of Africa to the Caribbean, the Gulf of Mexico, and to the southern United States ~~(Hsu et al., 2012; Hsu et al., 2013; Hsu et al., 2014; Hsu et al., 2014; Hsu et al., 2015)~~(Hsu et al., 2012; H The satellite data also indicate that the dust transport to the Western Atlantic in winter and spring is comparable, but the dust is largely confined to the southern latitudes of Barbados with a plume axis crossing the coast of South America in the region of French Guiana and Surinam. In addition, satellite data indicate a decrease of 50% in AOD and a decrease of 0.1–0.2 in the dust-only optical depth during the transport (Kim et al., 2014). The ITCZ acts as an efficient removal mechanism (Prospero et al., 2014) and thus as a barrier to the transport of dust to the southern Atlantic (Huang et al., 2009, 2010; Adams et al., 2012). To characterize the transatlantic dust transport, many studies have used satellite observations (Liu et al., 2008; Ben-Ami et al., 2009, 2010; Adams et al., 2012; Ben-Ami et al., 2012; Ridley et al., 2013; Alizadeh-Choobari et al., 2014; Kim et al., 2014; Yu et al., 2015, among others). However, the estimation of the satellite-based dust flux has large uncertainties, primarily because of ~~uncertainties~~ ambiguity associated with the derived dust-only optical depth (Yu et al., 2009, 2013) and the dust mass extinction efficiency. Both parameters are used for calculating the dust mass loading (Kaufman, 2005).

~~Therefore, the modeling of the dust~~ One cause of uncertainty is the chemical aging of mineral dust. For instance, the condensation of inorganic acids, such as nitric acid (HNO_3), can alter the particle size due to changes in hygroscopicity of the dust particles (Metzger et al., 2006; Karydis et al., 2016) . HNO_3, which is an oxidation end product of combustion processes and lighting NO_x, and therefore ubiquitous in the atmosphere, readily reacts with the calcium of the mineral dust surface. The neutralization product, calcium nitrate, additionally takes up ambient water vapour, which can change the particle (wet) radius. This process of water uptake can become significant, since it already starts at a relative humidity as low a 50% (the relative humidity of deliquescence (RHD) of $Ca(NO_3)_2$ is 48% at T=298 K). In strong contrast, dust coating by sulphuric acid (H_2SO_4) does not lead to such hygroscopic particles, since the RHD of $CaSO_4$ is close to 100% (at any T). Thus, especially the coating by nitrates can determine the hygroscopicity of mineral dust particles in case of a polluted atmosphere (Bauer et al., 2007; Sullivan et al., 2007; Li and Shao, 2009; Tobo et al., 2009, 2010; Li et al., 2013) . The growth of the particles increases the scattering cross sections and therefore alters the AOD, it indirectly affects the cloud scavenging efficiency (Lance et al., 2013; Wu et al., 2013; Li et al., 2013) , overall potentially increasing the wet and dry removal of the dust particles (Abdelkader et al., 2015) .

Therefore, the dust cycle and the associated impacts are found to be challenging for global and regional ~~models~~ modeling, because the complex dust processes have to be parameterized using a suite of simplifications ~~(Astitha et al., 2010; Nowottnick et al., 2010; Hu~~ Although most sophisticated atmospheric models can reproduce the transatlantic dust transport plumes, ~~but~~ the patterns differ in magnitude and seasonality. Generally, the models show better performance in summer than in winter for the transatlantic dust transport (Huneeus et al., 2011). It has been observed that large uncertainties particularly exist between model simulations of the dust deposition (wet and dry) (Schulz et al., 2012). The atmospheric models that are applied in the AeroCom model intercomparison activity (http://aerocom.met.no/) show that the mean normalized bias of the AOD ~~model~~ varies within a wide range from –0.44 to 0.27 ~~(Huneeus et al., 2011)~~ Huneeus et al. (2011)), which is caused by large discrepancies in the dust-related processes (emission~~and~~ , horizontal and vertical distributions and the parameterization of chemical aging) that affect the dust transport from Northern Africa over the Atlantic ocean (Prospero et al., 2010). This indicates that in ~~present~~ these models the dust removal is very efficient during the transatlantic ~~dust~~ transport (Kim et al., 2014) and that the development of the model requires ~~a more~~ comprehensive representation of the dust related processes. Though the incorporation of ~~the~~ satellite products helps ~~in improving the modeling~~ improving the model results, a deeper understanding of the key factors that determine the transport of the dust is also required. This study aims at examining the factors that can affect the transatlantic dust transport~~by explicitly considering the~~, i.e., the emission flux, convection schemes and the chemical aging of ~~the mineral dustduring long-range transport in a state-of-the-art atmospheric chemistry climate modelsetup.~~ mineral dust, by using the EMAC model.

2 Model Description

We ~~have used~~ use the EMAC (~~The~~ ECHAM5/MESSy2 ~~atmospheric chemistry General Circulation Model) in a setup~~ Earth System Model) following Abdelkader et al. (2015). The EMAC model describes the tropospheric and middle atmosphere processes and their interactions with land and oceans considering various submodels (Joeckel et al., 2010) – those used in this study are listed in Table 1. The mineral dust particles are emitted in two log-normal distribution modes (accumulation and coarse) with median diameters of 0.5 µm and 5.0 µm and a modal standard deviation of 1.59 and 2.0 for the accumulation and coarse modes respectively (Abdelkader et al., 2015). The anthropogenic emissions are based on the EDGARv4.0 inventory (Pozzer et al., 2012) and ~~include~~ includes the greenhouse gases, NO_x, CO, non-methane volatile organic compounds (NMVOCs), NH_3, SO_2, black carbon (BC) and organic carbon (OC) from fossil fuel and biofuel use. The monthly large-scale biomass burning emissions of OC, BC and SO_2, are based on GFED version 3 (Global Fire Emissions Database) (van der Werf et al., 2010). The emissions drive a comprehensive atmospheric chemistry mechanism (Sander et al., 2005), which calculates major inorganic acids (H_2SO_4, HNO_3, HCl) online with meteorology. Organic acids are not considered in this model setup since their concentrations over Sahara during dust outflow are very low, ~~however~~though, many modeling studies reported the uptake of organic acids by dust particles ~~(Metzger et al., 2006; Möhler et al., 2008; Liu et al., 2013; Alexander et al., 2015; Wang et al., 2015; Alexander et al., 2015)~~

The chemical aging of the dust depends on the condensation of inorganic acids and the associated uptake of water vapor. ~~Considering inorganic acids increases the level of dust aging, water uptake, particle size , removal rate and eventually may further decrease the dust lifetime~~ This increases the dust particle mass, particle size and the removal rates, which tends to decrease the lifetime of chemically aged dust. The condensation of acids ~~yields anions, i.e.,~~ in our model yields the anions sulfate (SO_4^{2-}), bi-sulfate (HSO_4^-), nitrate (NO_3^-), and chloride (Cl^-), whereas the condensation of ammonia (NH_3) yields a semi-volatile cation, ammonium (NH_4^+), that reacts with the inorganic anions in competition with the mineral cations Na^+, Ca^{2+}, K^+, Mg^{2+} (Metzger et al., 2006). However, in this study the cations are considered as reactivity proxy for natural aerosols, such as sea salt, biomass burning, or mineral dust~~. –~~, where we follow Abdelkader et al. (2015) and use a fixed percentage. These fractions have been derived from a comprehensive sensitivity study (which will be presented in a separate study) to achieve the best agreement of the cation and anion concentrations with various station observations for the period 2000-2012 (see Section 3). The anion–cation neutralization products (salt compounds), simulated by the ~~ISORROPIA-II aerosol thermodynamics model (Fountoukis and Nenes, 2007)~~ aerosol thermodynamic models, ISORROPIA-II (Fountoukis and Nenes, 20 EQSAM4clim (Metzger et al., 2016) , can alter the hygroscopicity of the atmospheric dust particles, but the effect strongly depends on the atmospheric residence time, region ~~, and concentration~~ and concentrations of acids. Generally, dust chemical aging changes the solubility, which controls the water uptake and in turn alters the aerosol size distribution (Metzger et al., 2006). The ~~latter~~ water uptake is a key parameter and important for aerosol-radiation feedback, aerosol in-cloud processing (nucleation scavenging), and below-cloud (impaction) scavenging. ~~Our~~ The EMAC scavenging processes include detailed pH-dependent aqueous phase chemistry (Tost et al., 2006a) which is fully coupled with the aerosol and gas-phase chemistry, liquid cloud water ~~,~~ and ice crystals. In addition to the aerosol hygroscopic growth and scavenging, the dust size distribution can change by coagulation, and smaller particles can grow into larger sizes for both the soluble and insoluble aerosol modes (Pringle et al., 2010)~~. Aerosol~~ , whereas aerosol hygroscopic growth is only allowed in the soluble modes (Abdelkader et al., 2015). Dry deposition and particle sedimentation can remove all particles from the atmosphere depending on the particle size (Kerkweg et al., 2006a). Thus, the representation of the dust cycle in our EMAC setup couples the dust emissions, loading, and lifetime with the radiative forcing and model dynamics. As a result, changes in the dust loading feed back to the surface wind speed, soil moisture, cloud formation and precipitation, and in turn the dust emission flux. Overall, the level of air pollution controls the dust cycle because it determines the level of dust chemical aging by inorganic acids and water vapor. A Newtonian relaxation approach is used to nudge the model meteorology in the free atmosphere (i.e., above the boundary layer) to achieve a realistic simulation of the surface wind speed and tracer transport (Abdelkader et al., 2015). Nudging significantly improves the surface dust mass concentration over the Caribbean compared with dust observations (Astitha et al., 2012). The model spectral resolution is T106 ($\approx$ 110 km) and for the longterm simulations it is T42 ($\approx$ 280 km). Both model resolutions use 31 vertical levels. Figure 1 summarizes the representation of the dust cycle and ~~air-pollution-dust-aging-radiation~~ air-pollution-dust-chemical-aging-radiation feedbacks in our EMAC model setup.

3 Long-term evaluation

This study aims at examining the key factors that affect the transatlantic dust transport ~~considering a~~ for a major dust-outflow event in July 2009 with a model resolution of T106, which is presented in Section 4. Before we focus on the sensitivity study, we present in this section the key findings of a comprehensive model evaluation, which was performed for the period 2000–2012 with a ~~model~~ coarser resolution of T42~~($\approx$ 280)~~. For the ~~model~~ long-term evaluation, we use the following satellite and ground station AOD products:

- AErosol RObotic NETwork (AERONET) ~~(Holben et al., 1998)~~ – Holben et al. (1998) ;
- Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) ~~(Winker et al., 2009, 2007)~~ – Winker et al. (2009, 20
- MODerate resolution Imaging Spectroradiometer (MODIS) platforms Aqua and Terra (product collection 6, L3 gridded data~~, Kaufman et al. (1997))~~) – Kaufman et al. (1997) ;
- Precipitation data from Tropical Rainfall Measuring Mission (TRMM) (product version 31 L3 gridded data~~, Diner et al. (1998))~~) – Diner et al. (1998) ;

Figure 2 shows the seasonal average of the ~~modeled~~ simulated dust burden and the precipitation rate over a 13-year simulation period. Both the dust burden and the precipitation rate peak during the summer season (JJA), where the dust plume is located relatively far north from the equator, in agreement with remote sensing observations (Prospero et al., 2014; Yu et al., 2015). During the winter season (DJF), the dust burden and the precipitation rate show a minimum, whereas during the spring season (MAM), the dust plume and the ITCZ are shifted southward. In winter and spring, the dust transport shifts southward to 0°-10°N and affects South America significantly, whereas during summer, the dust transport occurs predominantly at 10°-20°N, substantially affecting the Caribbean (Yu et al., 2015). During boreal winter the enhanced precipitation over the Northern part of South America results in higher and localized dust scavenging because the precipitation along the dust transport from the Western Africa into the Caribbean is at minimum. In contrast, during boreal summer, the dust spreads to a larger extent into the ITCZ because of the stronger ~~dust emissions (Prospero et al., 2014) and is associated with~~ emissions (Prospero et al., 2014) while it is subject to enhanced dust scavenging. The strong southward gradient of the dust burden ($\approx 100\,\mathrm{mg\,m^{-2}\,deg^{-1}}$) is collocated with precipitation in the western part of the Sahel and the ITCZ region. During the winter months, dust is primarily scavenged over Southeast America. As a result, the extent of the dust outflow is primarily ~~affected by the~~ controlled by precipitation in the ITCZ region. Figure S1 in the Supplement shows the dry and the wet removal of the dust particles. It shows that the dry removal dominates the northern part of the dust outflow region, whereas the wet removal dominates the southern part.

To indicate the region where the dust interacts with the ITCZ, we introduce the dust-ITCZ (DIZ) zone ~~. In this region, for~~ which is shown in Figure 3 – the DIZ is marked by a blue line, and the AERONET station locations used to evaluate the simulated AOD are included. In the DIZ region, the transatlantic dust transport ~~, the dust and cloud interactions are more important~~ is controlled by dust–cloud interactions and the dust scavenging is ~~very efficient. We~~ most efficient. Accordingly, we refer to the region ~~where the sedimentation and the dry deposition dominate the dust removal as the adjacent dust transport over the Atlantic Ocean (DTA)zone.~~

~~Figure 3 shows the DIZ and DTA zones, and the AERONET stations used in this study to evaluate the modeled AOD. The area bounded by the blue line represents the area where the dust interacts with the ITCZ. This region largely controls the transport of the dust into Caribbean (??) , and therefore in this study it is introduced as the DIZ zoneto illustrate the modeled transatlantic dust transport~~of the pre-dominant dry removal process (sedimentation), as the DTA zone.

