1 Response to D. Lunt, Executive editor

Dear Executive editor,

Thank you for your review of our article. Your remarks and suggestions appear below in italics, together with our responses to these remarks and suggestions. Our proposed amendments to the text of our paper appear in bold.

1. *The paper must be accompanied by the code, or means of accessing the code, for the purpose of peer-review. If the code is normally distributed in a way which could compromise the anonymity of the referees, then the code must be made available to the editor. The refereeeditor is not required to review the code in any way, but they may do so if they so wish.*

   All papers must include a section at the end of the paper entitled "Code availability". In this section, instructions for obtaining the code (e.g. from a supplement, or from a website) should be included; alternatively, contact information should be given where the code can be obtained on request, or the reasons why the code is not available should be clearly stated

   We have added the following section at the end of the paper:

   Code availability:

   A number of model codes developed at CNRM, or in collaboration with CNRM scientists, are available as Open Source code (see https://opensource.cnrm-game-meteo.fr/). However, this is not the case for the aerosol code presented in this paper. This code is nevertheless available upon request from the authors of the paper.

2. *All papers must include a model name and version number (or other unique identifier) in the title. In your instance, 'the code' would refer to just the aerosol component, rather than the whole CNRM model.*

   We have modified the title of the article as follows: **Development and basic evaluation of a prognostic aerosol scheme (v1) in the CNRM Climate Model CNRM-CM6**. We have amended the text of the abstract and of the article accordingly, referring to these model versions.

2 Response to Anonymous Referee #1

Dear Referee,

We are very grateful for your very careful and very detailed review. All your questions and suggestions, which appear below in italics, will lead to a clearer and stronger paper. Please also find below our responses to these remarks and suggestions. Our proposed amendments to the text of our paper appear in bold. Page and line numbers refer to the version of the paper you reviewed.

1. *The new description of mineral dust is presented as an important improvement. However, compared to the ranges derived from the AeroCom models analyzed by Huneeus et al. (2011), the new dust emission module produces very high emission totals (both globally and in most regions shown in Table 3), especially in the free-running simulation. To substantiate their claims, the authors should include an evaluation of the dust size distribution. This can be done by comparing Angstrom coefficients obtained with the new and original scheme against measurements from selected AERONET stations, dominated by dust.*

   Thank you for your suggestion to combine Angstrom coefficient and AOD information to infer evaluation of dust emission loads. Such a methodology is detailed and used in *Huneeus et al.* (2011), for instance. In given cases, such as a simultaneous underestimation of the AOD and underestimation of the Angstrom coefficient points to an overestimation of the mass emissions, the methodology can be conclusive and can point out an under or overestimation of the emissions. Unfortunately, our current model development does not allow for retrieval of Angstrom coefficients for the individual bins of our aerosol model. Indeed, only the AOD at 550 nm is computed for all individual bins, the three dust bins for instance, then the model adds up bin AODs to derive an aerosol type AOD at 550 nm. Dust is one of these types. AODs needed at other wavelengths for the radiation code are computed for these aerosol types. Angstrom coefficients are thus given for aerosol types only.

   It is true that our emission totals with this new dust scheme appear to be at the high end of the values reported in *Huneeus et al.* (2011) for 15 AEROCOM models, with particularly high emissions

over the Middle East and the Australian regions. As indicated in the article, we have adopted for our simulations here a value of 5.10^{-7} for the c_α coefficient proportional to the vertical to horizontal flux ratio, involved as such in the calculation of the dust emission flux. We computed this scaling factor, which differs largely among models (*Todd et al.*, 2008), to bring our 2004 AODs in the Sahelian region, the major global source of dust, into reasonable agreement with the satellite and AERONET observations. We could have adopted various such scaling factors depending on the region, as done by other modelling groups such as *Tosca et al.* (2013). We will keep that in mind as further developments of our model are put in place.

Moreover, one of the reasons for replacing the original scheme is that it did not performed well. For this the authors refer to "preliminary results using the original GEMS/MACC dust scheme". This should be explained in more detail. If possible, the authors should include one or more references to scientific papers in which the quality of the original scheme is analyzed.

The original scheme did not perform so well, neither within ARPEGE-Climat, nor within the ECMWF IFS (Integrated Forecast System). In the case of ARPEGE-Climat, dust simulations with this scheme are shown in the current paper (i.e. FreSim and NudSim simulations), while results obtained with a very similar scheme in IFS are analysed in *Melas et al.* (2013) and in *Huneeus et al.* (2011). Both articles underline that modelled dust AOD underestimate observations, even in the MACC Reanalysis where MODIS total AOD is assimilated in the course of the simulation. *Huneeus et al.* (2011) further point out that ECMWF IFS has the lowest emission load among 14 models, and then infer from a combined Angstrom coefficient and AOD analysis that ECMWF IFS dust emissions over Africa are underestimated.

We have now the following sentences in our paper page 6271 l15:

Dust aerosols simulated with ARPEGE-Climat and the dust scheme described in Section 2.2 confirmed the underestimation of dust aerosols already outlined by *Melas et al.* (2013) and *Huneeus et al.* (2011) when using a similar dust scheme within the IFS ECMWF model. Therefore, as a more complex scheme could be put into place in view of the detailed soil characteristics parameters available in ARPEGE-Climat from the ECOCLIMAP database (*Masson et al.*, 2003), an additional dust emission parameterisation has been included in the aerosol scheme, allowing for comparisons between the two parameterisations.

Similarly, it should be explained in more detail why the bins describing the size distribution of mineral dust have been shifted compared to the original description. Are there any references the authors can refer to?

The sizes of each bin have indeed been shifted from the original GEMS/MACC description to the description based on *Kok* (2011) and used in the regional climate model RegCM (*Zakey et al.*, 2006; *Nabat et al.*, 2012). With RegCM and ARPEGE-Climat using the same *Marticorena and Bergametti* (1995) dust emission scheme, we thought that the same choice of size distribution in both models was adequate. To confort us in that choice, very recently *Nabat et al.* (2014c) validated it within the coupled regional climate system model CNRM-RCM5, which is a regional version of CNRM-CM, in particular against aircraft observations.

We have added the following sentence after line 10 p 6271:

This size distribution adjustment was based on work done with the regional climate model RegCM (*Zakey et al.*, 2006; *Nabat et al.*, 2012); it has been recently validated in a regional version of CNRM-CM by *Nabat et al.* (2014c).

2. *A more detailed analysis of the global budget of the different aerosol components should be included. Currently, the authors compare global mean mixing ratios for the 12 aerosol tracers in the simulations with results from the MACC reanalysis. They should also compare the simulated global loads/burdens and lifetimes or deposition rates for both wet and dry deposition (the latter including sedimentation) for the different aerosol components with ranges estimated by other models (e.g. Textor et al., 2006; Tsigaridis et al., 2014). This will also be helpful to evaluate the impacts of the change of the parameter settings shown in Table 4.*

   As suggested, we have added to our paper an additional table (see Table 4) that shows diagnostics related to various global budgets. We have added at the end of section 4.1.2 the following text in our revised paper to go along with this table:

   Further insight into the behaviour of both types of simulations is provided in Table 4, which shows global annual means of the burden, residence time and ratios of various

sinks of the five aerosol types for the FreSimd2, NudSimd2, and MACC Reanalysis, while an estimation of the modelling range of these quantities is provided by *Textor et al.* (2006); *Huneeus et al.* (2011). Burden and residence times are higher for the NudSimd2 than for the FreSimd2 simulation for all aerosol types except SS, which is coherent with the results of Figure 1 analysed above in the same section. Values for both simulations are within the *Textor et al.* (2006); *Huneeus et al.* (2011) mean$\pm 2\sigma$ range, except in FreSimd2 for SO_4 with too low burden and residence time, and in both simulations for SS with too large burdens. However, *Grythe et al.* (2014) report a spread of more than 70 Pg yr^{-1} in the "best" SS source functions studied, which would generate much higher burdens than those of *Textor et al.* (2006). While the dry dep./wet dep. ratios are similar to lower for the FreSimd2 simulation than for the NudSimd2 simulation, the conv dep./wet dep. ratios are about 2 to 3 times smaller for FreSimd2, and the wet dep./total sink ratios a little larger for FreSimd2. Finally, the sed dep./dry dep. ratio, not null only for the coarser SS and DD bins, are the same for both simulations as dry deposition and sedimentation of large particles are independent from meteorology. In the end, more NudSimd2 results than FreSimd2 results shown in this table are closer to the AEROCOM means. Figures computed from the MACC Reanalysis diagnostics are also presented in Table 4 but should be taken as indicative only, as an error has been identified in the wet deposition amounts (up to 50% maximum), leading to an overestimation of the wet deposition diagnostics that results, for instance, in smaller MACC Reanalysis residence times. Apart from that error, Reanalysis burden amounts appear too high for SS and SO_4.

3. *In Figs. 6-9, the authors show maps of simulations results next to maps of climatological datasets from MODIS Aqua, MODIS Deep Blue, MISR, and Kinne et al. (2013). Rather than showing the AOD maps for these evaluation datasets, Figs. 6 and 7 should show the AOD map for the simulation in the first panel, and in the other four panels show the bias between the simulation and each of the evaluation datasets. Figs. 8 and 9 could then show the corresponding relative biases, rather than the relative differences between the evaluation datasets. The sign of the biases in these maps should be opposite to that currently shown in the top left panel of Figs. 8 and 9. Instead of showing both absolute and relative biases, the authors can also decide to keep only the absolute biases.*

   Thank you for your suggestions, which follow a slightly different logic than the one we first adopted. We now present in our article in Fig. 6 and 7 absolute values for the model and all reference datasets to illustrate characteristics of these datasets, and in Fig. 8 and 9 relative differences between the simulation and the evaluation datasets, as you suggested. We have amended the text of the article page 6284, lines 1 to 15 as follows:

   In the case of MISR, which has the largest spatial coverage of the satellite data we used, the model underestimation is lower in JJA than in DJF, with a relative mean bias of -41% and -52%, respectively (see Figures 8 and 9). This low bias is mainly driven by the oceanic values. In contrast, the model overestimates the observations in DJF in areas such as Central Africa, parts of Saudi Arabia and Northern Africa, and in JJA over the Arabian Sea and large parts of South America. Areas of model overestimation seem to follow the trace of biomass burning in tropical regions, while dust appears overestimated over the Arabian Sea. Over continents in JJA, at mid to northern latitudes, the bias appears quite patchy, with both positive and negative values.

   MISR and MODIS differ by more than 20% over large parts of the oceans, and they contrast even more over continents (not shown). The same comment applies to MODIS Deep Blue over continents, and is even more true for the *Kinne et al.* (2013) climatology. As a consequence, relative biases between model outputs and the other two satellite data sets, i.e. the MODIS Aqua and the Deep Blue products, yielded different results, see Figures 8 and 9. This is particularly the case over South America and Australia with large areas of observed low AODs (lower than 0.1). Over mid to high latitude oceans, the bias between *Kinne et al.* (2013) and our simulation is lower (around 10 to 50%) than the bias between MISR and our simulation (around 30 to 70%).

4. *In Fig. 10, the authors show maps for both the coarse and the fine aerosol fraction. Since these*

are complementary (they add up to 1), one of these rows should be removed. I suggest to keep the results for the fine mode, as in Kinne et al. (2013).

As you suggested, we kept the results for the fine mode only, changing also the difference between model and observations, for the sake of clarity.

5. *The comparison of both "anthropogenic sulphate" and "natural aerosols" with the climatology from Kinne et al. (2013) doesn't make sense and should be removed. Without a reference simulation using pre-industrial emissions or the use of tagged tracers, it is impossible to diagnose the anthropogenic sulphate and natural aerosols from the presented simulations, so including such a comparison and concluding there are large discrepancies is only misleading. Thus, the last two rows of Fig. 10 and the corresponding discussions (Page 6284, line 23-25; Page 6285 line 5-15; Page 6292, line 14-17) should be removed.*

   Thank you for these remarks and suggestions to which we agree to a large extent. We fully agree that a definite identification of the anthropogenic and natural aerosols would best be done with at least a reference simulation using pre-industrial emissions. This is indeed the methodology adopted when the anthropogenic aerosol radiative forcing is analysed, as for instance in *Schulz et al.* (2006); *Myhre et al.* (2013). Methodologies to isolate a single component, here for instance the anthropogenic sulfate, differ between models. However, some papers present results without this reference simulation, as in *Bellouin et al.* (2012), which analyses the aerosol radiative forcing from the MACC Reanalysis outputs only, with a specific method to identify for instance anthropogenic aerosols.

   In our case, we have no objective of radiative forcing whatsoever, but we are interested in comparing what comes out of our model and the *Kinne et al.* (2013) data, even though it can be argued that this comparison is crude for the anthropogenic sulfate. However, it is not that 'crude' as *Boucher et al.* (2013) write "In the present-day atmosphere, the majority of BC, sulphate, nitrate and ammonium come from anthropogenic sources, whereas sea salt, most mineral dust and terrestrial primary biological aerosol particles are predominantly of natural origin. Primary and secondary organic aerosols (POA and SOA) are influenced by both natural and anthropogenic sources." Clearly, the question of recombining aerosols in categories other than those of the model itself is a challenge, and as another example, *Sessions et al.* (2015), which make use of the MACC aerosol scheme, here used as a quasi-operational aerosol model, consider the MACC sulfate, similar to ours, in the pollution sulfate category.

   We therefore now present in the revised paper, the fine, anthropogenic sulfate and natural aerosol fractions. We have added some text as follows to better reflect the uncertainties described above:

   and we grouped our aerosol scheme "bins" to comply to the extent possible to these fractions. Total AOD has been separated ....(in our case DD and SS aerosols). This grouping may not appear fully satisfactory, the anthropogenic sulfate aerosols would for instance have been best identified running a supplementary simulation with pre-industrial conditions (*Schulz et al.*, 2006; *Myhre et al.*, 2013), or applying more complex grouping methodologies such as in *Bellouin et al.* (2012); *Sessions et al.* (2015), but the comparison detailed below is intended as a first estimation of our model outputs.

6. *I consider it a weak point that "re-evaporation is not applied in the free-running simulations". Instead of just switching it off, it would have been better to try to solve the underlying problem.*

   We fully agree to that, and we investigated the problem until we had no more options than to not apply the re-evaporation process. In short, the problem was identified quite late in the course of the validation of the aerosol module as it took place quite rarely: a couple of times at the most during a ten year simulation. Then it did not provoke any breakdown of the model, but caused the concentration of certain aerosols to increase gradually to abnormally high values until it would decrease again and return to normal stable values. The increase would take place in the course of a few hours, at a model grid-point, and then propagate with transport. We identified that it was related to the shape of the vertical large-scale precipitation profile and nothing else. Our scientists of the physics of the model, with whom we work very closely, had no other solution for this issue than not considering the re-evaporation process in the aerosol modelling. Simulations without this process lead to lower aerosol concentrations than simulations with this process, but we estimated that this relative difference was generally lower than 10%.

7. *The discrepancies between the simulated BC concentrations and the MACC reanalysis, shown in Fig. 1, seem to be inconsistent with the change of the hydrophilic emission fraction from 20 to 80%, as indicated in Sect. 2.3.1 and Table 4. Could it be that these numbers got mixed up?*

   As you noted, numbers got mixed up in the paper. The correct fraction of the hydrophilic emission fraction is 20%. We have corrected this throughout the paper.

8. *A constant set of dry deposition velocities is assumed, independent of the meteorological conditions. To improve their results, the authors have modified the applied deposition velocities compared to the original implementation. A more realistic approach would have been to extend the description by including the effect of the aerodynamic resistance. This could be mentioned in the final paragraph of the Conclusions.*

   We have added the following lines in the final paragraph of the Conclusions:

   Implementing a more realistic description of dry deposition velocities by including the effect of the meteorology through the aerodynamic resistance should also be a step forward.

9. *Please clarify in the text that DMS is emitted as sulfate precursor. In reality part of the sulfur from DMS will be removed from the atmosphere as MSA before it gets converted to SO4. Please include a short discussion of the errors one can expect when this is neglected.*

   DMS and H2S are emitted in our simple model as sulfate precursors, in addition to SO2 itself. The lifetime of the so-called sulfate precursor in our model, which is considered as one "species" only, has been approximated, as done in *Huneeus et al.* (2007) whose model was calibrated on the results of the full LOA/LMD-Z sulfur model of *Boucher et al.* (2002). We will not add details in our paper on the sulfur cycle and errors with our approximation as we think it would go beyond its purpose, but we have now added the following sentences (p 6273 line 5 of the original paper):

   As in *Boucher et al.* (2002); *Huneeus et al.* (2007), we added an H_2S source as an additional sulfate precursor, which we scaled to the SO_2 anthropogenic source (5%), and we considered a direct emission of sulfate (5% of the emitted SO_2, *Benkovitz et al.* (1996)). In summary, our model adds up SO_2, DMS and H_2S emissions in our so-called sulfate precursor.

   and in our conclusions (p 6283 l 18):

   the inclusion of a simple sulfur cycle, considering prescribed monthly distributions of chemical constituents (e.g., OH, or O_3), could yield to better concentrations of sulfate, of primary interest to climate, as processes linked to the seasonal or day/night dependence of the chemical reactions that produce sulfate, or linked to the presence/absence of clouds involved in the sulfur aqueous chemistry would then be considered.

10. *The conversion of sulfate precursors to sulfate aerosols is described by a simple exponential function with a time constant depending on latitude only. In reality, the level of oxidants will also depend on the season. For instance, the oxidation of SO2 in the gas phase occurs by reaction with the hydroxyl radical (OH), the concentration of which is strongly dependent on the amount of sunlight.*

    See the modification of our conclusions just above.

11. *Minor comments and technical corrections:*

    1. *I suggest to change "CNRM Climate Model" to "CNRM-CM6.1 global climate model" in the title.*

    We have modified the title that is now: **Development and basic evaluation of a prognostic aerosol scheme (v1) in the CNRM Climate Model CNRM-CM6**

    2. *Abstract, line 2-3: Please change "in the CNRM-GAME/CERFACS climate model" to "in CNRM-CM, the climate model of CNRM/GAME and CERFACS".* OK

    3. *Abstract, line 5-6: Remove hyphen in "sea-salt".* OK

    4. *Abstract, line 9: Change "of 2004 conditions and" to "time slice simulations for 2004 conditions and".* OK

    5. *Abstract, line 13-15 / Page 6279, line 23-25 / Page 6291, line 19-20: Why does the internal variability have little impact on the seasonal climatology of the AODs? In my opinion, low internal variability just means that small ensembles or short timeslice simulations are sufficient to calculate the model's climatology. Please clarify or rephrase.*

We have rephrased these lines as follows: **The uncertainty of aerosol type seasonal AOD due to model internal variability is low over large parts of the globe,**

6. *Page 6265, line 7: Change "largely" to "strongly".* OK

7. *Page, 6265, line 10: Remove "the" in "the aerosol-cloud effects".* OK

8. *Page 6265, line 12: Change "of aerosol distribution" to "of the aerosol distribution".* OK