~~Figure 4 summarizes~~ Summarizing the long-term evaluation results~~for the transatlantic dust transport. The figure compares the modeled AOD with the AERONET observations and shows~~, Figure 4 shows: (i) the transatlantic dust transport region with skill scores at each station (see Appendix A for the evaluation metrics), (ii) the time series of the six selected stations that provide long-term data with three stations each in the Caribbean (left) and around ~~the Western~~ West Africa (right)~~(for the station locations see Figure 3)~~, and (iii) the corresponding scatter plots for both sides of the Atlantic Ocean. Table 2 summarizes the model performance for both regions over the entire period (2000–2012). ~~The~~ Generally, the simulated AOD compares well with the AERONET observations – the 13-year average (based on 5 ~~hours intervals~~hourly output) of the ~~modeled~~ simulated AOD for the Western Africa sites is 0.16±0.27 (one standard deviation), which is ~~comparable to~~ slightly lower than the observation of 0.24±0.37. ~~However, the~~ The difference is larger compared ~~with the Caribbeansites, which is represented~~

~~by an average modeled AOD of~~ to that for the Caribbean, for which the average simulated AOD is 0.12±0.18 and ~~with~~ 0.14±0.22 ~~by~~ according to the observations. At both sides of the Atlantic, the lower variability of the model is primarily a result of the relatively coarse model resolution (280 km), which was used for these long-term simulations because of ~~higher computation cost of the higher~~ significantly larger computational burden for the higher T106 resolution. The skill score (SS1 ~~(Taylor, 2001)~~ Taylor (2001)) has a value of 0.73 and 0.70 for the Western Africa and the Caribbean ~~sites~~stations, respectively. ~~A rather good comparison is shown by~~

Additionally, the correlation coefficients (R) (Table 2) compare reasonably well, although the values are lower because R is more phase sensitive than the SS1 ~~,~~ (i.e., more sensitive to time lags between ~~modeled~~ simulated and observed AOD). The higher R value for ~~Western~~ West Africa (0.61) compared with ~~the~~ Caribbean (0.41) mainly results from the overall higher ~~dust AOD contribution~~ contribution of dust AOD to the total AOD~~, compared with the Caribbean~~. Typically, the Caribbean is strongly influenced by the uncertainty ~~of the~~ associated with long-range transport and the ~~associated dust aging (~~dust chemical aging, with potential failures causing a time shift of ~~the~~ dust peaks during the transport~~)~~. These differences are~~best described by time series. Overall,~~ , however, best revealed by the station time series (Figure 4). This shows that the model captures the variability of AOD at all stations, ~~but around Western~~ and only around West Africa the model slightly underestimates the AOD peaks, especially at Dakar which is ~~on~~ at the edge of the DIZ zone. Over the Caribbean, the model seems to agree slightly better~~but it generally~~ , while it generally somewhat underestimates the AOD during the dust outflow periods, e.g., seen at the AERONET station La Parguera. This underestimation could be ~~either caused by an insufficient representation of the~~ realted to the representation of dust emissions and ~~the~~ related processes in the source region of the ~~Western~~ West Africa (Huneeus et al., 2011; Shao et al., 2011; Cuevas et al., 2015), ~~an~~ by overestimated removal during ~~the~~ transport (Schulz et al., 2012; Prospero et al., 2014), or ~~insufficient~~ due to low-biased dust transport from the boundary layer into the free atmosphere (Khan et al., 2015). In addition, the underestimation of ~~the~~ AOD could be also due to the missing ~~large coarse~~ fraction of giant mode particles (larger than 10 µm)~~in the model, which could lead to the~~ , which may contribute to an underestimation of AOD ~~on the Western Africa side whereas these large~~ near the dust source region. However, giant particles are not transported far over long distances ~~. In this study , we focus on the sensitivity of the~~ and hence not really relevant for the long-range transport and our sensitivity study on the emission flux and ~~the removal mechanisms, whereas the effect of large coarse particles and their radiative forcing is a subject of future study~~removal mechanisms.

4 Sensitivity studies

To ~~resolve the impact of various~~ study the key factors that ~~control the dust modeling~~may affect the transatlantic dust transport, we focus ~~on a period with relatively strong dust outflow that occurred during July2009 with a surface concentration up to 600at Dakar~~in this section on a major dust-outflow event that occurred in July 2009. We study the impact of various key factors with a relatively high model resolution (T106). The dust outflow is, in terms of AOD ,close to most observations, as indicated by monthly ~~mean AOD observations at Dakar and Capo Verde (~~means highlighted by the red bar in Fig. 4~~). For this month, the model and observations show a relatively large difference and therefore this period is~~ . However, near the source region at Dakar and Capo Verde, the AOD observations are underestimated for this month. During this period a major outflow event occurred, and therefore it seems suitable to test various model parameters~~that may affect the transatlantic dust transport~~, i.e., (a) the dust emission flux, (b) the ~~convective~~ convection parameterization, and (c) the level of dust chemical aging. ~~The sensitivity studies conducted using EMAC are all based on a higher spectral resolution than the long-term evaluation (i.e., T106, or ≈ 110). Table 1a and table 1b in the supplement show the evaluation metrics for the AOD for sensitivity studies over West Africa and the Caribbean, respectively.~~

~~During the transatlantic dust transport, the ITCZ represents a strong barrier for the dust outflow and therefore controls the meridional extent of the dust plume (Yu et al., 2015) . The ITCZ acts as a major sink that depends on the amount of precipitation (Prospero et al., 2014; Schlosser et al., 2014) and the removal might be enhanced depending on the dust aging (Abdelkader et al., 2015) .~~ Figure 5 shows the dust burden and the total mean precipitation for July 2009 ~~using~~ from the reference EMAC simulation, which includes the dust cycle and chemical aging as shown in Fig. 1. The simulated dust surface concentration reaches on average up to 600 $\mu g\,m^{-3}$ at Dakar, indicating that the model captures in principle the strong outflow event. Generally, two strong precipitation areas are visible with one peak centered at 15°W with a monthly average of 20 $mm\,day^{-1}$, i.e., one at the coast of West Africa and the other peak area is located in the Caribbean at 50°W. with a monthly average of 25 $mm\,day^{-1}$. These precipitation maxima influence the dust loading. ~~The precipitation~~ During transatlantic dust transport, the ITCZ represents a strong barrier for the dust outflow and therefore controls the meridional extent of the dust plume (Yu et al., 2015) . The ITCZ acts as a major sink that depends on the amount of precipitation (Prospero et al., 2014; Schlosser et al., 2014) and the removal might be enhanced depending on the dust chemical aging (Abdelkader et al., Clearly, the precipitation within the ITCZ coincides with the steep gradient of the dust burden in the meridional direction over the Western Africa. Along the zonal extent of the dust plume, the collocation of the dust plume and precipitation ~~indicates~~ corroborates that the meridional extent of the dust is primarily ~~affected~~ controlled by the location of the ITCZ. Fig. S1 in the Supplement summarizes the monthly average dust removal during July 2009. Table 1a and table 1b in the supplement additionally show some evaluation metrics for the AOD of the sensitivity study over the West African and Caribbean stations.

Typically, African dust outflow reaches the Caribbean ≈ 5 days later (Gläser et al., 2015) and the surface dust concentration is significantly lower at the Caribbean side compared to Western Africa. Figure 6 shows the time series of the size-resolved surface dust concentrations. Two main dust outflows on the 2^{nd} and 12^{th} July are simulated at the Capo Verde station, indicated by dust concentrations higher than or close to 300 $\mu g\,m^{-3}$ (equivalent particle cutoff diameter of 5 µm) and another weaker dust outflow is simulated on 24^{th} July, indicated by a lower concentration peak around 100 $\mu g\,m^{-3}$. The former two dust outflows are seen at Dakar with twice the concentration (up to 600 $\mu g\,m^{-3}$) at slightly different ~~times~~ time periods due to different transport. ~~These dust outflows reach the Caribbean ≈ 5 days later,~~ Eventually, the dust outflow reaches the Caribbean with a significant lower ~~surface~~ concentration of around 60 $\mu g\,m^{-3}$ ~~.~~ at the earth surface.

Despite chemical aging, the model ~~concentrations show~~ simulates a majority of the dust particles in the insoluble coarse (ci) mode, which indicates that the dust particle concentration is high ~~or the level of inorganic acids is low, not allowing for complete~~ and/or the inorganic acids concentration is relatively seen too low for complete chemical aging. This is especially valid for ~~high dust outflows~~strong dust outflows, such as studied here. On the other hand, the fraction of the aged dust, i.e., the ratio of the coarse mode soluble to insoluble particles (cs/ci), is somewhat higher in the Caribbean because of the continuous chemical aging during long-range transport. The aged dust fraction over West Africa is about 10% of the total dust mass and twice ~~of~~ that at the Caribbean sites. The same is true for the dust in the accumulation modes (ai and as), but the mass concentrations are an order of magnitude lower compared ~~with~~ to the coarse mode ~~concentration~~ concentrations and therefore they are not ~~visible~~ discernable at the linear scale. At higher elevations, this fraction can be different because of different dust and precursor gas concentrations.

To investigate the vertical distribution, the ~~modeled~~ simulated dust extinction is compared with the dust subtype classification of the CALIPSO retrievals. Figure 7 shows a comparison for the second dust outbreak on 12 July 2009. The figure shows a subset of four collected CALIPSO tracks and includes a qualitative comparison of the dust layer height. The scatter plot attached to each panel represents the point-to-point comparison, colored by the height of each observation point whereas the area plots show the dust burden interpolated in time to the CALIPSO overpass time~~which is~~ , indicated by a solid black line. Additional CALIPSO tracks are shown in Fig. S2a–Fig. S2e in the Supplement. Both EMAC and CALIPSO show that ~~the~~ dust over the Sahara reaches an elevation up to 7 km. The dust burden is very low (as indicated by the area plot) south of 10°N, which coincides with a very low AOD observed by CALIPSO. Both EMAC and CALIPSO ~~shows~~ show that the dust plume is limited to the area between 14°to 22°N and the top of the dust layer is ~~reduced~~ lowered to 5 km ~~in~~ over the middle of the Atlantic. This is primarily a result of the prevailing deposition (gravitational settling + wet removal), which is further discussed in the following sections. Once the dust reaches the Caribbean, the plume spreads over a considerably larger area, which extends from 5°to 28°N as a result of ~~change~~ changes in meteorological conditions. The dust plume eventually reaches the Caribbean with a top layer height of $\approx$ 5 km. In Fig. 7, the comparison with CALIPSO (and Fig. S2a–Fig. S2e in the ~~supplement~~Supplement) shows that the model ~~nicely~~ captures the vertical structure of the dust outbreak during the transport over the Atlantic Ocean. Nevertheless, the model tends to systematically overestimate the dust extinction at lower altitudes, whereas at higher altitudes the model tends to underestimate the CALIPSO extinction (considering all CALIPSO tracks in Fig. 7 and Fig. S2a–Fig. S2e in the Supplement). This indicates that EMAC might remove the dust too efficiently during transport. The reason can be manifold and ~~caused by different insufficiently represented~~ related to different processes of the dust cycle (Figure 1). ~~Next~~Therefore, the key factors are investigated further in greater detail.

4.1 Dust emission flux

A successful representation of the dust cycle first of all depends on an accurate dust emission flux. However, the ~~correctness of the modeled~~ simulated emission flux critically depends on many model parameters, where some of them are resolution dependent. Using EMAC, the dust emissions are calculated considering the ~~frictional~~ friction velocity following Astitha et al. (2012). To test the sensitivity of the transatlantic dust transport to the dust emission parameterization, several sensitivity simulations were performed, which are summarized in Table 3. The ~~dust mass~~ total dust mass, emitted during July 2009 within the region between 20°W to 10°E and 15°N to 30°N, is 0.6133 $\mathrm{kg\,m^{-2}}$ for the reference case~~is 0.6133~~ .

The first test case (B1E1) represents a redistribution of emission bins between the coarse and accumulation modes so that dust particles are shifted from the coarse to the accumulation mode while conserving the total dust mass. In this case, a ~~higher~~

larger amount of dust in the accumulation mode is transported over ~~larger~~ extended distances compared with the reference case "EMAC". "EMAC" considers the same total dust mass with a larger fraction in the coarse mode. Additional sensitivity runs, B1E2 to B1E7, change the total dust emission flux by increasing the emission flux according to different factors shown in Table 3. The horizontal dust emission flux is described by Eq. 1 (Marticorena and Bergametti, 1995; Astitha et al., 2012)

$$H = \frac{c\rho_{air} u_*^3}{g}\left(1+\frac{u_t^*}{u^*}\right)\left(1-\frac{u_t^{*2}}{u^{*2}}\right), u^* > u_t^* \quad (1)$$

With the tuning parameter c = 1 representing the reference case "EMAC" following (Darmenova et al., 2009; Astitha et al., 2012), g is the gravitational acceleration, ρ_{air} the air density, u^* the friction velocity, u_t^* the threshold friction velocity. For case B1E8, the horizontal mass flux is increased by a factor of 2.6 ~~– as this is another "tuning" factor for the emission scheme~~ (parameter c in Eq. 1). The cases highlighted in Table 3 are shown in Fig. 8, whereas the other cases are shown in the Supplement (Fig. S3).

Due to the different dry and wet deposition characteristics of the accumulation and coarse mode particles, significant differences ~~might be~~ are expected. Figure 8 shows that the AOD time series at the selected AERONET stations are rather insensitive to the emission flux modifications except for case B1E3 (and B1E4, which is shown in the Supplement). This is valid for both sides of the Atlantic, where the AOD at the Caribbean stations seems even less sensitive than the AOD for the ~~Western Africa~~ West African sites. Only for the cases where the coarse mass flux is significantly increased (factor of 5.3), the AOD shows a higher sensitivity. The large increase in the coarse mode mass for case B1E3 results in a significant increase in AOD (exceeding 2.0) on both sides of the Atlantic Ocean. Case B1E8 (modification of the horizontal mass flux) shows better agreement with the AERONET observations at both sides of the Atlantic Ocean despite the very high AOD values obtained on 21 July at Saada station. The model captures the AOD during the two dust outflow events (2 July and 12 July) at Capo Verde as well as the first dust outflow at Saada on 4 July. For the Caribbean sites, case B1E8 shows the best agreement with AERONET for the three stations.

The sensitivity simulations show that the accumulation mode fraction of the dust contributes much less to the AOD on both sides of the Atlantic Ocean because even an increase by a factor of 5.3 in the dust emission flux is not sufficient to match the observations. Instead, such an increase (by a factor of 5.3) in the emitted dust mass flux results–regionally and globally–in an unreasonable dust budget shown by Astitha et al. (2012). On the other hand, this sensitivity study shows that the AOD is more sensitive to the dust mass in the coarse mode and that the AOD over the Caribbean is much less sensitive to the total dust emission flux. Clearly, the model sensitivity is higher for the West African sites because these AOD results are ~~primarily~~ more directly controlled by the Saharan dust outbreaks. To match the elevation at which this outflow occurs is ~~also importantas the~~ equally important. The comparison with the CALIPSO observations ~~showed~~ (Fig. 7) ~~providing~~ reveals that EMAC overestimates the dust extinction at lower elevations~~whereas it underestimates~~, whereas the values at higher elevations ~~. Instead, for long-range transport, this~~ are underestimated. This finding points to the strong contribution of ~~the dust removal on the~~ dust removal during transatlantic dust transport, and is largely controlled by the convection scheme.

4.2 Convection schemes

The scavenging of dust particles by precipitation is another key factor that controls the transatlantic dust transport (Kim et al., 2014). In order to study the impact of the convection and the associated precipitation during the dust outflow, different convection schemes ~~that are implemented by Tost et al. (2006b) in the EMAC model~~ (implemented in EMAC by Tost et al. (2006b)) are compared. ~~Different convection parameterization schemes have been evaluated and the~~ The default scheme (TIEDTKE convection with NORDENG closure) provides realistic water vapor distributions on the global scale, which is crucial for radiative transfer processes and atmospheric chemistry (Tost et al., 2006b, 2010; Rybka and Tost, 2013). However, the radiative effect of aerosols has not been considered in these studies. Table 3 ~~presents~~ includes the sensitivity tests by using ~~the available convective schemes of~~ several convective schemes available in the EMAC model. The principal cases are shown in Fig. 9, whereas the other cases are shown in the Supplement (Fig. S4).

Figure 9 ~~demonstrates that~~ depicts the AOD time series for the stations shown in Fig. 3 ~~are more sensitive~~ and shows a larger sensitivity to the convection ~~parameterization than~~ compared to the emission flux parameterizations (Sec. 4.1). ~~For the convection parameterization~~In particular, the AOD is more ~~sensitive over West Africa~~ sensitively influenced over the West African than over the Caribbean sites, which is primarily a result of the decreasing dust burden due to the removal of the dust during transport (Fig. 6). Generally, the AOD is underestimated at all stations, except for Saada, ~~for~~ in the reference simulation (EMAC)~~, which is significantly improved for~~ . During the period 20–25 July 2009, this significantly improves in the sensitivity simulations (B1T3 and B1T5)~~during the period 20–25 July 2009. During this period~~. However, the model also simulates a dust outflow ~~, which is not shown by AERONET observations. Over~~ event that is not observed by the AERONET stations. Overall, over the Caribbean, case B1T5 (ECMWF operational convection scheme) ~~shows better~~ yields the best results for all dust outflow events. ~~Generally, the~~ The main differences between the schemes appear in the tropical region, ~~and~~ while the maximum difference is obtained during ~~the~~ boreal summer. For these conditions (location + time), the EMAC reference setup ~~shows the maximum difference~~ is associated with relatively large discrepancy in the precipitation amount (Tost et al., 2006b). As a result, the scavenging of aerosols, including dust particles, is overestimated due to the high precipitation rates. Consequently, this over-removal of the dust results in an underestimation of the AOD over the Caribbean.

~~To illustrate this finding,~~ Figure 10 ~~shows (from left) the~~ illustrates this finding. The total cloud fraction, precipitation, dust surface concentration, and the dust burden (monthly mean) ~~for~~ are shown for the different convection parameterizations in comparison to MODIS cloud fraction and TRMM precipitation. In general, the model ~~exhibits~~ reproduces the main features of the cloud cover observations; however, ~~the~~ EMAC (reference~~setup) model underestimates the~~) underestimates cloud cover over the Atlantic Ocean. Over the tropical areas in Africa, B1T5 (ECMWF) leads to more realistic results compared to MODIS ~~relative~~ and compared to B1T4 (~~shown in the~~ also ECMWF but with shallow convection closure, shown in Fig. S5 in the ~~supplement)simulation (for this region and season)~~Supplement). Over the ~~Ocean~~ocean, B1T5 considerably underestimates ~~the~~ cloud cover and ~~the precipitation rate that has limited impact on the dust transport~~precipitation rates. Over the Caribbean sites, B1T5 overestimates ~~the~~ cloud cover, whereas the other schemes ~~show better~~ produce more realistic results. On the other hand, the calculated precipitation (second column) generally shows an overestimation for all schemes except B1T5 ~~that shows~~ with an underestimation over the ocean. As a result of the differences in the cloud cover and precipitation rates, ~~the model shows a different magnitude~~ all model simulations show different magnitudes of the dust plumes (third and fourth columns) which is ~~more~~ most pronounced for the dust burden. For the reference simulation (EMAC in Table 3 and Figure 10), the dust plume extends to 60°W with a dust burden of 200 mg.m^{-2}, whereas for simulation B1T3 (TIEDTKE) the same dust burden is obtained at 80°W and westwards. The difference in the dust plume magnitude merely results from different removal efficiencies because of different precipitation rates.

For a quantitative comparison, the average meridional dust burden in the dust outflow over the Atlantic Ocean region (10°– 25°N) is shown in Fig. 11 for different convection parameterizations~~, together with the precipitation (middle panel)~~. Additionally, the precipitation and the column averaged aged dust proxy (ADP)~~(bottom panel), is shown in Fig. 11~~, which was introduced by Abdelkader et al. (2015), are included. The ADP simulations, which represents the ratio between aged and non-aged dust particles, indicates the level of dust ~~aging (~~chemical aging (i.e., the mass fraction of the aged to the total dust mass). A zero ADP value indicates ~~no aged dust particles (~~"pristine" or freshly emitted ~~particles in the insoluble mode~~insoluble particles (no aging), whereas a value of one indicates that all dust particles are ~~considered to be aged (all particles are coated and present in the soluble dust mode~~chemically aged (fully coated and transfered from the insoluble to the soluble modes).

~~The~~ First, the dust burden shows a very steep gradient westward over the Atlantic Ocean~~because of the~~. This is mainly a result of dust removal by deposition (sedimentation and scavenging mechanisms) during ~~the~~ long-range transport. Over the Atlantic (within DTA), ~~the~~ this gradient is linear in the logarithmic scale, whereas the gradient is nonlinear over the Western and Eastern Atlantic (especially within DIZ). The dust burden over West Africa (~~eastern to~~ east of 10°W) ~~shows roughly~~ is about 1000 µg m^{-3} ~~and~~ but declines to 50 µg m^{-3} over the Caribbean. The different parameterization schemes show more than a factor of 2 difference between the dust burden over Western Africa~~and~~, and about a factor of 3 over the Caribbean. This is primarily a result of different precipitation rates and ~~different associated~~ the associated differences in dust removal. The two precipitation peaks (over Western Africa and the Caribbean), ~~as~~ shown in Fig. 5, ~~can also be~~ are also seen in Fig. 11~~, but to a lesser extent~~. They are, however, weaker because the averaging is performed over a wider area (dust plume) that is not associated with precipitation. The higher precipitation rate over the western and eastern parts of the Atlantic results in ~~an~~ enhanced dust scavenging. Over the Atlantic, the precipitation is lower and therefore the removal by sedimentation is ~~relatively stronger~~ stronger during July 2009 (≈2 g m^{-2} compared to ≈0.2 g m^{-2}~~during July 2009), whereas the~~, respectively). The elevated precipitation over the Caribbean ~~shows the maximum dust depositiondue to scavenging~~causes maximum wet deposition. As a result, the dust burden is an order of magnitude lower over the Caribbean compared ~~with~~ to West Africa. In addition, there is a clear ~~anticorrelation~~ anti-correlation between the dust burden and the precipitation amount over both sides of the Atlantic. The comparison of precipitation with TRMM observations ~~shows~~ reveals that the EMAC model gives more realistic results over West Africa compared with the Caribbean for all convection schemes.

~~The~~ Second, the ADP (Fig. 11) illustrates the effect of convection schemes on the transatlantic dust transport~~and shows the highest sensitivity for the convection parameterization~~. Over West Africa, the dust is already aged with ADP values between 0.2 and 0.4, whereas over the Caribbean the ADP values are ~~higher ranging between~~ with 0.3 and 0.5 only slightly higher. The lower ADP values over West Africa ~~indicate~~ can be attributed to the higher dust loadings, which ~~require a~~ requires a much larger amount of condensable material to ~~age~~becomes fully aged. Over the Caribbean, the dust loading is ~~much~~ considerably lower due to ~~removal during the transport which is~~ the removal processes along dust transport, which takes about 5 days~~, for instance Gläser et al. (2015) , long enough~~. This time is sufficiently long for coating by acids and other soluble materials ~~which cause~~ (Gläser et al., 2015) , and causes the dust to become more aged~~(ADP = 0.6) compared to the Western African side (ADP=0.35). The~~. On the other hand, the high precipitation amount at 15°W over the Western Africa region results in higher scavenging of the aged dust particles compared with the ~~"pristine" (nonaged~~–pristine– (non-aged) dust particles~~and~~. This results in a decrease in the ADP values~~that are~~, in agreement with the results of Abdelkader et al. (2015). ~~Western to~~ West of 15°W, the dust is transported over the Atlantic ~~at~~ into a region where ~~the precipitation is~~ precipitation is much lower (middle panels). ~~This results in an increase in the aging levels.~~ Consequently, the level of chemical aging increases. The EMAC reference simulation (with ~~higher precipitation) shows~~ too strong precipitation) therefore shows a higher ADP (0.35 compared to 0.2)~~values as~~, which is a result of the lower dust burden ~~, which is caused by~~ and mainly caused by a too efficient wet removal.

~~The~~ Thus, the convection sensitivity analysis ~~indicates a very strong removal of the dust during transatlantic transportwith the EMAC~~ points to a too strong removal mechanism of the mineral dust particles along transatlantic transport, when the default convection scheme ~~, which is indicated by the underestimation of the AOD over the Caribbean~~is used in EMAC. In addition, the level of dust ~~aging controls~~ chemical aging seems to control the efficiency of dust scavenging. Higher levels of aged dust, and higher precipitation amounts, significantly decrease the dust burden and thus the AOD over the Caribbean. This ~~suggests that improving~~ further suggests that modeling the transatlantic dust transports requires improved convection parameterization ~~and~~ (i.e., more realistic precipitation rates~~in parallel with the improved dust~~), and in parallel a realistic representation of dust chemical aging.

4.3 Dust chemical aging

~~The level of dust aging depends on the availability of inorganic acids, i.e., volatile and semivolatile compounds.~~ To further investigate the impact of the dust chemical aging on the transatlantic dust transport, ~~dust aging~~ this process was excluded for an additional sensitivity study. ~~For this~~ The level of dust chemical aging depends on the availability of condensable acids (see Sec. 2). For the "No Aging" case, the condensation of acids on insoluble dust particles is excluded, which suppresses water uptake by dust particles. Figure 12 shows the AOD time series at the AERONET stations on both sides of the Atlantic for the two cases, i.e., ~~"Aging " and "No Aging"~~Aging and No Aging. Generally, the ~~"Aging "~~ Aging case systematically shows a higher AOD ~~as compared with the "No Aging "~~ compared to the No Aging case, which emphasizes the importance of this process and the associated water uptake in agreement with the results of ~~Pozzer et al. (2015) . The dust~~ Abdelkader et al. (2015) . However, the dust chemical aging has a stronger impact on the AOD over ~~Western~~ West Africa, especially at the Capo Verde and Dakar stations during the two dust outbreaks discussed above. The Aging case shows about 0.2 higher AOD compared with the ~~"No Aging "~~ No Aging case as a result of the larger particle size and the associated water uptake. This increases the scattering cross section and thus the AOD. Over the Caribbean, the dust chemical aging shows a smaller impact on the AOD; the ~~"Aging "~~ Aging case shows only about 0.05 higher AOD because of the lower contribution of the dust to the overall AOD values (which includes the contribution of other aerosol species, sea salt, etc., for instance). During the high dust outbreaks, the concentration of the soluble compounds required to coat such a large amount of dust is not available according to the EMAC model. The aged dust particles are removed more efficiently during transport and relatively more uncoated dust particles reach the Caribbean. As a result, the dust chemical aging has a limited effect on the AOD over the Caribbean AERONET stations.

Figure 13 shows the regional difference (monthly mean) for (a) the dust burden, (b) AOD, (c) dust emissions averaged over the region from 18°-22°N, and (d) the dust-only AOD (~~"No Aging " minus "Aging "~~ No Aging minus Aging case). The results show a higher dust burden over the ~~dust~~ source regions in ~~Western~~ West Africa for the ~~"No Aging " case as~~ No Aging case compared with the reference ~~case ("Aging" case~~(Aging). For the ~~"No Aging "~~ No Aging case, the dust plume ~~is slightly extended~~ slightly extends further to the west over the Caribbean because of the ~~lower~~ reduced dust removal during transport. The difference between the two simulations decreases during ~~the~~ transport, which is supported by the differences in the dust-only AOD. In contrast, the difference in the total AOD shows lower ~~AOD~~ values over the dust source region compared with the ~~"Aging "~~ Aging case, which indicates a significant contribution of the dust chemical aging to the total AOD.