9. *Page 6265, line 22: Remove "/Welcome.html".* OK

10. *Page 6266, line 13: Change "such an issue" to "this issue".* OK

11. *Page 6266, line 16: Change "simulation" to "simulations"* OK

12. *Page 6267, line 4: Change "evolution" to "upgrade".* OK

13. *Page 6267, line 6: Change "Integrated Forecast System" to "Integrated Forecasting System".* OK

14. *Page 6267, line 6-8: Change formulation to "forecast models of : : :", followed by the two institutes.* OK

15. *Page 6267, line 10: Change "specificities of" to "changes in" or something similar.* OK

16. *Page 6267, line 17-18. Change to "The land surface of ARPEGE-Climat is modelled with". The current formulation is misleading, because it suggests that SURFEX is run offline.*

We changed the formulation to : **The surface parameters are computed by the surface scheme SURFEX (v7.3), already in place for CMIP5 simulations.**

17. *Page 6268, line 6: Replace "onto" by "to", and specify the reduced Gaussian grid, e.g. "an Nxx reduced Gaussian grid".*

We changed the formulation to : **with the physics calculated to a N84 reduced Gaussian grid equivalent ...**

18. *Page 6268, line 26: Remove hyphen in "sea-salt", and change "3 size-bins particles" to "three size bins".* OK

19. *Page 6269, line 2: Change "separate a" to "separate into a".* OK

20. *Page 6269, line 3-4: Change "a sulfate precursor, named SO2, and a sulfate aerosol, named SO4, cohabit" to "a gaseous sulfate precursor, mainly representing sulfur dioxide (SO2), and a sulfate aerosol (SO4) are included". It is confusing to refer to the precursor as SO2. For instance, in Table 2, SO2 is used for sulfur dioxide only.*

We agree to that and we changed the text along your lines, specifying SO2 when adequate, and sulfate precursor when adequate.

21. *Page 6269, line 4: Change "adds up" to "adds".* OK

22. *Page 6269, line 6: Change to "The scheme describes a number of physical aerosol processes, including".* OK

23. *Page 6269, line 8: Please clarify what is meant with "as a function of the aerosol". Is there a dependence on both aerosol size and type?*

Yes, there is dependence on both the aerosol size and type, though not all 12 bins of the scheme have different deposition velocities in the current configuration of the model. We reformulated the sentence to:

assuming constant dry deposition velocities depending on the aerosol bin and on the surface type ...

24. *Page 6269, line 11: According to Sect. 2.3.1, in the original scheme described here not 80% but 20% of the black carbon is assumed hydrophilic, so these percentages should be interchanged.*

We have corrected throughout the paper the mismatch in these numbers.

25. *Page 6269, line 14: Change "chemical species" to "explicity chemistry", and "but is done along with an exponential function" to "but is done assuming exponential decay".* OK

26. *Page 6269, line 19: Change "model lowest level" to "lowest model level".* OK

27. *Page 6270, line 3-6: Move this part up, e.g. it can be included at the beginning of the third paragraph of this section. Also, change "transports" to "transport".*

We have done that.

28. *Page 6270, line 8: Change "the list" to "a list".* OK

29. *Page 6270, line 26-27. Please clarify what is meant with "The efficiency of scavenging rates corresponds to the lowest values of Textor et al. (2006)." I don't see how this is achieved, given that these efficiencies depend on the aerosol tracer, and that these are model specific.*

From table 8 of *Textor et al.* (2006), we derived the scavenging rate of the aerosols type as the "1-Interstitial" fraction of components, these components being aerosol types in our aerosol scheme.

30. *Page 6271, line 1: Change "Huneeus et al. (2007)" to "Huneeus (2007)".* OK

31. *Page 6271, line 9: Please also give the size boundaries of the bins.*

We have now the following text: **with 0.32-0.75-9.0 μm and 0.2-1.67-11.6 μm mean bin radii respectively in the GEMS/MACC and in our version (new bin boundaries of 0.01-1.0 μm, 1.0-2.5 μm, 2.5-20 μm)**

32. *Page 6271, line 27: Change "normalized" to "normalization".* OK

33. *Page 6272, line 1: Change section title to "Prescribed anthropogenic and natural emissions".* OK

34. *Page 6272, line 8: IPCC has already been introduced in the Introduction. Better to refer to the IPCC reports as "Assessment Reports" in the introduction as well.* OK

35. *Page 6272, line 18: Remove "Apart from these anthropogenic sources".* OK

36. *Page 6272, line 23-25: It is mentioned that the applied climatology of DMS emissions has the same temporal and spatial characteristics as the data set applied for volcanic emissions. If that is indeed the case, it cannot be correct. Please clarify.*

We changed the text: to **The *Kettle et al.* (1999) dimethylsulfide (DMS) climatology, emitted from the oceans, is a monthly, 1 deg. horizontal data set, and is therefore independent from the surface meteorological conditions in our simulations.**

37. *Page 6273, line 6-7: Change "considered" to "included". It should be stated that the total emitted sulfur remains unchanged.*

line 6-7: done.

As in *Boucher et al.* (2002) we scaled H2S emissions to anthropogenic SO2 emissions, adding an additional source of sulfur. We amended our text that is now as follows: **As in *Boucher et al.* (2002); *Huneeus et al.* (2007), we added an H_2S source as an additional sulfate precursor, which we scaled to the SO_2 anthropogenic source (5%), and we included a direct emission of sulfate (5% of the emitted sulfate precursor, *Benkovitz et al.* (1996)).**

38. *Page 6273, line 10: "noting that the option was rejected". It is hard to believe that the possibility that any of the sinks is overestimated, even at a regional level, can be excluded. Please adapt this statement.*

We reformulated this sentence as follows:

noting that this option was qualified as "unlikely-but possible-" by *Kaiser et al.* (2012) who also worked with the *Morcrette et al.* (2009) model,

39. *Page 6273, line 22-23: Clarify in the text why the sulfate precursor emissions are scaled and why biomass burning is excluded here. Change "excepted" to "except".*

Results presented in the paper you reviewed did include a scaling factor of 0.7 on the sulfate precursor emissions. This factor was chosen somehow hastily, and as both reviewers questioned that choice, we have rerun all our simulations without rescaling these emissions. In the end, in all diagnostics analysed in this paper, model outputs are closer to the observations in the no-rescaling case. Therefore we now show in the revised version of the article this second set of simulations. We have accordingly amended Table 2 of the paper, which shows totals of static emissions, to which we have also added a couple of references.

It has to be noted that some differences in the figures/tables between the original and the revised articles are caused by the use of a different/updated version of the atmospheric model, the aerosol model remaining unchanged. Overall, as this update was relatively minor, most of the analysis presented in the original paper is valid in the revised version.

40. *Page 6273, line 26-28: Doesn't that mean that the scaling factors applied to the emissions from biomass burning are too high. Please clarify.*

Independently from the scaling factors, the monthly biomass burning emissions we use include, very occasionally in space and time, very high grid point values, several times higher than the other relative peaks of the time series. This caused the model to compute unrealistically high AODs. We clipped these values under the maxima given in the text, paying attention that the time series still clearly showed biomass burning events, and that the total monthly emissions remained very similar.

We have amended the text as follows: **as higher values, reached very occasionally in space and time during very intensive biomass burning events or volcanic eruptions, generated unrealistic high AOD (higher than 10) in the model. The impact of this limitation on the monthly or yearly total emissions, and on most biomass burning events, is very small.**

41. *Page 6274, line 2: Change "small" to "relatively small". In fact, the impact is not that small, because doubling the amount of SOA emissions from Dentener et al. (2006) would give 38.2 Tg OM/yr, while according to Table 2 only 34.7 Tg/yr is emitted in the model. Please explain what causes this difference.*

We rephrased Page 6274, line 2, see response to comment 40.

With regards to the SOA emissions, we do not know what causes the difference between our totals and those provided by Dentener et al. (2006). This difference does not come from the limitation to peak values we use as the SOA emissions did not have such high values. We obtained this SOA data set from JJ. Morcrette and we transformed it to our model's horizontal grid, ensuring a conservative regridding, and then we computed the total emitted, which we present in the Table "Totals emitted".

42. *Page 6274, line 6: Remove "in" in "in within".* OK

43. *Page 6274, line 7: It is not true that "both the intra and inter-annual variabilities come from the biomass burning emissions". Natural emissions also have seasonal variability, and trends in anthropogenic emissions contribute to interannual variability. Please adapt the text. Also change "variabilities" to "variability".*

Thank you for this remark. The text is now as follows:

A significant part of the intra and inter-annual variabilities comes from the biomass burning emissions ...

44. *Page 6274, line 21: Change "consists in" to "consists of".* OK

45. *Page 6274, line 22: Change "with a spectral" to "with spectral".* OK

46. *Page 6274, line 25: Remove "as classically in nudged simulations", and include the reference to Zhang et al. (2011) to the end of the sentence.* OK

47. *Page 6275, line 1-2: Change "the comparison of modelled aerosols is the most realistic one" to "modelled aerosols are most realistic".* OK

48. *Page 6275, line 6: Change "Nudging, or not, the humidity" to "Whether or not humidity is nudged".* OK

49. *Page 6275, line 13: Change "that are our evaluation sets" to "used in our evaluation".*

50. *Page 6276, line 4: Please clarify if the anthropogenic emissions applied in the MACC reanalysis as the same as the data sets used in the simulations presented in this study. If so, this should be mentioned in Sect. 2.3.3. If not, what are the differences?*

We cannot say that our anthropogenic emissions (*Diehl et al.*, 2012) are the same as those used in the MACC Reanalysis (*Granier et al.*, 2011). Both datasets are based however on the *Lamarque et al.* (2010) dataset. We also cannot compute annual anthropogenic emissions from the MACC Reanalysis diagnostics as we only have access to total (anthropogenic plus biomass burning) amounts, which we show in Table "Totals emitted for static emissions...". This Table also highlights a number of differences between emissions for our simulations and emissions of the MACC Reanalysis.