Interestingly, the negative feedback between the AOD and the radiation scheme results in higher dust emission over the region from 10°E to 0°and thus causes a higher dust burden. The average dust emission during July 2009 over the region from 18°N to 22°N (lower panel) shows that the dust emission for the "Aging" case is on average higher by about $3\,\mathrm{g\,m^{-2}}$, which results in a higher dust burden by $1\,\mathrm{g\,m^{-2}}$ while the remaining amount of the dust ($2\,\mathrm{g\,m^{-3}}$) is deposited. The higher AOD in the "Aging" case results in stronger scattering of short-wave solar radiation, lower surface radiation fluxes but higher surface wind speed (as shown in Fig. S7 in the supplement), and eventually stronger dust emission of $2\,\mathrm{g\,m^{-2}}$. The increased wind speed (more than $0.25\,\mathrm{m\,s^{-1}}$ on monthly average) could result either from the increase in the surface temperature because of the absorption of the dust particles and the resultant increase in the surface pressure (Menon, 2002; Mishra et al., 2014) or from a change in the horizontal temperature gradient that also increases the local wind speed (Rémy et al., 2015). On the other hand, the more efficient removal of the large dust particles in the ~~"Aging "~~ Aging case by both scavenging and sedimentation results in lower dust burden and thus the lower AOD. The balance between the two competing processes defines the impact of dust chemical aging on AOD. The difference in the dust-only optical depth is shown in the lower right panel of Fig. 13 and indicates that the "No Aging" case has higher dust optical depth as a result of the lower dust removal as compared with the "Aging" case. The difference is at a minimum within a region between 18°N to 22°N. However, the total AOD shows that the "No Aging" case leads to a lower AOD, which is significant over Western Africa and less pronounced over the Caribbean sites. Note that the AOD, as compared with AERONET stations, shown in Fig. 12 does not resolve this large difference because the AERONET stations are all located in the DTA region where the differences are obviously lower.

The substantial higher AOD for the "Aging" case (0.3 on ~~a monthly meanbasis~~monthly mean) primarily results from the dust chemical aging because of the associated water uptake. Figure 14 shows the monthly averaged burden for lumped gas-phase acids (sum of HCl+$\mathrm{HNO_3}$+$\mathrm{H_2SO_4}$) and the difference between both simulations. The figure also shows the corresponding lumped inorganic aerosol mass (sum of $\mathrm{SO_4^{2-}}$+ $\mathrm{HSO_4^-}$+ $\mathrm{NO_3^-}$+ $\mathrm{NH_4^+}$+ $\mathrm{Cl^-}$+ $\mathrm{Na^+}$+ $\mathrm{Ca^{2+}}$+ $\mathrm{K^+}$+ $\mathrm{Mg^{2+}}$) and the aerosol associated water mass. For the ~~"Aging "~~ Aging case, the burden of ~~the lumped~~ acids is very low over the dust source region because of the uptake by dust particles – an important effect which has been also recently studied with the EMAC model by

Karydis et al. (2016) for the nitric acid uptake (also included here). Consequently, the ~~burden of the lumped aerosols~~ aerosol burden is higher over the dust source region and over the ~~dust~~ outflow region, because of the additional neutralization of the calcium ions by anions and the associated absorption of water vapor by the resulting calcium salts. As a result, the aerosol-associated water increases by more than 255 $\mathrm{mg\,m^{-2}}$ for the ~~aged~~ Aged case. The effect of dust chemical aging is a result of ~~the~~ gas–aerosol partitioning that clearly affects the AOD. It is best observed in the differences (right column of Figure 14)~~.~~ ~~This shows~~ , which reveal that the impact of ~~the dust~~ dust chemical aging can be ~~very high,~~ significant, but mainly due to the associated uptake of aerosol water. We refer to this effect as the "direct effect of dust chemical aging." In addition, we refer to the higher removal of aged dust (by both sedimentation and scavenging), and the consequently shorter dust lifetime, as the "indirect effect of dust chemical aging" – both effects are introduced in this study.

To obtain ~~better~~ improved statistics for the effect of dust chemical aging, the same analysis (~~"Aging " versus "No Aging"~~Aging versus No Aging) was applied to the entire evaluation period (2000–2012) at lower ~~model resolution (i. e., T42, or ≈ 280).~~ T42 model resolution. Figure 15 shows the long-term meridional dust burden mean and the model precipitation for TRMM observations over the DTA and DIR zones (as discussed above). The ~~"No Aging "~~ No Aging case consistently shows higher dust ~~burden~~ burdens in the DIR zone as a result of more efficient scavenging for the ~~"Aging "~~ Aging case. Even for this long-term average, the dust burden is three times higher for the "No Aging" case than the "Aging" case over the Caribbean sites. However, the impact of scavenging ~~of the "Aging " case is higher~~ in the Aging case is stronger in the region between 10°W and 20°W, which corresponds with the ~~high~~ precipitation peak in the ~~Western~~ West Africa region.

5 Conclusions

Tansatlantic dust transport is a major large-scale atmospheric phenomenon. Although the EMAC model mostly reproduces the dust pattern during the transatlantic dust transport, the dust loadings and AOD can deviate in magnitude and seasonality from observations. To examine ~~different key~~ the controlling processes, the dust outflow region has been divided into two subregions: (1) the dust-ITCZ (DIZ) zone and (2) the adjacent dust transport over the Atlantic Ocean (DTA) zone. In the former, the dust is removed primarily by scavenging, whereas in the latter region sedimentation is predominant. Considering the two subregions allows the distinction of factors that affect the transatlantic dust transport.

Several sensitivity studies were conducted using the EMAC model ~~reference setup that was implemented by Abdelkader et al. (2015) , which uses an~~ following Abdelkader et al. (2015) – with a comprehensive setup which includes a fully coupled online dust emission scheme and ~~explicit dust chemical aging . The modeled~~ an explicit chemical aging of the atmospheric dust particles. First, the simulated AOD is sensitive to the emission flux parameterization~~but it is even more sensitive~~ , and even more to the choice of the convection scheme. The ~~modeled AOD is more sensitive to the~~ dust emission flux affects the AOD over West Africa ~~compared with~~ more strongly compared to the Caribbean sites. ~~EMAC can use several convection schemes, and~~ On the other hand, the dust burden shows a very steep gradient westward over the Atlantic Ocean. This is mainly a result of dust removal by deposition (sedimentation and scavenging) during long-range transport. Over the Atlantic (within DTA), this gradient is linear in the logarithmic scale, whereas the gradient is nonlinear over the Western and Eastern Atlantic (especially within DIZ). The dust burden over West Africa (east of 10°W) is about 1000 $\mu g\,m^{-3}$ but declines to 50 $\mu g\,m^{-3}$ over the Caribbean. The different convection parameterization schemes show more than a factor of 2 difference in dust burden over West Africa, and about a factor of 3 over the Caribbean. This is primarily a result of different precipitation rates and the associated differences in dust removal. Overall, the dust outflow into the Caribbean is best represented by the ~~dust outflow to the Caribbean best represented if the~~ ECMWF convection scheme~~is used – mainly because the~~, as a result of more realistic representation of precipitation within the ITCZ ~~is better reproduced compared to the other schemes (~~(compared to other schemes available in EMAC ~~). As a result of the~~ and relative to TRMM observations). The more realistic precipitation ~~, the~~ subsequently improves the dust removal (compared to the reference EMAC simulations) and subsequently the AOD on both sides of the Atlantic Ocean ~~is significantly improved,~~ significantly within the DIZ zone, a region which is largely controlled by wet removal processes~~within the DIZ zone, especially when dust aging is considered~~. Considering the dust chemical aging amplifies this effect.

~~Dust~~ To study the impact of dust chemical aging, we use the calcium cation as a proxy for the overall chemical reactive dust fraction and consider the uptake of major inorganic acids (i.e., H_2SO_4, HNO_3, HCl) and their anions, i.e., sulfate (SO_4^{2-}), bi-sulfate (HSO_4^-), nitrate (NO_3^-), and chloride (Cl^-)) on the surface of mineral particles. The subsequent neutralization reactions with the calcium cation forms various salt compounds that causes the uptake of water vapour from the atmosphere, which leads to the chemical aging of dust particles. Dust chemical aging changes the particle sizes because of the additional amount of condensed inorganic acids and the associated uptake of water vapor by the neutralization products (salts). Therefore, the aged dust particles are larger and scatter ~~more light~~ light more efficiently, whereas they are more ~~efficiently~~ rapidly removed by dry and wet removal processes. To ~~distinguish~~ analyze these effects, we ~~introduce the *"direct effect of dust aging"* and the *"indirect effect of dust aging"*. These effects clearly show the differences between the "Aging " and " No Aging " simulations, and the result of~~ performed Aging and No Aging simulations, for which we distinguish between the direct and indirect effect of dust chemical aging on AOD.

In our senitivity simulations, the dust chemical aging shows the ~~air-pollution–dust interactions that can regionally strongly influence the AOD. Dust aging has the~~ largest impact on the AOD over West Africa and on the dust burden in the ITCZ. The ~~higher~~ larger impact on the AOD results from the increase in the aerosol burden (more than 120 $mg\,m^{-2}$) due to the uptake of acids and associated water by the originally insoluble dust particles. This directly increases the AOD by 0.15 ~~on monthly average~~(monthly average). As a result of the radiative feedback on the atmospheric dynamics and circulation, the dust emission regionally increases. On the other hand, the aged dust particles are more efficiently removed in our EMAC reference setup compared with the ~~"~~non-aged ~~"~~ dust particles case. The enhanced removal of aged particles decreases the dust burden and lifetime, indirectly affecting the AOD. Both processes are significant and the net effect depends on the region and the level of dust chemical aging, which is controlled by the strength of the dust outflow and the collocated air-pollution levels. In order to improve the dust cycle in climate models, we recommend an explicit treatment of dust chemical aging, at least by considering the calcium cation as a proxy for the overall chemical reactivity of the mineral dust particles.

Appendix A: Evaluation metrics

- RMSE – Root Mean Square Error between the model (m) and the observations (o):

$$RMSE = \sqrt{\frac{1}{N}\sum (X_m - X_o)^2} \tag{A1}$$

- σ – Standard deviation of the model (σ_m) and the observation (σ_o) for variable (X_i) with average of ($\bar{X}$) with N the number of observations:

$$\sigma = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(X_i - \bar{X})^2}, \qquad where \quad \bar{X} = \frac{1}{N}\sum_{i=1}^{N} X_i \tag{A2}$$

- R – Correlation coefficient between the model (m) and the observations (o):

$$R = \frac{\sum_{i=1}^{N}(X_i^m - \bar{X^m})(X_i^o - \bar{X}^o)}{\sum_{i=1}^{N}(X_i^m - \bar{X^m})^2 \sum_{i=1}^{N}(X_i^o - \bar{X}^o)^2} \tag{A3}$$

- r geometric mean of the model (r_m) and the observations (r_o).

$$r = \sqrt[n]{\Pi_{i=1}^{N} X} \tag{A4}$$

- MBE – Mean Bias Error between the model and the observations:

$$MBE = \frac{1}{N}\sum (X_m - X_o) \tag{A5}$$

- GFE Growth Factorial Error

$$GFE = \frac{1}{N}\sum \frac{|(X_m - X_o)|}{X_m + X_o} \tag{A6}$$

- SS1 – Skill score between the model (m) and the observations (o) (Taylor, 2001):

$$SS1 = \frac{4(1+R)}{(\sigma_f + 1/\sigma_f)^2(1+R_0)}, \qquad where \quad \sigma_f = \frac{\sigma_o}{\sigma_m} \quad R_0 = 0.0 \tag{A7}$$

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Table 1. EMAC sub-models used in the current study and the corresponding references.

Submodel	Description	Reference
AEROPT	Aerosol optical properties	Lauer et al. (2007); Klingmüller et al. (2014); Pozzer et al. (2015)
CLOUD	ECHAM5 cloud scheme as MESSy submodel	Roeckner et al. (2006)
CONVECT	Convection parameterizations	Tost et al. (2010)
CVTRANS	Convective tracer transport	Tost et al. (2006b)
DDEP	Dry deposition	Kerkweg et al. (2006a)
EQSAM4clim	Fast aerosol thermodynamics	Metzger et al. (2016)
GMXe	Aerosol dynamics and microphysics	Pringle et al. (2010)
ISORROPIA-II	Aerosol thermodynamics	Fountoukis and Nenes (2007)
JVAL	On-line photolysis rates	Landgraf and Crutzen (1998)
LNOX	NO_x production from lightning	Tost et al. (2007)
MECCA	Gas phase chemistry	Sander et al. (2005)
OFFEMIS	Prescribed emissions of trace gases and aerosols	Kerkweg et al. (2006a)
ONEMIS	On-line calculated emissions	Kerkweg et al. (2006b); Astitha et al. (2012)
RAD	ECHAM5 radiation scheme as MESSy submodel	Roeckner et al. (2006); Joeckel et al. (2010)
SCAV	Comprehensive scavenging of aerosols and gases	Tost et al. (2006a)
SEDI	Sedimentation of aerosols	Kerkweg et al. (2006a)
TNUDGE	Newtonian relaxation of species	Kerkweg et al. (2006a)
TROPOP	Tropopause and other diagnostics	Joeckel et al. (2006)

Table 2. Long-term EMAC model evaluation for the period 2000-2012 for AOD. Statistics are given for both sides of the Atlantic, based on the selected AERONET sites around ~~Western~~ West Africa and the Caribbean (station average). The ~~sites are shown in Figure 3 and the~~ evaluation metrics are defined in ~~the~~ Appendix A, while the station locations are shown in Figure 3.