51. *Page 6276, line 8-9: Were there really no sulfur emissions from volcanoes or oceans used in the MACC reanalysis? Seems strange, so please check this. Also, change "volcanos" to "volcanoes", and "no specific direct H2S or sulfate" to "no direct sulfate". Note that because there is no H2S tracer in IFS, it should be obvious that direct H2S emissions were not included in the reanalysis. Moreover, the only H2S emissions accounted for in the ARPEGE-Climat simulations are from volcanoes. If these were not included in the reanalysis, this implies that H2S emissions were not considered.*

We checked with the scientists at ECMWF, who confirmed that no sulfur emissions from volcanoes or oceans were considered in the MACC Reanalysis. We corrected the wording as you suggested. Note that our H_2S emissions are scaled to the SO_2 anthropogenic source (5%) as in *Boucher et al.* (2002); *Huneeus et al.* (2007).

52. *Page 6276, line 12: Also give corresponding resolution in degrees, as before.*

We added : **and a T255 spectral truncation corresponding to a reduced N128 Gaussian grid with a horizontal resolution of approximately 80 km (0.7 deg.).**

53. *Page 6276, line 20: Change "largely used in the modelling aerosol community" to "widely used in the aerosol modelling community".* OK

54. *Page 6276, line 21: Change to "monthly product of total AOD at 550 nm".* OK

55. *Page 6276, line 27: Change "Kan" to "Kahn"* OK

56. *Page 6277, line 6: Remove "(1o)".* OK

57. *Page 6277, line 7: Remove "courtesy of B. Koffi" and include a statement in the Acknowledgments.* OK

58. *Page 6277, line 23: Include space after "using the".* OK

59. *Page 6277, line 28-29: Include "monthly" before "climatology".* OK

60. *Page 6278, line 6: Change "issued from" to "produced by".* OK

61. *Page 6279, line 8: Remove "timescale".* OK

62. *Page 6278, line 15: Change to "As a preliminary step, we looked".* OK

63. *Page 6278, line 17: Change to "mean global monthly" to "global monthly mean".* OK

64. *Page 6279, line 3: Change "ARPEGE-Climat internal variability" to "internal variability in ARPEGE-Climat".* OK

65. *Page 6279, line 4: Change "FreSImd2" to "FreSimd2".* OK

66. *Page 6279, line 5: Change "response of" to "variability in".* OK

67. *Page 6279, line 10: Change "over west" to "west".* OK

68. *Page 6279, line 11: Include ", respectively" after "DD".* OK

69. *Page 6279, line 18: Change "of the central" to "in the central".* OK

71. *Page 6279, line 20: Remove comma after "large", include comma after "seasons".* OK

72. *Page 6280, line 4: Remove quotation marks around "bins", here and in other places.* OK

73. *Page 6280, line 13: Change "release suppressed for" to "which is suppressed in".* OK

74. *Page 6280, line 18-20: However, also the distributions of SS and DD determine the relative impact of wet scavenging.*

We have now in the text: **An explanation for that, in addition to the intrinsic distributions of SS and DD, is the smaller importance of wet scavenging on total losses for SS than for DD, with efficiencies...**

75. *Page 6281, line 10: Change "other three simulations" to "other two simulations, as well as the MACC reanalysis".* OK

76. *Page 6281, line 15: Change "of proportion of bare soil" to "for the bare soil fraction".* OK

77. *Page 6281, line 22: An enhancement factor of 20.9 is enormous. Can the authors explain why they expect it to be reasonable?*

Indeed, this factor of 20.9 is enormous. It reflects both a change in the dust emission scheme, and a change in the emitted dust size distribution (*Kok*, 2011). We have not done the four simulations that would allow us to draw conclusions about the relative importance of these changes. In the end, what is important to us is the final result with the new dust scheme and new size distribution, and that emissions look reasonable. In the end also, the NudSimd2 modelled AOD appear satisfactory compared to observations.

Please note that we corrected incorrect values in the Table "Dust emissions" of AEROCOM Median, min and max values over the globe.

78. *Page 6282, line 4: Remove "brother".* OK

79. *Page 6282, line 8-10: Please include a reference to the study where this is shown. Is it Cesnulyte et al. (2014)? Also, I would propose to already include such a statement in Sect. 2.3.2.*

No, it is *Melas et al.* (2013). We have moved the reference in the paragraph so the text is clearer.

80. *Page 6282, line 14: Change to "fairly made as an unrealistic hydrophilic/ hydrophobic fraction was assumed".* OK

81. *Page 6282, line 16: Change "tropospheric "bin" concentrations" to "tropospheric binned concentrations".* OK

82. *Page 6282, line 21: Correct "three dust bins" to "two coarser dust bins".* OK

83. *Page 6282, line 25: Change "lat-lon plots" to "global maps".* OK

84. *Page 6282, line 26-27: The authors claim that the transport is more efficient with the meteorological fields in the MACC reanalysis. However, this cannot be concluded, because also the representation of the aerosols is different, e.g. their size distributions.*

By writing that "transport away from the sources is more efficient with the MACC Reanalysis meteorology than with the meteorological conditions of our nudged simulation. " we meant that the combination of transport/sinks away from the aerosol sources lead to higher concentrations far from the sources, even with the sources being stronger in our simulations.

85. *Page 6283, line 3: Remove "within continents".* OK

86. *Page 6283, line 5: Change "SS of" to "SS in".* OK

87. *Page 6283, line 6: The claim that the new dust scheme performs much better than the original one" is not substantiated sufficiently (see main comment above).*

See our response to the main comment above.

88. *Page 6284, line 1: The fact that the model performs better in JJA than in DJF could be related to the fact that the time constant for sulphate production is assumed independent of the season (see main comment above).*

It could be, but here we have not investigated the relative importance of the various components of the AOD by aerosol type.

89. *Page 6284, line 1: Change to "mean relative bias", implying that the relative biases are averaged.* OK

90. *Page 6284, line 2: Since these percentages indicate negative biases, a minus sign should be included.* OK

91. *Page 6284, line 6-7: Please comment on the positive biases observed over the Arabian Sea and South America in summer.*

These positive biases are related in the Arabian Sea to dust aerosols and in South America to OM aerosols. We have amended our text as follows:

In contrast, the model overestimates the observations in DJF in areas such as Central Africa, parts of Saudi Arabia and Northern Africa, and in JJA over the Arabian Sea and large parts of South America. Areas of model overestimation seem to follow the trace of biomass burning in tropical regions, while dust appears overestimated over the Arabian Sea.

92. *Page 6284, line 22-25: Change "the fine mode ... aerosols)." to "and the fine mode." (see main comment above).* OK

93. *Page 6285, line 19: Change "aerosol "bin" AODs" to "binned AODs".* OK

94. *Page 6286, line 2: Change "Kanpur North India" to "Kanpur, northern India".* OK

95. *Page 6286, line 6: Change to "around 1".* OK

96. *Page 6286, line 7: Change "model outputs is that, (...), the model shows a nul to low bias" to "model is that, (...), it shows a low bias".* OK

97. *Page 6286, line 22: Change to "The underestimation".* OK

98. *Page 6286, line 24: Change to "under the influence of dust storms".* OK

99. *Page 6287, line 22 / Page 6293, line 1: A correlation coefficient of 0.5 means that 25% of the variance is explained. It would be better to use a higher threshold value to distinguish good from bad performance.*

We have changed our criteria as your suggested. The stations presented now as performing well have CC>0.7 and 0.5<rVar<1.5, while those performing poorly have the opposite criteria. Hence, three stations shown are different in the revised version of the paper.

100. *Page 6287, line 28: Change "badly" to "poorly", and remove the quotation marks.* OK

101. *Page 6288, line 1: Change "quality" to "data quality".* OK

102. *Page 6288, line 7: Remove quotation marks around "well" and "near-by". Remove hyphen in "nearby".* OK

103. *Page 6288, line 5: Change "thumbnail" to "graph".* OK

104. *Page 6288, line 9: Change "correctly" to "well".* OK

105. *Page 6288, line 10-12: Rephrase this sentence and remove the last part.*

The sentence in the revised version is now: **In these regions the model appears to perform well over large areas. Similarly, the behaviour of the model is coherent at the Taihu station in China and at the corresponding station of Xianghe (*Cesnulyte et al.*, 2014), with the same underestimation of the observations.**

106. *Page 6288, line 13-15: Why are these station names written with capital letters? If not too much work, change the names in Figure 14 and in the text.*

We have not changed the names in the figures, and therefore in the text.

107. *Page 6288, line 14: Change "badly" to "poorly".* OK

108. *Page 6288, line 17: Are all four self-references needed?*

We kept the 2013 and 2014c references.

109. *Page 6288, line 18: Change to "investigation regarding specific conditions, representativity, and quality of the site, which"* OK

110. *Page 6288, line 21: Change "repartitions of" to "component contributions to"* OK

111. *Page 6288, line 22: Change section title to "Evaluation of vertical distributions".* OK

112. *Page 6288, line 27: Change "We output" to "We show".*

We changed to : **We diagnosed ...**

113. *Page 6289, line 7: Remove "to total aerosols".* OK

114. *Page 6289, line 8: Change to "the model is biased low"* OK

115. *Page 6289, line 9: Change "quasi-nul" to "insignificant".* OK

116. *Page 6289, line 11: Please clarify why CAT is mentioned here.* OK

It is true that lines 10-12 were better placed later in this section. We have done that.

117. *Page 6289, line 17: Change "load" to "extinction".* OK

118. *Page 6290, line 5-6. Please explain why this S curve shape is not observed in the free-running simulation.*

We have no definitive idea of why the shapes of the model curves are so different. This could have been caused by different wet scavenging sinks, or by the fact that re-evaporation is suppressed in the FreSimd2 simulation, or by more active vertical transport in the NudSimd2 simulation. We do not have in hand the diagnostics to confirm, or not, these hypotheses.