	Western Africa	Caribbean
Mean_m	0.16± 0.27	0.12± 0.18
Mean_o	0.24± 0.37	0.14± 0.22
r_m	~~0.16~~0.13± ~~0.27~~ 0.40	~~0.12~~0.11± ~~0.18~~ 0.27
r_o	~~0.24~~0.29± ~~0.37~~ 0.35	~~0.14~~0.13± ~~0.22~~ 0.29
RMSE	0.35	0.23
R	0.61	0.43
MBE	-0.19	-0.11
GFE	-0.24	-0.12
SS1	0.73	0.70
PF2	0.59	0.81
PF10	1.00	1.00
NPOINTS	50288	15827

Table 3. Description of the transatlantic dust transport sensitivity simulations for two key-processes: (i) Emission flux (Sec. 4.1) and (ii) convection scheme (Sec. 4.2). Highlighted cases are shown in the manuscript (for all cases see the Supplement, Fig. S3–S4). The emitted dust mass during July 2009 for the reference case is $0.6133\ \mathrm{kg\,m^{-2}}$.

	Case	Description
Emission	EMAC	Reference simulation
	B1E1	Redistribution of dust between accumulation and coarse modes
	B1E2	As EMAC, accumulation fraction incased by a factor of 2.61
	B1E3	As EMAC, the coarse mode increased by a factor of 5.3
	B1E4	As EMAC, the accumulation mode increased by a factor of 5.3
	B1E5	As EMAC, the accumulation mode increased by a factor of 10.6
	B1E6	As EMAC, the accumulation and coarse modes increased by a factor of 10.6 and 2.61 respectively
	B1E7	As EMAC, the accumulation and the coarse modes increased by a factor of 2.61
	B1E8	As EMAC, factor=2.61 in the horizontal flux
Convection	EMAC	Reference simulation; TIEDTKE convection with NORDENG closure
	B1T2	TIEDTKE convection with TIEDTKE closure (Tiedtke, 1989)
	B1T3	TIEDTKE convection with HYBRID closure (Tiedtke, 1989)
	B1T4	ECMWF operational convection scheme (Bechtold et al., 2004) with the shallow convection closure of Grant and Brown (1999)
	B1T5	ECMWF operational convection scheme (Bechtold et al., 2004)
	B1T6	Zhang-Hack-McFarlane convection scheme (Zhang and McFarlane, 1995; Hack, 1994)

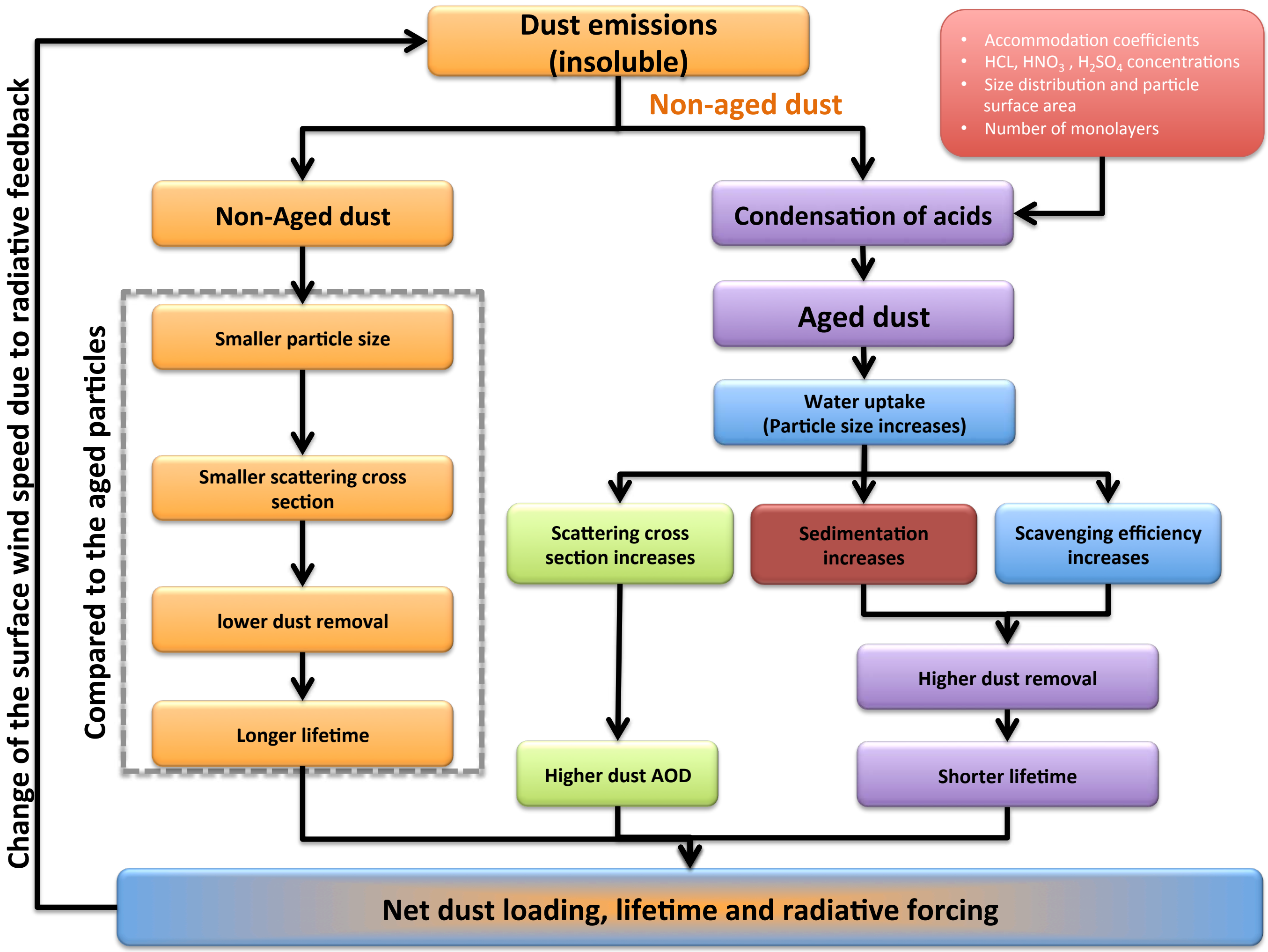

Figure 1. Schematic representation of the dust cycle and ~~air-pollution-dust-aging-radiation~~ air-pollution-dust-chemical-aging-radiation feedbacks in EMAC. Air pollution controls the chemical aging of dust particles, whereby the consequent water uptake increases the dust particle scattering cross section, enhances the dust deposition (wet and dry) which ~~decreases~~ tends to decrease the dust lifetime. The net radiative differences between aged and non-aged dust particles are indicated.

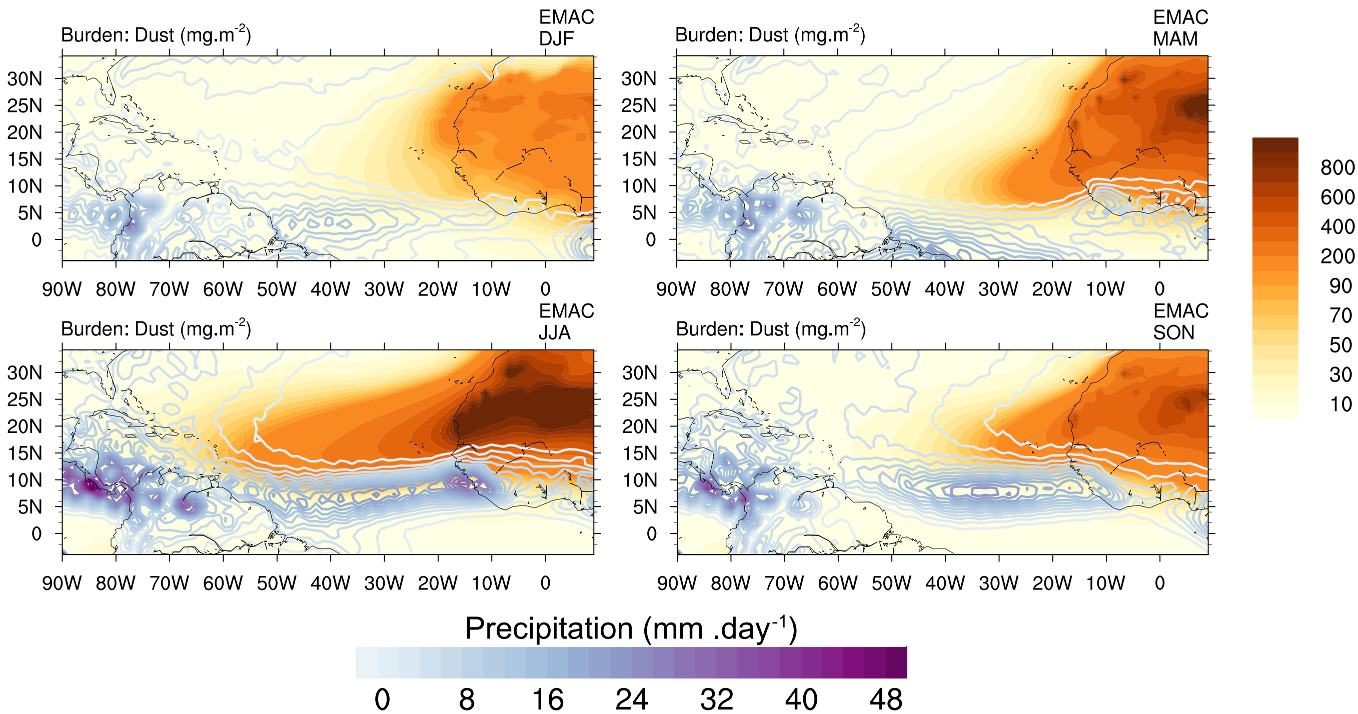

Figure 2. Seasonal averages of the dust burden and precipitation representing the transatlantic dust outflow for the entire model evaluation period (2000-2012). Dust burden and precipitation are at maximum during boreal summer and at minimum during winter. The orange color ~~represent~~ represents the dust burden while the purple color (contour lines) ~~show the~~ depicts precipitation.

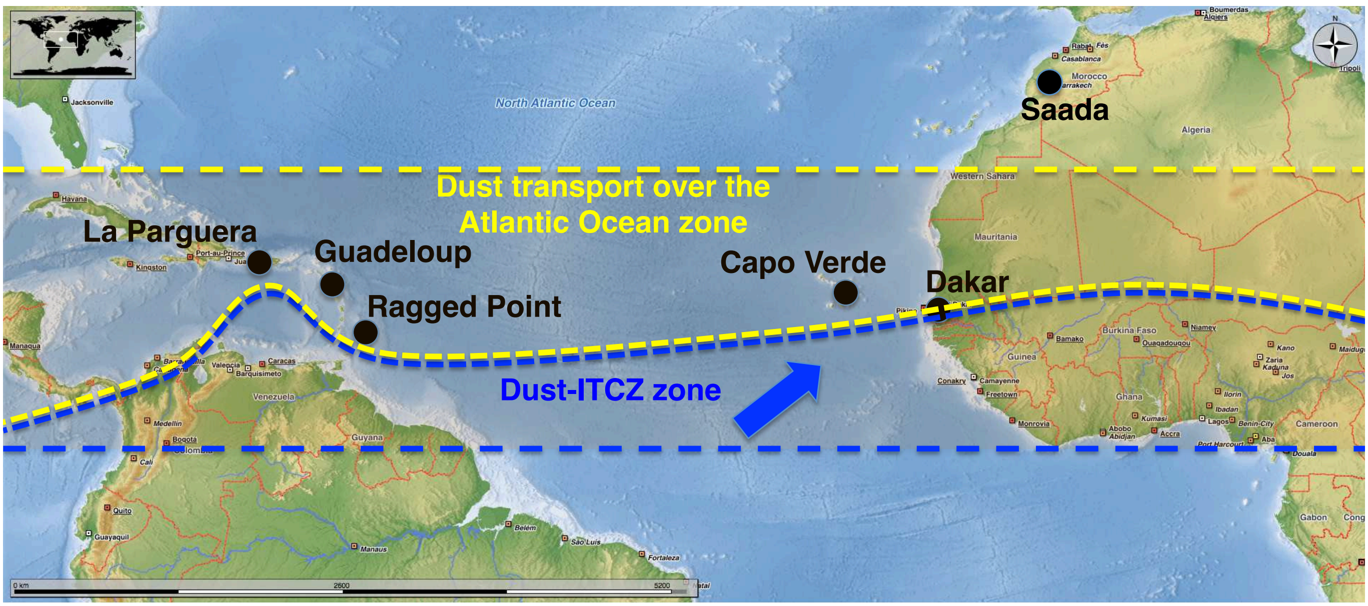

Figure 3. The location of selected AERONET stations used in the transatlantic dust transport study. Stations: Saada, Capo Verde and Dakar are as *"West Africa"* and stations: La Parguera, Guadeloup and Ragged Point as *"Caribbean"*. The upper blue line shows the approximate northern bound of the ITCZ, The yellow box ~~shows roughly~~ indicates the adjacent dust transport region (DTA) zone. The region ~~in~~ within the blue bounds represents the dust-ITCZ interaction zone (DIZ). These regions are defined according to the predominance of the dust removal mechanism shown in Fig. S1 in the supplement.

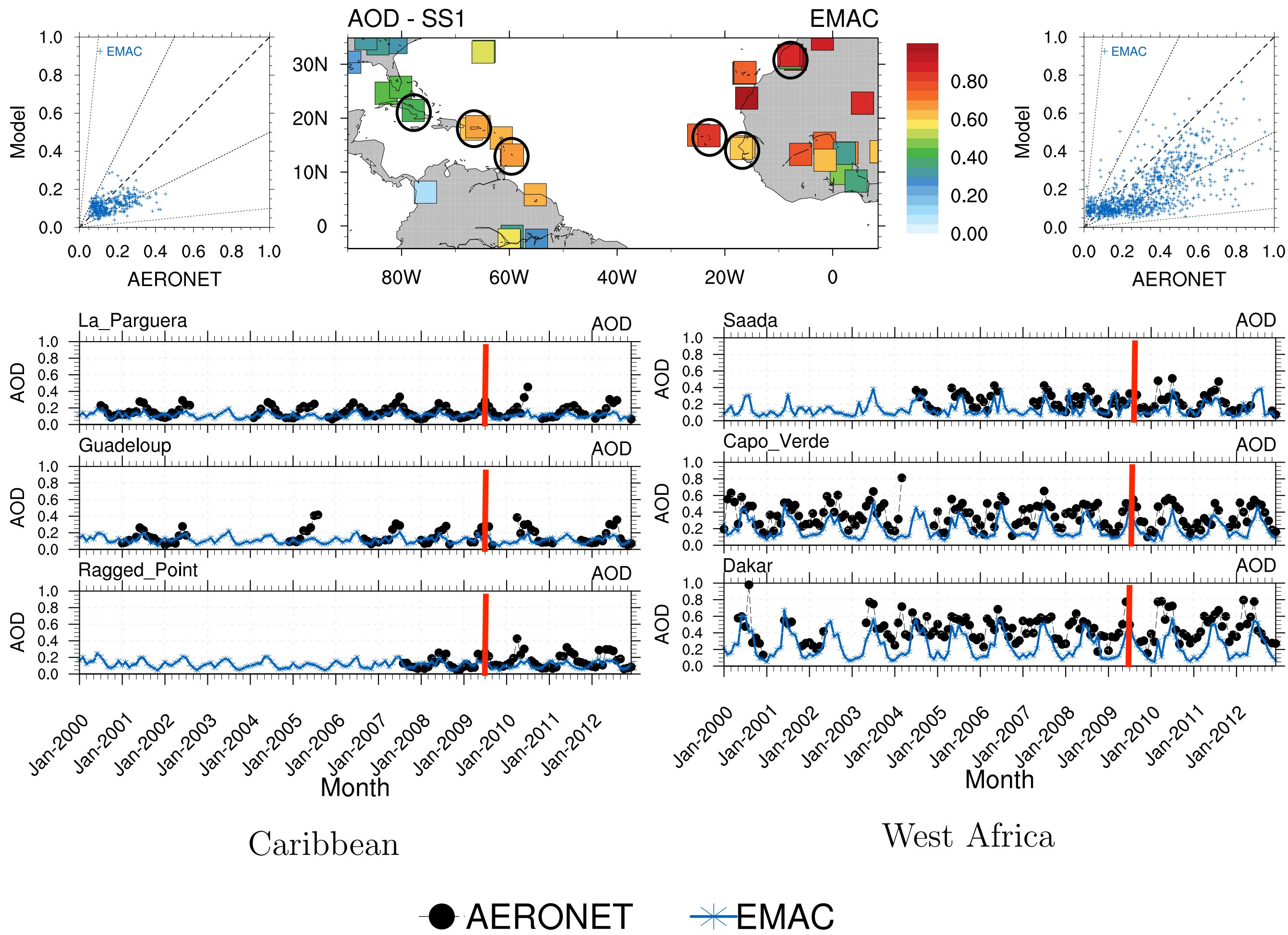

Figure 4. Long-term evaluation for AOD (2000-2012) over western Africa and the Caribbean: (Top panel) scatter ~~plot~~ plots (left for the Caribbean, right for the ~~Western~~ West Africa region) and skill score (SS1) defined in ~~the~~ Appendix (A): (Lower panel) time series for stations ~~in~~ at both regions (monthly means of ~~5hour~~ 5 hour averages for model and AERONET AOD). The red bars represent the July 2009 dust outflow period and the black circles ~~show~~ depict the selected AERONET stations shown in Fig. 3 ~~, both are used in~~ within observations for the period of our sensitivity simulations.

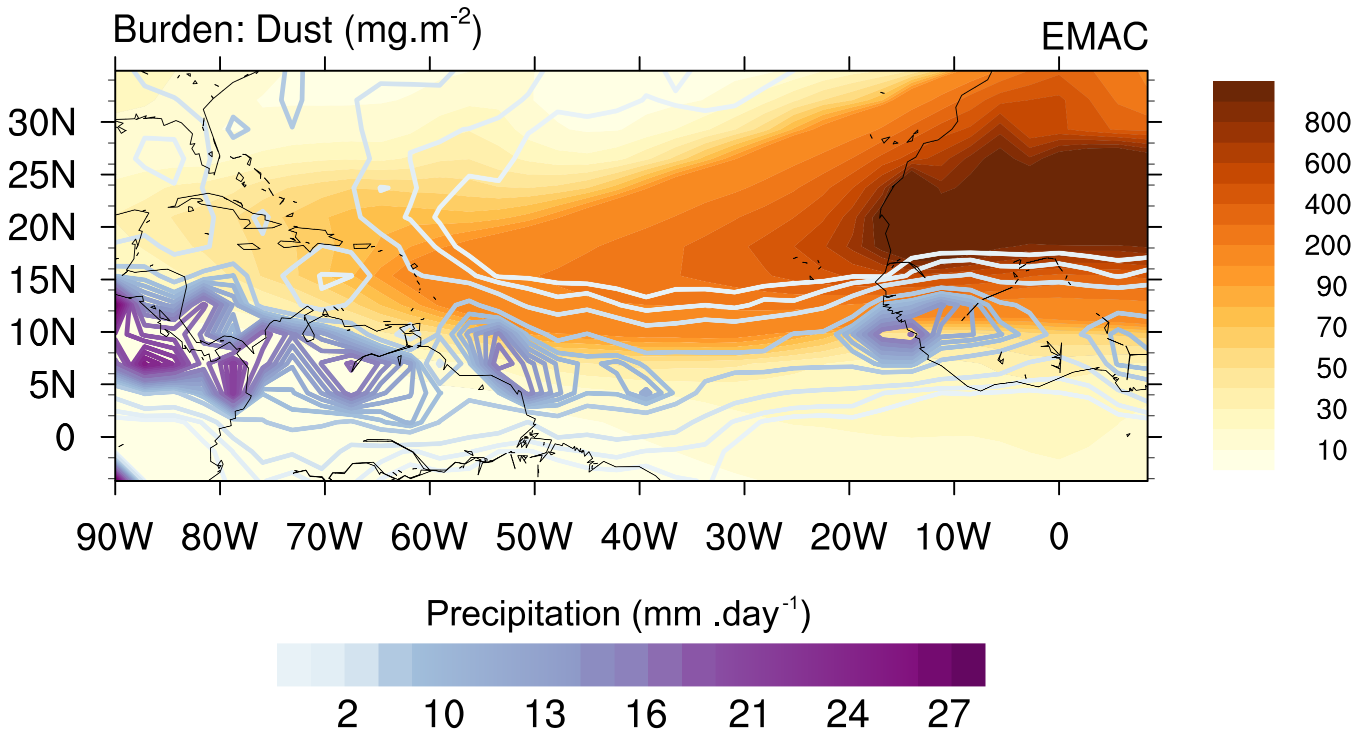

Figure 5. The EMAC computed spatial distribution of the dust burden (orange) and total precipitation (purple lines) for the reference simulation for July 2009 (monthly mean).~~The distribution indicates the dust outflow area over the Atlantic Ocean.~~

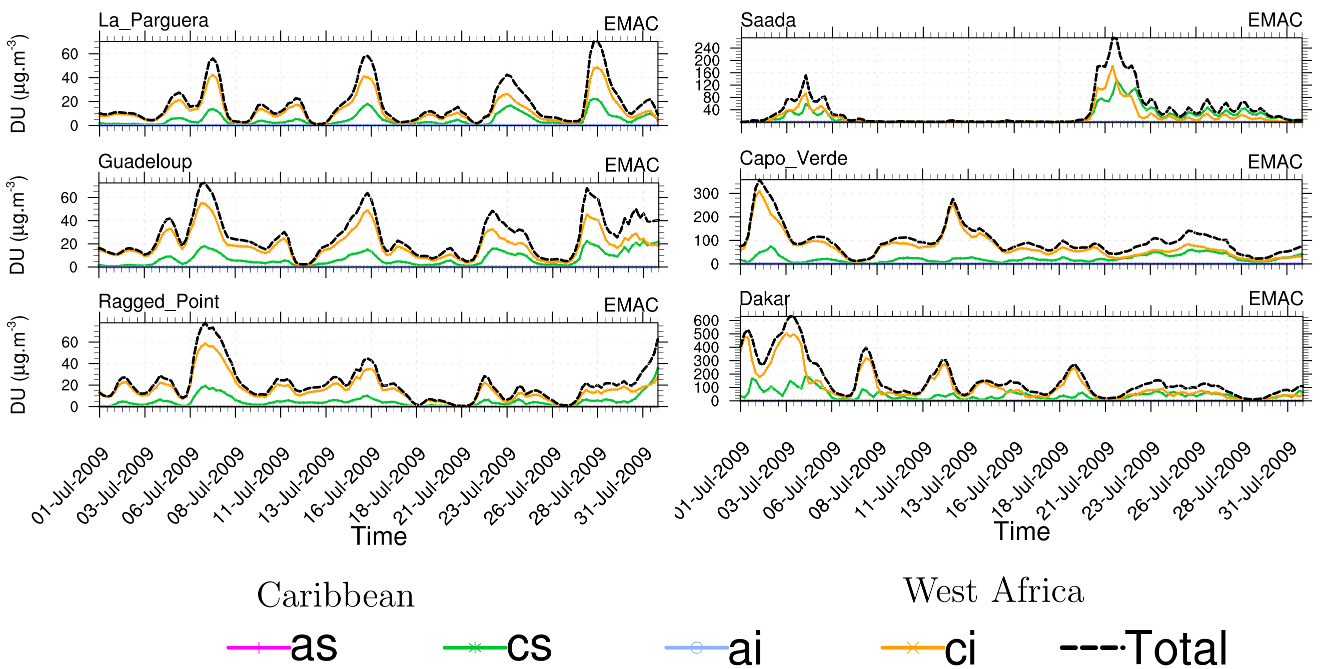

Figure 6. Time series of size-resolved surface dust concentrations for the different AERONET stations shown in Figure 3. Aerosol modes: accumulation soluble (as); coarse soluble (cs); accumulation insoluble (ai); coarse insoluble (ci). Note the different scaling which reflects the wide range of concentrations at these stations. The accumulation mode dust fraction has a much lower contribution to the total dust concentration.

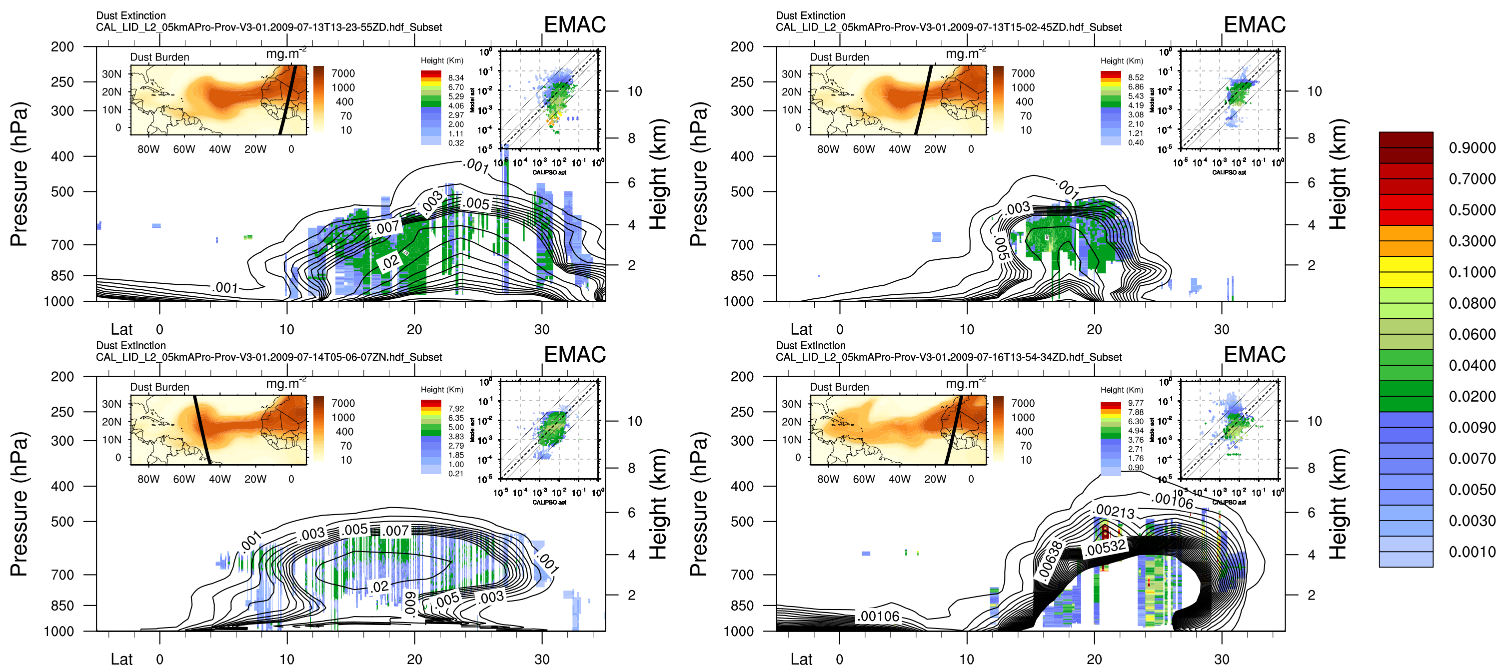

Figure 7. Collocated EMAC and CALIPSO observations of dust extinction and burden for four different CALIPSO overpasses during the second dust outbreak over the Atlantic Ocean. The time of the overpass is shown in the upper left corners (13-16th July 2009). The solid lines show the ~~modeled~~ simulated extinction and the colored contours shows the observed CALIPSO extinction, which are complemented by the scatter plots for point-to-point comparison colored by the corresponding elevations of each observation (distinguished by the colors). The lines in the scatter plots ~~show~~ delineate the one-by-one~~line~~, the factor ~~, of~~ two and the factor of ten intervals. All available comparisons with CALIPSO overpasses for this period are shown in the Supplement (Fig. S2a – Fig. S2e).