119. *Page 6290, line 12: Change "correct" to "good".* OK

120. *Page 6290, line 12-14: Please also mention that the agreement is bad in all other regions.*

We rephrased our text to : **Agreement between model and observations is good for WEU, with very low extinction coefficients, and for instance for CAF in DJF or for CAT in the 2-4 km layer in JJA. Agreement is poor for other regions/layer depths such as the DJF CAT 0-2km range.**

121. *Page 6290, line 17: Please change "the climate model" to "the CNRM-CM global climate model".* OK

122. *Page 6290, line 18: Change "aerosol AODs" to "aerosols".* OK

123. *Page 6290, line 21: Change "from 2005" to "since 2005".* OK

124. *Page 6290, line 23: Change "12 bins" to "twelve tracers" (or "12 tracers", if you prefer).* OK

125. *Page 6290, line 26: Change "Large" to "Large-scale (advection)"* OK

126. *Page 6291, line 1: Change "transports" to "transport".* OK

127. *Page 6291, line 5: Change "(1995); Kok" to "(1995) and Kok".* OK

128. *Page 6291, line 7: Change "as a common" to "as is common".* OK

129. *Page 6291, line 16: Change "site as from" to "site, from".* OK

130. *Page 6291, line 19-24: In this paragraph, it should also be mentioned that there is a problem with the re-evaporation in the free-running simulation.* OK

We have amended the text as follows: **Differences in AODs between a free-running and a nudged simulation, linked to different meteorologies and to the suppression, in free-running simulations, of the release of aerosols when re-evaporation of stratiform precipitation, appear lower than 0.05 over most of the globe.**

140. *Page 6291, line 27: Change to "by 14 using the new scheme". Correct "dependant" to "dependent".* OK

141. *Page 6292, line 1: Change to "The spatial distributions".* OK

142. *Page 6292, line 4: Change "static" to "prescribed", and remove quotation marks.* OK

143. *Page 6292, line 9: Change "simulation" to "model".* OK

144. *Page 6292, line 13-17: Change to "underestimates the coarse fraction over continents, except over dust emitting areas." Remove the next sentence (see main comment above).*

We have amended the text according to the figures we show (see main comment above).

145. *Page 6292, line 18: Change "the various aerosol types" to "total AOD".*

We have provided some analysis on the aerosol types. We therefore did not change this text.

146. *Page 6292, line 22-23: Change to "a systematic low bias".* OK

147. *Page 6292, line 23: Change to "This seems to".* OK

148. *Page 6292, line 25: Change "close to 0" to "small".* OK

149. *Page 6293, line 4-5: Change to "comparing for summer and winter total and dust extinction".* OK

150. *Page 6293, line 8-9: Change to "However, most regions".* OK

The text in the revised paper is now: **A number of regions where the CALIOP interannual variability is very large (e.g., the Western China WCN region) appear really hard to simulate.**

151. *Page 6293, line 9-10: Remove "but there ... large", since this is no excuse for the discrepancies found.*

See comment 150.

152. *Page 6293, line 11: Change to "this simple prognostic aerosol scheme is promising". Given the deficiencies in the descriptions of both natural and anthropogenic aerosols, I don't think it is fair to say that the current scheme is suitable for aerosol climate studies (see main comment above).*

Ok for promising!

153. *Page 6293, line 14-18: Change sentence to "Over the continents, there is room for improvement in the modelling of SOA, and the inclusion of a simple sulfur cycle, using prescribed monthly distributions of oxidant fields (e.g., OH, O3, and H2O2), could improve the description of sulfate, which is of primary interest to climate.".* OK

154. *Page 6293, line 14-18: Please also mention that the current scheme does not describe nitrate, which is expected to be of growing importance (see main comment above).*

We have added the following sentence: **Finally, for longer term simulations, nitrate, expected to be of growing importance in the future, should also be considered.**

155. *I would propose to re-order the Tables and Figures following the order in which they appear in the text.*

We reordered the Tables following the order in which they appear in the text. This is also the case for the figures when considering the main comments related to each figure.

156. *Table 1: Please change to "dust emission scheme".* OK

157. *Table 2: Change to "Prescribed emission totals". Remove "Range" after "1993- 2012". Correct "litterature" to "literature".* OK

158. *Table 4: I suggest to include the full variable descriptions in the table entries, rather than in the caption. Change "Efficiency for scavenging" to "Efficiency for incloud scavenging", and include "by" before "rain" and "snow". Please explain the meaning of dust emission potential in the main text. Please indicate that the hydrophylic/hydrophobic ratio is applied to emissions only. This should also be clarified on page 6271, line 6. Please also indicate on page 6269, lines 9-12, that ageing of OM and BC is included using a constant conversion rate from the hydrophobic to the hydrophilic fractions, and refer to the table for the value assumed for this. What is the unit of this conversion rate given in the table? Please check it.*

Given the small amount of space available in the first column, we will not include the full variable description in the table entries. We have corrected the wording according to your suggestions.

We added in the text :**...and depends on the soil upper layer wetness, the albedo, the model's lowest level wind speed and the particle radius. It is proportional to the dust emission potential (see Table 1), which is one of the terms of the source function of *Morcrette et al.* (2009).**

We clarified the hydrophylic/hydrophobic ratio as suggested.

We also added in page 6269 : **hygroscopic growth or ageing of OM and BC is included using a constant conversion rate from the hydrophobic to the hydrophilic fractions (see Table 1),** .

The conversion rate unit (RGRATE is Table 4) is s^{-1}. It corresponds to an aging time constant of 1.63 days.

159. *Figure 1: Sea salt seems to be more sensitive to the meteorology than mineral dust. Is this explained in the text? Are the differences between the simulations and the MACC reanalysis consistent with the previous evaluations of the MACC reanalysis? Please comment in the text.*

It is hard to conclude whether meteorology has a larger impact on sea salt or on mineral dust. Relative differences in zonal means for instance (not shown in this article), appear to of the same importance over the emitting aeras. We would need much further analysis to conclude on this subject.

160. *Figure 4: Change "Mean 2004 dust AOD" to "Mean dust AOD for 2004". Is it explained in the text why the dust AOD pattern obtained with the new emission scheme is much more inhomogeneous than with the old scheme (this Figure) and the MACC reanalysis (Figure 5)? If not, please do so.*

We have added the following text in the revised paper: **The corresponding changes in AOD, for the three dust bins and the total dust aerosol are shown in Figure 4. The figure highlights also that dust AOD pattern obtained with the new emission scheme is much more inhomogeneous than with the old scheme (this Figure) and the MACC reanalysis (Figure 5). This is in better agreement with the satellite MISR and Deep Blue output (Figures 6 et 7), and it reflects the soil characteristics taken into account in the new dust scheme (see section 2.3.2).**

161. *Figures 11, 12 and 14: If it is not too much work, can the underscores be removed in the station names?*

We have not changed the names in the figures, and therefore in the text.

162. *Figure 14: Change "good performing" to "well performing".* OK

163. *Figure 15-17: Please indicate the full names of the regions, and increase the size of these Figures.*

We have increased the size of the individual plots, to the extent possible, and indicated the full names of the regions.

164. *Unless specified otherwise above, please change "specificities" to "characteristics", "specific features" or similar words, and "outputs" to "output" throughout the paper.* OK

3 Response to Anonymous Referee #2

Dear Referee,

Thank you for your review, and for your questions and suggestions, which appear below in italics, together with our responses to these remarks and suggestions. Our proposed amendments to the text of our paper appear in bold. Page and line numbers refer to the version of the paper you reviewed.

1. *General comments ...*

   ... MACC aerosol module is a simple aerosol module developed by Morcrette et al. at 2009. The transformation of SO2 into sulfate is done without any explicit chemistry. Nitrate and ammonium are not considered in the model. Lamarque et al. (2012) incorporated the online full chemistry scheme MOZART into the CESM model system. They used the Bulk Aerosol Model (BAM) coupling with MOZART to simulate sulfate, sea salt, dust, and carbonaceous species mass concentrations by considering major chemical and physical processes including emission, transport, gas phase chemistry, aqueous phase chemical reactions, dry deposition, and wet scavenging. Liu et al. (2012) introduced a modal aerosol module (MAM) in CESM which can be used to study aerosol size distribution and both internal and external mixing between aerosol components. Comparing to these previous works, the treatments of aerosol chemistry, mixing state, and scavenging in MACC aerosol module, which was incorporated in the CNRM climate model by this work, are too simple. When the authors calculate aerosol optical depth, how do they deal with particle growth caused by the uptake of nitrate, ammonium and aerosol water? And I do not find any new improvements to address these problems in this work.

   We do not claim to show a development with regards to aerosol modelling comparable to what exists in some climate models, such as the CESM model mentioned in the above comment. We present a modest development, as (1) the work was initiated from scratch just less than four years ago, and (2) we have very limited man resources to devote to this subject. Back five years ago, the CNRM climate model included, concerning chemistry, the linear scheme of *Cariolle and Teyssèdre* (2007), and prescribed aerosols. Since then, an interactive chemistry scheme has been introduced on-line (*Michou et al.*, 2011). We are not pretending to possess not even a state of the art aerosol module, but a simple scheme that would interact with other parts of the model, notably radiation, keeping the numerical cost low for multi-annual simulations, and bringing aerosols from the prescribed state to an interactive one.

   Table 9.1 in *Flato et al.* (2013) provides some insight into the characteristics of the CMIP5 models, particularly in relation to aerosols and atmospheric chemistry. Among about 40 models, half included fully interactive aerosols, while six used prescribed aerosols, and the rest used semi-interactive aerosols that consisted in climatologies calculated with a version of the model run that included an interactive aerosol scheme. For the atmospheric chemistry, 1/4 of the models included a chemistry scheme.