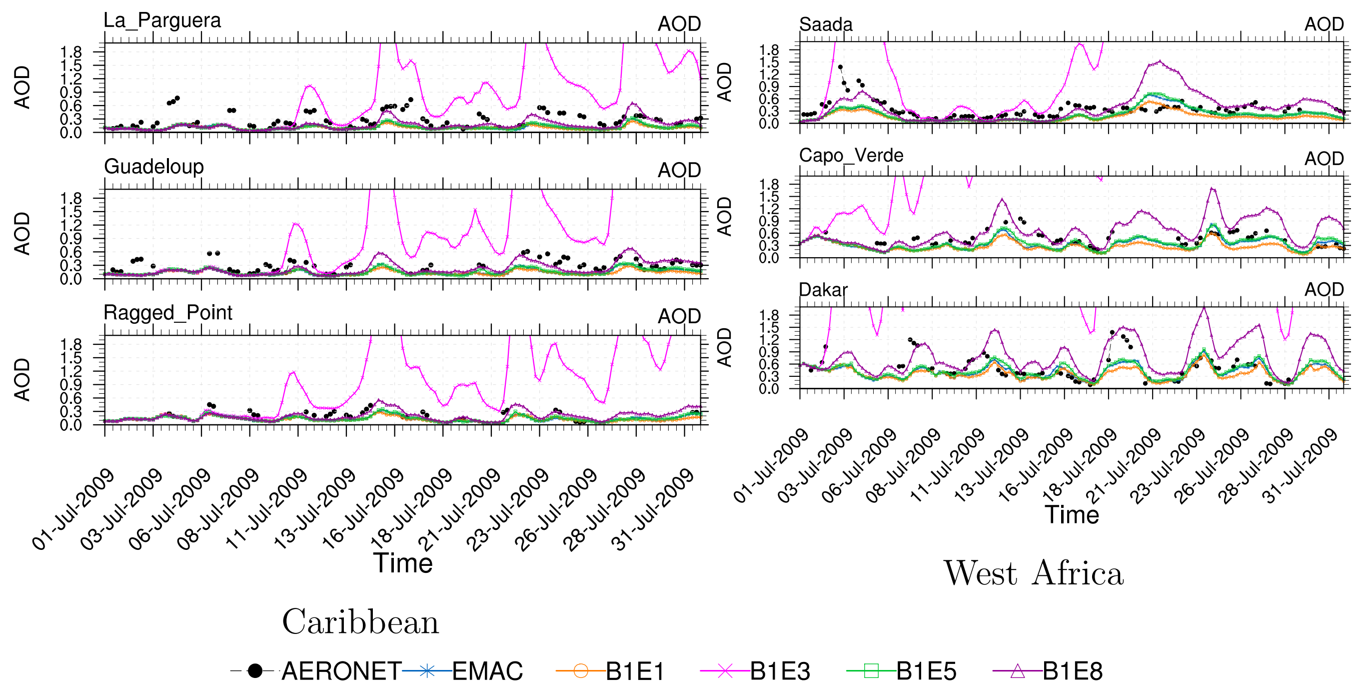

Figure 8. EMAC and AERONET AOD for the western Africa (right) and Caribbean (~~right~~left) sites based on different dust emissions (Table 3).

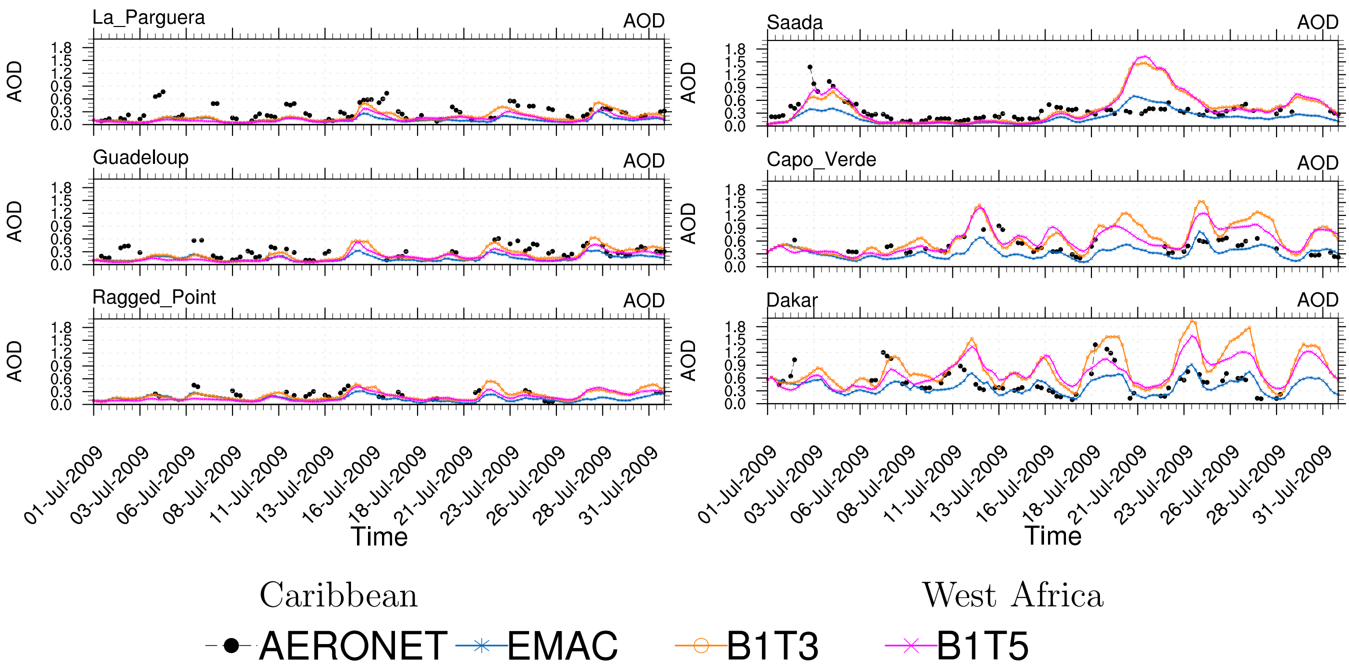

Figure 9. EMAC and AERONET AOD for ~~the western~~ West Africa (right) and Caribbean (~~right~~left) based on different convection schemes (Table 3).

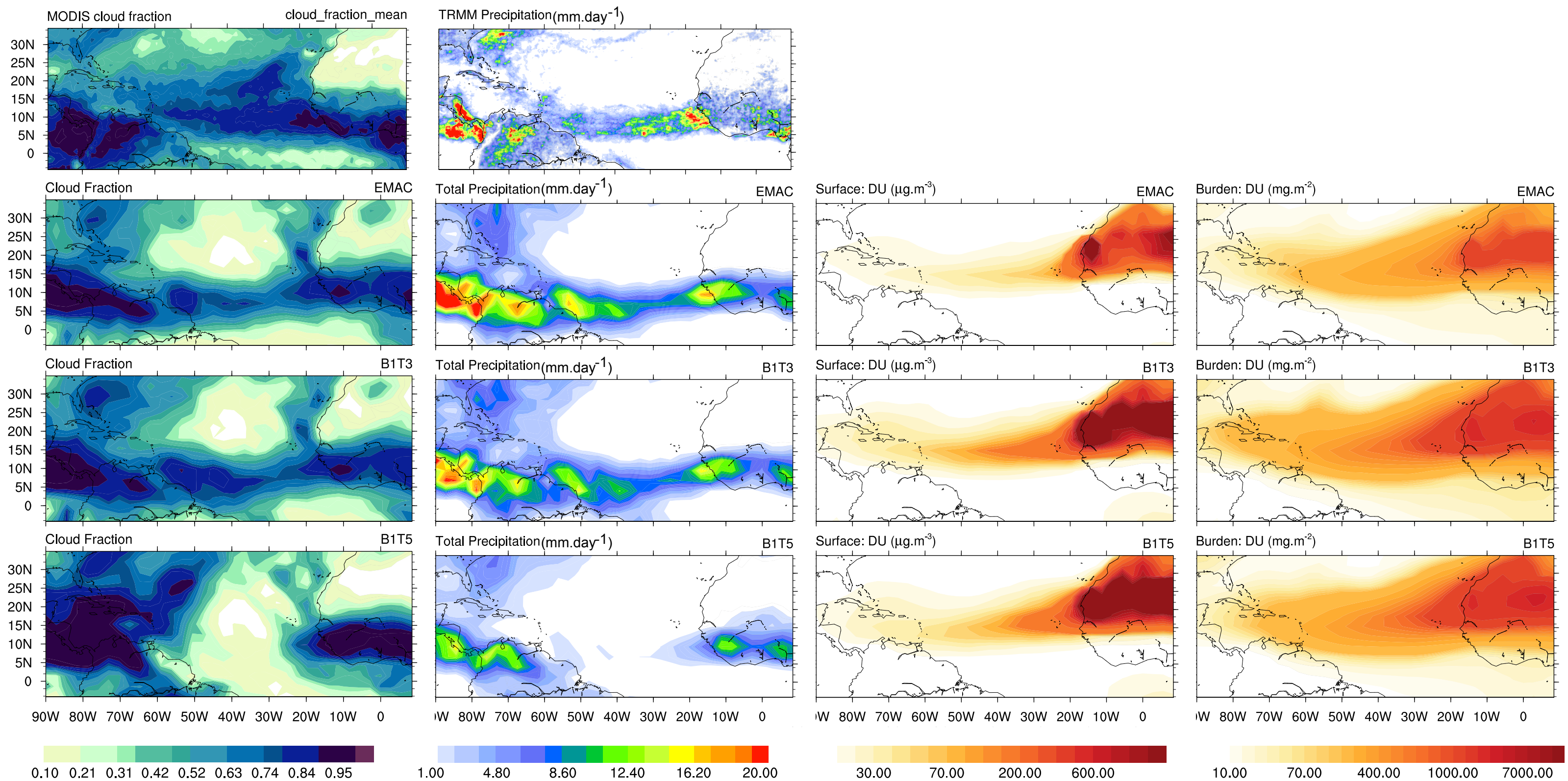

Figure 10. (Top) MODIS cloud fraction and TRMM precipitation (July 2009 monthly mean); (below) EMAC results (from left to right) cloud fraction, precipitation, surface dust concentration, and dust burden for different convection schemes (2nd–4th row) highlighted in Table 3. The model precipitation and cloud cover agrees for our EMAC set-up best with TRMM and MODIS observations with the ~~ECMWF and~~ TIEDTKE (B1T3) and ECMWF (B1T5) convection schemes.

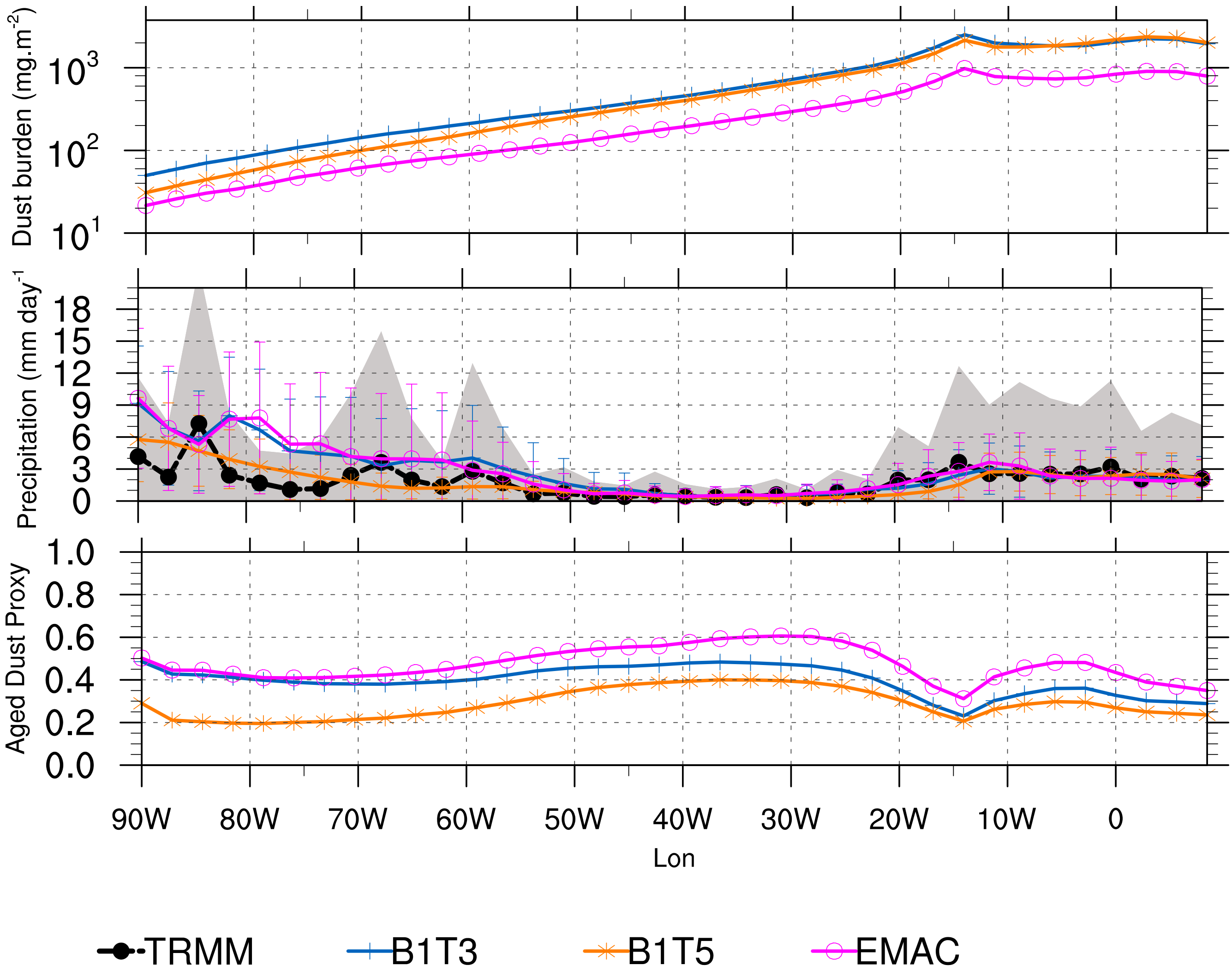

Figure 11. Comparison of observed and calculated meridional means over the dust outflow over the Atlantic Ocean region (10°–25°N) for: (~~Top~~top) dust burden, (middle) precipitation, (bottom) aged dust proxy (ADP) for July 2009 (monthly mean). The ADP represents the ratio between aged and non-aged dust particles. The shaded area represents one standard deviation of the TRMM-precipitation and the bars shows one standard deviation of the model results.