   Our strategy to put in place an aerorol component in ARPEGE-Climat laid upon published developments integrated in the ARPEGE/IFS system, as the atmospheric component of the CNRM climate model is based on this system. When we started this, the only choice was to go for the *Morcrette et al.* (2009) model. Since then other schemes have been introduced, or are being introduced, in the IFS or IFS related systems, see for example a description of the M7 model in EC-Earth in *Noije et al.* (2014), or of the GLOMAP system *Mann et al.* (2012) in the IFS. We may benefit from these developments in the future, although the question of the level of complexity required with regards to aerosols in climate models is still under debate (*Boucher et al.*, 2013).

2. *The consideration of aerosol and climate interaction processes is important for the implementation of aerosol module in climate model. Based on current manuscript, I cannot obtain enough information to let me understand how the authors deal with aerosol and climate interactions. I would like the authors to provide more details about the treatments of aerosol direct and indirect effects in the manuscript. And what are the values of simulated aerosol radiative forcing and cloud radiative forcing in this study? Do the authors consider the impacts of aerosol on clouds and precipitation?*

   We agree that the ultimate interest of aerosols in a climate model is the two-way interactive aerosol-radiative scheme, and indeed this article does not deal with aerosol and climate interactions. The objective here is the validation of the prognostic aerosol distributions obtained with the aerosol module imbedded in the climate model, with a one-way interaction of the meteorology upon aerosols only. A coupling of the prognostic aerosol scheme and the radiative scheme is not active here. The

framework of our simulations is that of a Chemistry Transport Model (CTM), which is the case for a number of the AEROCOM, or even ACCMIP models. The interaction of prognostic aerosols and radiative forcing in our model will be explored in another article.

The setup of our simulations is detailed in section 3.1 "Simulations", which includes the following sentences: "The simulations performed (see Table 3 for a summary) include firstly an ARPEGE-Climat simulation with 2004 conditions for all forcing, namely SST, GHG gases and climatologies of aerosols. This climatology of aerosols is the one that interacts with the radiation scheme of ARPEGE-Climat, as in the CMIP5 simulations (see *Voldoire et al.* (2012); *Szopa et al.* (2012)), and such a configuration allows an evaluation of the prognostic aerosol distribution independently from their possible impact on the meteorology."

3. *IMPROVE and EMEP have provided multi-year sulfate, BC, OC mass concentration measurements at numbers of sites over USA and Europe. I would like the authors to do the comparison of their model results with these aerosol mass observations.*

   We have added in our revised article a figure (Figure 11) that compares surface mass concentrations of SO2 and sulfate. The following text has been added in the Evaluation data section:

   The EBAS is a database infrastructure (see http://ebas.nilu.no) operated by NILU - the Norwegian Institute for Air Research - that handles, stores and disseminates atmospheric composition data generated by international and national frameworks like long-term monitoring programmes, including IMPROVE (United States Interagency Monitoring of Protected Visual Environments) and EMEP (European Monitoring and Evaluation Programme) and research projects. For this article we downloaded and processed surface concentrations of SO_2 and sulfate. These data, depending on the network, include daily, or weekly values, and for the EMEP or IMPROVE networks, which provided most of the data we used, are representative of areas away from the sources. We present in this article annual means (for 2005) from all observations available.

   The following text accompanies this figure and has been added in the paper in the 4.2.2 section, after the paragraphs dedicated to the *Kinne et al.* (2013) climatology:

   Figure 11, which compares observations and NudSimd2_Trans outputs of annual (2005) surface concentrations of SO_2 and sulfate, provides additional information on the modelling of sulfate. Correlation between model outputs and observations is better for the European sites (red dots) than for the US sites (black dots), noting that in all cases it is lower than 0.4. While for sulfate the means of observations and model outputs are very close ($\sim$0.7), for SO_2 the mean model value is twice that of the mean observed value, some of this overestimation being related to our sulfate precursor including H_2S and DMS in addition to SO_2.

4. *In this work, the authors added a new mineral dust emission parameterization into the model. This new parameterization is based on the works of Marticorena and Bergametti (1995) and Kok (2011), As shown in Table 3, simulated dust emission by the new scheme is much higher than the values simulated by the old dust emission scheme in GEMS/MACC. What are the major reasons causing such differences of predicted dust emission? In additionally, the global averaged difference of the two scheme is more than a factor of 10. According to the authors understanding, which scheme is more realistic?*

   The approach based on *Marticorena and Bergametti* (1995) and *Kok* (2011) takes into account characteristics of the soil at the horizontal resolution of the model that the ECMWF approach based on Ginoux et al. 2001 does not consider. More specifically, the former considers the clay and sand content of the soil of the model to determine a soil structure, and the soil roughness that plays a role in the erodibility of the soil, where spatially broad empirical factors are used in the latter approach developed at a time where this soil information required for the *Marticorena and Bergametti* (1995) approach was not available (*Morcrette et al.*, 2009). Furthermore, by taking into account the results of *Kok* (2011), we correct for a general drawback of GCMs that is that previous model relations could overestimate the mass fraction of the dust fine mode while underestimating the fraction of coarser aerosols. For all these reasons, our preferred approach is that of *Marticorena and Bergametti* (1995) and *Kok* (2011).

   Emissions in the ECMWF dust scheme are proportional to a so-called dust emission potential factor. For the simulations presented in this article we used a value of 1.e-11 kg s^2m^{-5}, which is the value

adopted in the latest version of the aerosol code at ECMWF, but a value of 2.e-11 kg s^2m^{-5} was used for the MACC Reanalysis and discussions with our colleagues at ECMWF revealed that a value of 5.e-11 kg s^2m^{-5} had been used in specific cases.

Emission totals with this new dust scheme appear to be at the high end of the values reported in *Huneeus et al.* (2011) for 15 AEROCOM models, with particularly high emissions over the Middle East and Australian regions. As indicated in the article, we have adopted for our simulations here a value of 5.10^{-7} for the c_α coefficient proportional to the vertical to horizontal flux ratio, involved as such in the calculation of the dust emission flux. We computed this scaling factor, which differs largely among models (*Todd et al.*, 2008), to bring our 2004 AODs in the Sahelian region, the major global source of dust, into reasonable agreement with the satellite and AERONET observations. We could have adopted various such scaling factors depending on the region, as done by other modelling groups *Tosca et al.* (2013). We will keep that in mind as further developments of our model are put in place.

Differences of predicted dust emission by the two schemes reflect both a change in dust emission scheme, and a change in emitted dust size distribution. We have not done the four simulations that would allow us to draw conclusions about the relative importance of these changes. In the end, what is important to us is the final result with the new dust scheme and new size distribution, and that emissions look reasonable. In the end also, the NudSimd2 modelled AOD appear satisfactory compared to observations.

To conclude, although we are concerned by the large differences we present in our article with regards to the dust emission loads, results presented could have been very different if we had make use of different values for the two coefficients (i.e., dust emission potential factor and c_α) described above. Nevertheless, we think that the *Marticorena and Bergametti* (1995) and *Kok* (2011) is more realistic to use in the end, for the reasons detailed above.

5. *Based on current model validations provided by the authors, I think the incorporated MACC aerosol module in the CNRM climate model did not show good performances comparing to these observations/reanalysis. The validation of monthly mean global bin concentrations with MACC Reanalysis suggested the climate model simulation significantly underestimated mass concentrations of all the 3 sea salt bins, dust within the range of 0.03-0.5 micrometer, hydrophilic BC, and sulfate. The model significantly overestimated mass concentrations of dust within the range of 0.9-20 micrometer and hydrophobic BC. Simulated mean DJF 2003-2012 total AOD by this work is only half of the values from satellite retrievals, while simulated mean JJA 2003-2012 total AOD is about 60-70% of the values from satellite retrievals. The comparison of model simulation with AERONET observations also indicated that the model significantly underestimated AOD at numbers of sites.*

   We do not fully agree to all the comments made here, and we have several arguments for that: (1) indeed, outputs of the MACC Reanalysis and from our simulations are quite different, especially in the global mean diagnostics. This is an interesting result by itself: the same aerosol code can lead to different aerosols distributions depending on the meteorological model it is implemented in. However, this affirmation should be qualified as AOD information is assimilated within the MACC Reanalysis system, while it is not in our climate model. *Kaiser et al.* (2012) emphasize the impact of this assimilation within the very same aerosol model. Furthermore, comments made above concerning on the one hand BC (hydrophilic/hydrophobic) and on the other hand the coarser dust aerosol bin need to be refined as for BC the hydrophilic/hydrophobic emission ratio has the inverse value in our simulations (the MACC Reanalysis ratio was incorrect), and for dust the overestimation is related to the use of a fully different dust emission scheme. We have made this clear in our paper. (2) Comparisons of the mean global seasonal total AOD of satellites and of our simulation lead to the large discrepancies outlined by the reviewer. However, the analysis we make at a regional scale reveals more than that overall affirmation. And finally, (3) comparisons with AERONET observations, although revealing underestimation at a number of sites, showed also that, in the climatological perspective, the very diverse annual cycles of the total AOD, with varying dominant aerosol types, were well represented by ARPEGE-Climat for all the AERONET stations of *Cesnulyte et al.* (2014) chosen to evaluate the same aerosol module in the ECMWF weather model. In conclusion, we think that we have a promising tool in hand, and further use of it in comparative analysis exercises, such as AEROCOM or AerChemMIP, will be of particular interest in the aerosol modelling world.

Special comments

6. *P6266, L3: Can the authors give some discussions about aerosol modeling in CESM, GISS, ECHAM, and UKCA?*

   We have added the following sentences in our introduction (after p6266 line 10):

   While *Liu et al.* (2012) present in their introduction a review of aerosol treatments in global climate models, from the bulk to the sectional methods, some of which have been under development for a couple of decades, *Flato et al.* (2013) provide the references for the aerosol modules of the CMIP5 climate models (see Table 9.A.1).

7. *P6268, L28: Please check the size information about dust bins.*

   We checked the size information about dust bins. We have reformulated the text as follows: **sea salt discriminates three particle size-bins (boundaries of 0.03-0.5 μm, 0.5-5 μm, 5-20 μm); desert dust also has three size-bins (0.03-0.5 μm, 0.5-0.9 μm, 0.9-20 μm)**

8. *P6269, L7: This kind of treatment cannot reflect the impact of boundary condition on dry deposition velocity which is important for tiny particles such as sulfate, BC, OC, and first bins of sea salt and dust.*

   We fully agree to this. As a comparison, in *Textor et al.* (2006) 5 out of 16 models also used constant velocities, and we are talking here of full aerosol models that do not have the same constraints as climate models with aerosol modules in terms of computer time and therefore complexity. We have however added the following lines in the final paragraph of the Conclusions:

   Implementing a more realistic description of dry deposition velocities by including the effect of the meteorology through the aerodynamic resistance should also be a step forward.

9. *P6269, L12: How do the authors deal with hydrophobic and hydrophilic species in wet scavenging. Does aerosol bin size impact wet scavenging?*

   We have moved the sentences in page 6270 lines 7-11

   "A detailed description of the original GEMS/MACC aerosol scheme appears in *Morcrette et al.* (2009), and the list of parameters of the scheme, together with the values used for the MACC Reanalysis (see paragraph 3.2.1), is given in Table 1. These parameters are fully detailed in *Morcrette et al.* (2009), and for the sake of clarity parameter names in Table 1 correspond to the ones in *Morcrette et al.* (2009). "

   towards the beginning of this section so that interested readers have this information right away.

   As for wet scavenging, the scheme makes a distinction between in-cloud and below cloud scavenging, and input parameters of the scheme include, in addition to meteorological fields such as 3D large-scale and convective precipitation fluxes or the cloudy fraction of a model grid box, the fraction of aerosol included in droplets through dissolution or impaction (D parameter in *Morcrette et al.* (2009) and in Table 1) and efficiencies with which aerosols are collected by raindrops (α parameter in *Morcrette et al.* (2009) and in Table 1). Values for the various aerosols of our scheme appear in Table 1; they are the same for the three DD and SS bins.

10. *P6269, L14: It is very questionable. How do the authors divide gas phase chemistry and aqueous phase chemistry in their study. The production rates of sulfate from the two different processes are quite different from each other.*

    Our modelling is a crude first approach of reality. We do not consider explicit chemistry in our aerosol model, not to mention the gas phase and the aqueous phase chemistries. Lifetime of the so-called sulfate precursor of our model, which is considered as one 'species' only, has been approximated, as done in *Huneeus et al.* (2007) whose model was calibrated on the results of the full LOA/LMD-Z sulfur model of *Boucher et al.* (2002). This approach has been running in a pre-operational mode at ECMWF under the GEMS and MACC auspices since 2008, and it is also part of the four latest generation of quasi-operational aerosol models of the International Cooperative for Aerosol Prediction (ICAP) programme (see *Sessions et al.* (2015)).

11. *P6272, L23: Without chemistry, how do DMS emission impact the authors' simulation. Do the authors trace DMS transport in their model?*

    No, we do not have a specific DMS tracer in our model. The DMS emissions are simply added to the SO_2 and H_2S emissions to form the emissions of our so-called "sulfate precursor" aerosol.

12. *P6273, L2: How do the authors deal with SOA in their model?*

    We deal very simply with SOA in our model. As indicated in the text of our original paper, page 6273 lines 1-3 : "as our emission scheme does not describe the SOA formation, we prescribed the SOA inventory of *Dentener et al.* (2006), representative of the year 2000." These SOA monthly emissions are considered as a source for our organic matter aerosols.

13. *P6273, L22: Why do the authors rescale the sulfate precursor emissions?*

    Results presented in the paper you reviewed included a scaling factor of 0.7 on the sulfate precursor emissions. This factor was chosen somehow hastily, and as both reviewers questioned that choice, we have rerun all our simulations without rescaling these emissions. In the end, in all diagnostics analysed in this paper, model outputs are closer to the observations in the no-rescaling case. Therefore we now show in the revised version of the article this second set of simulations. We have accordingly amended Table 2 of the paper, which shows totals of static emissions, to which we have also added a couple of references.

    It has to be noted that differences in the figures/tables between the original and the revised articles are caused by the use of a different/updated version of the atmospheric model, the aerosol model remaining unchanged. Overall, as this update was relatively minor, most of the analysis presented in the original paper has remained valid in the revised version.

14. *P6273, L26: This kind of assumption is unreasonable to me. It may cover some potential problems in the authors' model simulation.*

    We analysed these high AODs, modelled at very limited grid points, and there was no doubt that they were directly related to the biomass burning emissions at these grid points. Indeed, the monthly biomass burning emissions we use include, very occasionally in space and time, very high grid point values, several times higher than the other relative peaks of the time series. This caused the model to compute unrealistically high AODs. We clipped these values under the maxima given in the text, paying attention that the time series still clearly showed biomass burning events, and that the total monthly emissions remained very similar.

    We have amended the text as follows: **as higher values, reached very occasionally in space and time during very intensive biomass burning events or volcanic eruptions, generated unrealistic high AOD (higher than 10) in the model. The impact of this limitation on the monthly or yearly total emissions, and on most biomass burning events, is very small.**

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Development and basic evaluation of a prognostic aerosol scheme (v1) in the CNRM Climate Model CNRM-CM6

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Abstract

We have implemented a prognostic aerosol scheme ~~in the~~ (v1) in CNRM-CM6, the climate model of CNRM-GAME ~~/CERFACS climate model~~ and CERFACS, based upon the GEMS/MACC aerosol module of the ECMWF operational forecast model. This scheme describes the physical evolution of the five main types of aerosols, namely black carbon, organic matter, sulfate, desert dust and ~~sea-salt~~sea salt. In this work, we describe the ~~specificities~~ characteristics of our implementation, for instance, taking into consideration a different dust scheme or boosting biomass burning emissions by a factor of 2, as well as the evaluation performed on simulation ~~outputs~~output. The simulations consist of time slice simulations for 2004 conditions and transient runs over the 1993-2012 period, and are either free-running or nudged towards the ERA-Interim Reanalysis. Evaluation data sets include several satellite instrument AOD products (i.e., MODIS Aqua classic and Deep-Blue products, MISR and CALIOP products), as well as ground-based AERONET data and the derived AERONET climatology, MAC-v1. The ~~internal variability of the model has little impact on the seasonal climatology of the AODs of the various aerosols~~uncertainty of aerosol type seasonal AOD due to model internal variability is low over large parts of the globe, and the characteristics of a nudged simulation reflect those of a free-running simulation. In contrast, the impact of the new dust scheme is large, with modelled dust AODs from simulations with the new dust scheme close to observations. Overall patterns and seasonal cycles of the total AOD are well depicted with, however, a systematic low bias over oceans. The comparison to the fractional MAC-v1 AOD climatology shows disagreements mostly over continents, while that to AERONET sites outlines the capability of the model to reproduce monthly climatologies under very diverse dominant aerosol types. Here again, underestimation of the total AOD appears in several cases, linked sometimes to insufficient efficiency of the aerosol transport away from the aerosol sources. Analysis of monthly time series at 166 AERONET sites shows, in general, correlation coefficients higher than 0.5 and lower model variance than observed. A large interannual variabil-

ity can also be seen in the CALIOP vertical profiles over certain regions of the world. Overall, this prognostic aerosol scheme appears ~~suitable~~ promising for aerosol-climate studies. There is room, however, for implementing more complex parameterisations in relation to aerosols.

1 Introduction

Tropospheric aerosols strongly influence the climate system (*Kaufman et al.*, 2002), in multiple and complex ways because of interactions with radiation and clouds. They have been known, especially since the ~~3^{rd} and 4^{th} IPCC reports~~ Third and Fourth IPCC (Intergovernmental Panel on Climate Change) Assessment Reports (*IPCC*, 2001, 2007), and are still known to contribute ~~largely~~ strongly to the uncertainties in climate system modelling (see the Clouds and Aerosols chapter of the ~~5^{th} IPCC report~~Fifth Assessment Report, *Boucher et al.* (2013)) for several reasons, as, for instance, the quantification of ~~the~~ aerosol-cloud effects (*Lohmann et al.*, 2005) or the representation of their optical properties (*Mallet et al.*, 2013) continues to be a challenge. A more basic uncertainty can be attributed to the inaccurate representation of the aerosol distribution in the atmosphere which is highly variable in space and time because of very diverse aerosol sources, themselves suffering from very different estimations (see e.g., *de Leeuw et al.* (2011)) and of a life time shorter than a few days. This uneven distribution in the atmosphere remains hard to simulate with current climate models (*Boucher et al.*, 2013).

Although a community of global aerosol modellers has been working together for more than 10 years under the AeroCom project (Aerosol Comparisons between Observations and Models), with coordinated simulation exercises analysed in a large number of papers (see *Kinne et al.* (2006); *Textor et al.* (2006) as first papers, and http://aerocom.met.no ~~/Welcome.html~~ for a list of publications), aerosol schemes within climate models, such as those used for phase 5 of the Coupled Model Intercomparison Project (CMIP5, *Taylor* (2009)), are still undergoing development and evaluation (see e.g., *Evan et al.* (2014)). Modelling requires a fundamental understanding of processes and

their representation in large-scale models, and a number of climate models continues to consider prescribed aerosol climatologies. Such climatologies have been continuously upgraded, from *Tanré et al.* (1984) to *Kinne et al.* (2013).