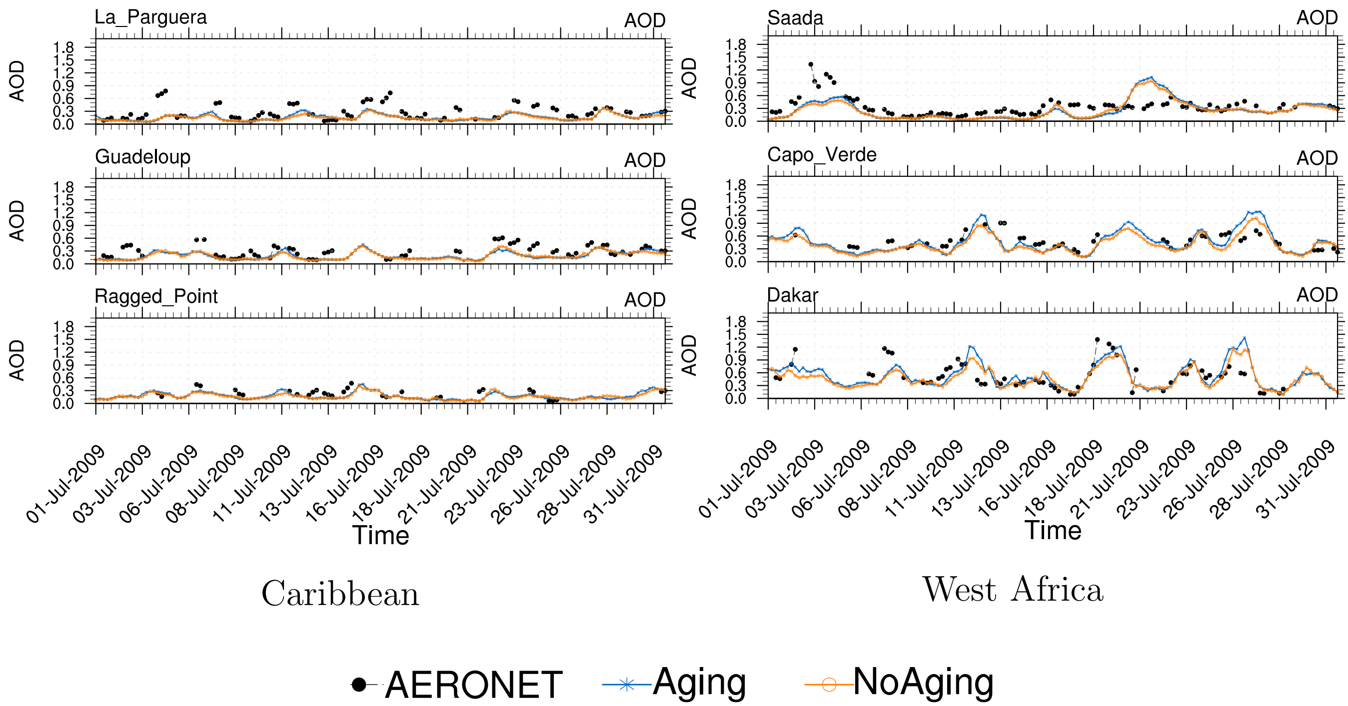

Figure 12. Comparison of observed (AERONET) and calculated AOD for western African and the Caribbean and for two EMAC simulations that include and exclude chemical aging (labeled "Aging" and "No aging", respectively).

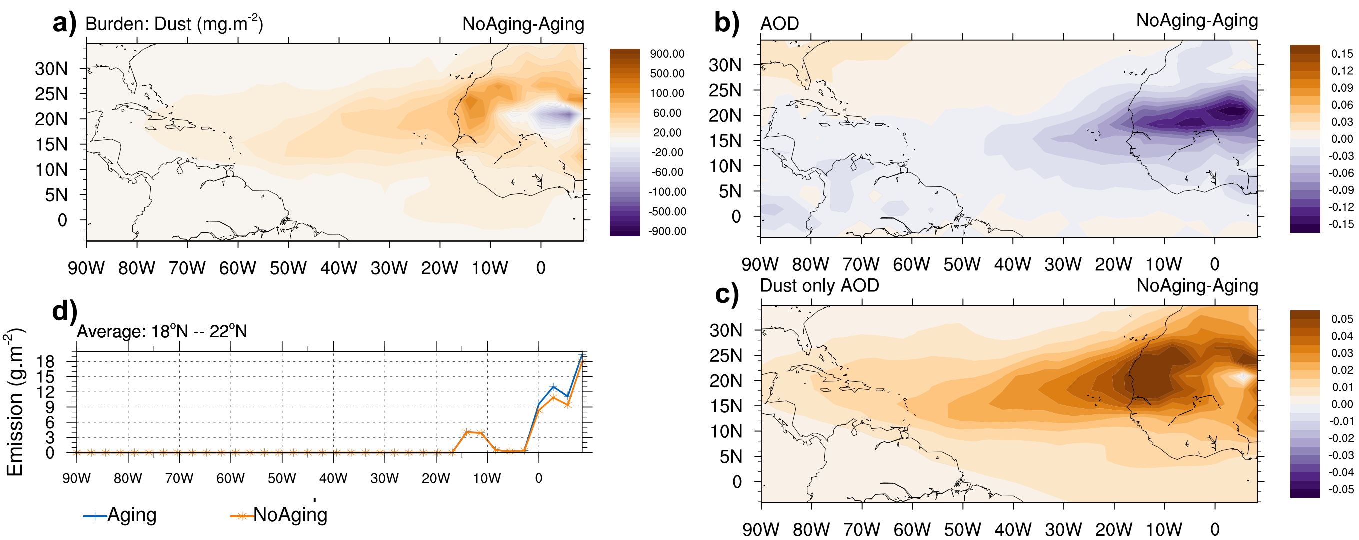

Figure 13. EMAC results (monthly mean for July 2009) for two simulations that include and exclude chemical aging (labeled "Aging" and "No aging", respectively). (a) difference in dust burden, (b) difference in AOD, (c) dust emission averaged over the region from 18°-22°N for both simulations, (d) difference in "dust only AOD". "Aging" is the reference case. The difference shows the results of the "No Aging" minus "Aging" case.

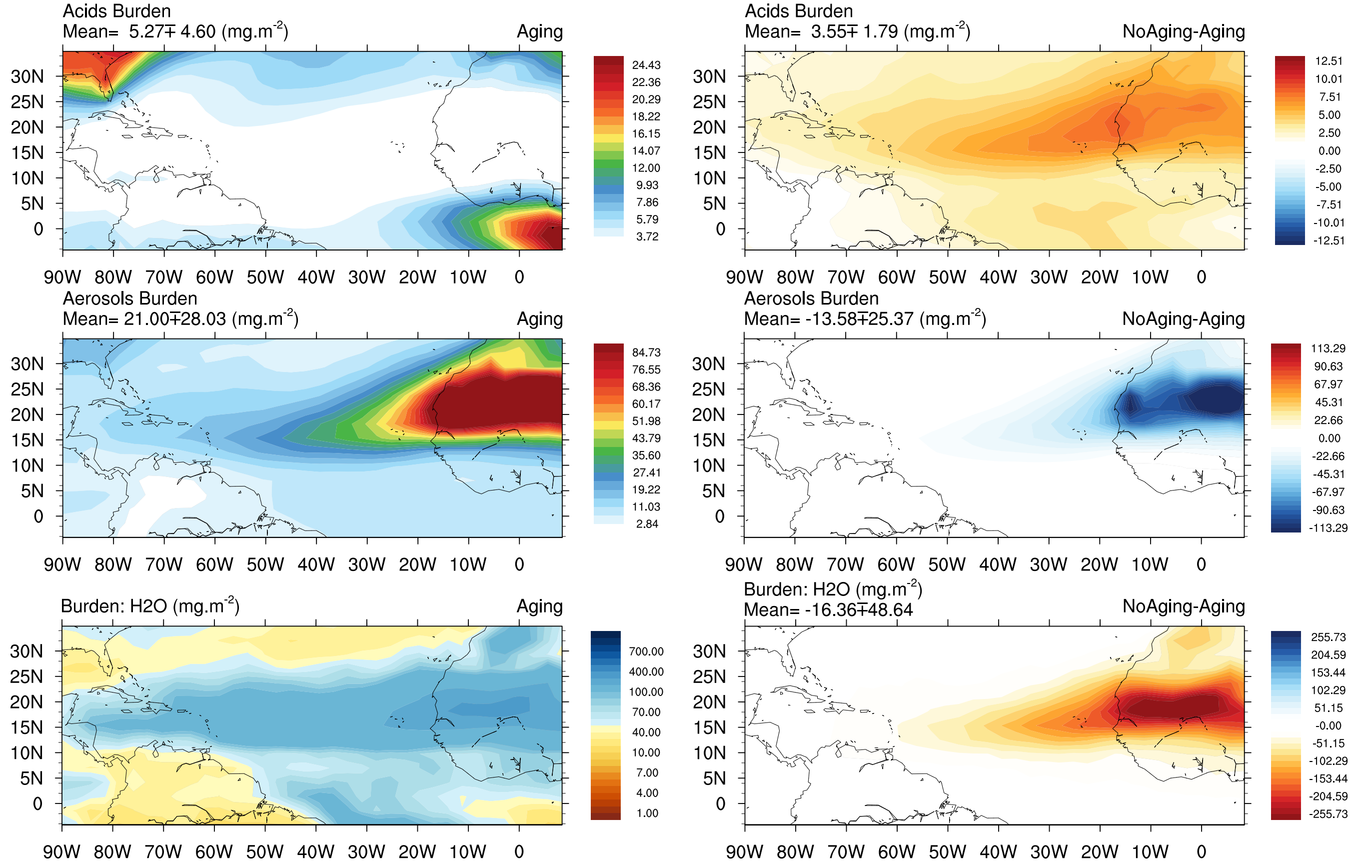

Figure 14. Monthly mean (~~Top~~July 2009) for: (top) burden of lumped inorganic gas-phase acids (sum of HCl+HNO_3+H_2SO_4), (middle) burden of lumped aerosols (sum of SO_4^{2-}+ HSO_4^-+ NO_3^-+ NH_4^++ Cl^-+ Na^++ Ca^{2+}+ K^++ Mg^{2+}), (bottom) burden of aerosol associated water mass (monthly mean). (Left column) reference simulation (Aging case), (right column) difference between reference and ~~the "~~No Aging ~~"~~case. Note the inverted color scales for the bottom two panels, where higher aerosol water mass is shown in blue and lower in red.

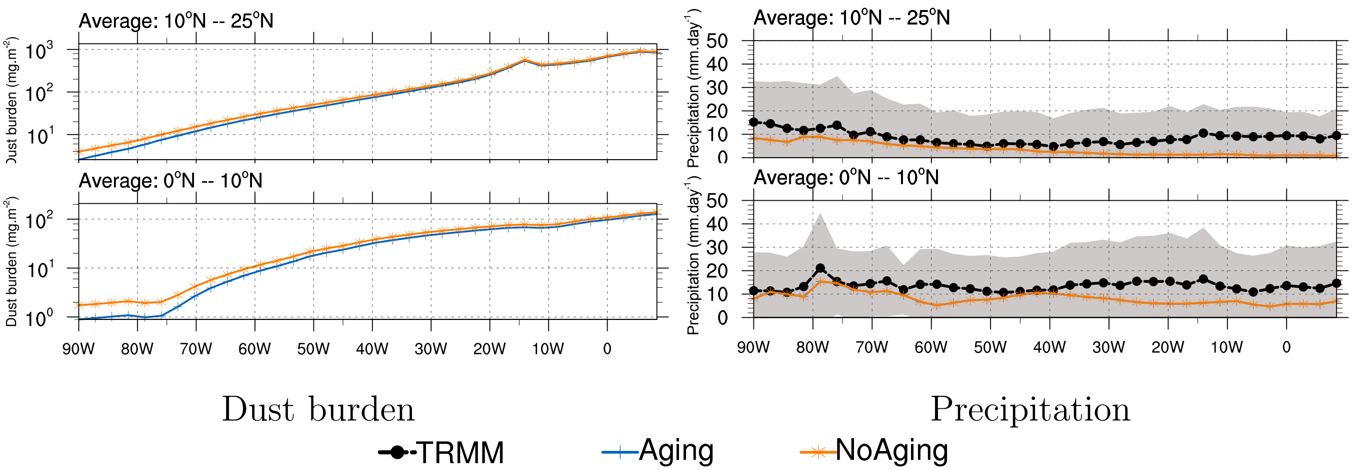

Figure 15. (Left) Dust burden, (right) precipitation for different regions: (Top) dust transport over the Atlantic Ocean zone, (bottom) dust-ITCZ zone 0°to 10°N. The shaded area represents one standard deviation of TRMM precipitation. The results show the long-term average of the entire evaluation period 2000-2012.