More recently, the ACCMIP (Atmospheric Chemistry and Climate Model Intercomparison Project, *Lamarque et al.* (2013)) analysed the aerosol forcing of about 10 free-running global models, in contrast to AeroCom models driven by meteorological analyses, looking at past and future reference periods in coordination with CMIP5 experiments (*Lee et al.*, 2013; *Shindell et al.*, 2013). In general, these ACCMIP models have less sophisticated aerosol physics than the AeroCom models, and the issue of the added value of an explicit aerosol module as part of the climate model is still under debate (*Ekman et al.*, 2014).

While *Liu et al.* (2012) present in their introduction a review of aerosol treatments in global climate models, from the bulk to the sectional methods, some of which treatments have been under development for a couple of decades, *Flato et al.* (2013) provide the references for the aerosol modules of the CMIP5 climate models (see Table 9.A.1).

We have implemented a prognostic aerosol module within the climate model of Météo-France that takes part in CMIP exercises in order to have the requisite tool to contribute to answering ~~such an~~ this issue. This tool will also improve our knowledge about aerosol-climate interactions. In this paper, we provide a description and an evaluation of this aerosol module. We describe the underlying General Circulation Model (GCM) and the aerosol scheme in section 2, the ~~simulation~~ simulations performed together with the evaluation data used in section 3, and the results from our evaluation, with firstly intrinsic ~~specificities~~ characteristics of our simulations, and then confrontation between simulation ~~outputs~~ output and observed data sets in section 4.

2 Description of the aerosol scheme

2.1 The underlaying GCM

The aerosol scheme has been included as one of the physical packages of the ARPEGE-Climat GCM. ARPEGE-Climat is the atmospheric component of the CNRM-GAME (Centre National de Recherches Météorologiques—Groupe d'études de l'Atmosphère Météorologique) and CERFACS (Centre Européen de Recherche et de Formation Avancée) coupled Atmosphere-Ocean General Circulation Model (AOGCM) CNRM-CM, whose development started in the 1990s.

We present in this work an evaluation of the aerosol scheme driven by version 6.1 (v6.1) of ARPEGE-Climat that is an ~~evolution~~ upgrade of v5.2, fully described in *Voldoire et al.* (2012), and used to contribute to CMIP5. ARPEGE-Climat v6.1 is based on the dynamical core cycle 37 of the ARPEGE-Integrated ~~Forecast~~ Forecasting System (IFS), the operational numerical weather forecast models of Météo-France ~~/~~and the European Centre for Medium-Range Weather Forecasts (ECMWF)~~operational numerical weather forecast models~~. The major differences between v5.2 and v6.1 consist of differences in their respective physics: that of v5.2 is described in *Voldoire et al.* (2012), while the ~~specificities of~~ changes in v6.1 are in summary as follows: the vertical diffusion scheme is a prognostic turbulent kinetic energy scheme (*Cuxart et al.*, 2000), where the microphysics is the detailed prognostic scheme of *Lopez* (2002), used both for the large-scale and convective precipitation. The shallow and deep convection are those of the Prognostic Condensates Microphysic Transport (PCMT) scheme described in *Piriou et al.* (2007); *Guérémy* (2011). Further details on ARPEGE-Climat, valid for both versions 6.1 and 5.2, which concern for instance the radiation scheme, appear in *Voldoire et al.* (2012). The surface parameters ~~of ARPEGE-Climat have been computed with the external surface scheme ,~~ are computed by the surface scheme SURFEX (v7.3), already in place for CMIP5 simulations. SURFEX can consider a diversity of surface formulations for the evolution of four types of surface: nature, town, inland water and ocean. A description of SURFEX is available in the overview paper

of *Masson et al.* (2013), from the simple to the quite complex parameterisations available. We considered for this paper a configuration of SURFEX very close to the one presented in *Voldoire et al.* (2012), except for the air-sea turbulent fluxes that are those of the COARE 3.0 iterative algorithm (*Fairall et al.*, 2003; *Masson et al.*, 2013).

The interactive aerosol scheme presented below is aimed at replacing the description of the tropospheric aerosols currently in place in ARPEGE-Climat, which was used for the CMIP5 simulations and consists of 2D monthly climatologies of the AOD (Aerosol Optical Depth) of five types of aerosols, namely sea salt (SS), desert dust (DD), black carbon (BC), organic matter (OM) and sulfate (SO_4) aerosols, with a vertical profile depending on the aerosol type (see *Voldoire et al.* (2012)).

The ARPEGE-Climat configuration used here is the one that CNRM and CERFACS scientists have agreed to probably serve as the basic configuration for future CMIP6 simulations: the ARPEGE-Climat spectral model is operated in a T127 triangular truncation, with the physics calculated ~~onto a~~ to a N84 reduced Gaussian grid equivalent to a spatial resolution of about 1.4° in both longitude and latitude. The vertical description consists of 91 hybrid sigma pressure levels defined by the ECMWF, as already adopted in a number of studies with ARPEGE-Climat (e.g., *Guérémy*, 2011), which include 9 layers below 500 m and 52 layers below 100 hPa, ensuring a correct description of the vertical distribution of the tropospheric aerosols, from the surface with the aerosols emissions up to the middle troposphere where the concentration of most aerosols reaches very low values. A time step of 15 min is used for the model integration.

2.2 The original GEMS/MACC aerosol scheme

The prognostic aerosol scheme of ARPEGE-Climat is based upon the GEMS/MACC aerosol description included in the ARPEGE/IFS ECMWF operational forecast model starting in 2005 as part as the European projects Global and regional Earth system Monitoring using Satellite and in situ data (GEMS, 2005-2009, *Hollingsworth et al.* (2008)) and its follow-up projects Monitoring Atmospheric Composition and Cli-

mat (MACC, MACC-II, 2009-, http://www.gmes-atmosphere.eu/), which provide a pre-operational atmospheric environmental service to complement the weather analysis and forecasting services of European and national organisations by addressing the composition of the atmosphere.

The GEMS/MACC aerosol scheme describes the physical evolution of the five main types of tropospheric aerosols mentioned previously (*Morcrette et al.*, 2009), in which various ~~"bins "~~ bins are considered: ~~sea-salt discriminates 3~~ sea salt discriminates three particle size-bins ~~particles (radius~~ (boundaries of 0.03–0.5 μm, 0.5–5 μm, 5–20 μm); desert dust also has ~~3~~ three size-bins (0.03–0.5 μm, 0.5–0.9 μm, 0.9–20 μm); the boundaries given are for dry particles, however, the ambient humidity is taken into account in the computation of the aerosol optical properties; organic matter and black carbon separate into a hydrophilic and a hydrophobic component; and for the representation of sulfate both a gaseous sulfate precursor, ~~named~~ mainly representing sulfur dioxide (SO_2), and a sulfate aerosol ~~, named~~ (SO_4~~, cohabit in the scheme~~) are included. Hence the aerosol scheme adds ~~up~~ 12 prognostic variables to the original prognostic meteorological variables. Large-scale and parameterized transport of the prognostic aerosols, e.g. convective and diffusive transport, are done in the same way as for any meteorological prognostic field (see paragraph 2.1).

~~The scheme allows~~ A detailed description of the original GEMS/MACC aerosol scheme appears in *Morcrette et al.* (2009) , and a list of parameters of the scheme, together with the values used for the MACC Reanalysis (see paragraph 3.2.1), is given in Table 1. These parameters are fully detailed in *Morcrette et al.* (2009) , and for the sake of clarity parameter names in Table 1 correspond to the ones in *Morcrette et al.* (2009) .

The scheme describes a number of physical ~~evolutions of the aerosols~~aerosol processes, including dry deposition at the surface, assuming constant dry deposition velocities ~~as a function of the aerosol and of~~ depending on the aerosol bin and on the surface type (land, ocean, ice); sedimentation with a settling velocity depending on the aerosol ~~"bin"~~bin; hygroscopic growth ~~of BC and OM ,~~ or ageing of OM and BC is included using a constant conversion rate from the hydrophobic to the hydrophilic fractions (see

Table 1), and assuming that OM is distributed between 50% hydrophilic and 50% hydrophobic when emitted, whereas BC is distributed between 80% hydrophilic and 20% hydrophobic when emitted; wet deposition in and below clouds, from large-scale and convective precipitation, with release of aerosols when precipitation re-evaporates in the atmosphere; and conversion from SO_4 precursors into SO_4 that ~~does not consider any chemical species,~~ is done without explicit chemistry but is done ~~along with an exponential function~~assuming exponential decay, with a time constant depending on the latitude. Sources of SS and DD are calculated at each model integration using model meteorological fields. For SS, an emission flux is considered only over full ocean grids, and for their open ocean fraction only excluding a possible sea ice fraction, as a function of the wind speed at the ~~model lowest~~ lowest model level. The SS mass flux is tabulated depending on the wind speed class, based on work from *Guelle et al.* (2001) (see other references in *Morcrette et al.* (2009)). For DD, the parameterisation is derived from that of *Ginoux et al.* (2001). DD is produced over selected model grids cells, i.e., snow free, fractions of bare soil/high and low vegetation above/below given thresholds respectively, and depends on the soil upper layer wetness, the albedo, the model's lowest level wind speed and the particle radius. It is proportional to the dust emission potential (see Table 1), which is one of the terms of the source function of *Morcrette et al.* (2009) . For the other aerosols, OM, BC and the SO_4 precursors, external monthly inventories are read in. The aerosol scheme separates between the biomass burning source, in order to allow for real-time updates of that source in the IFS model (see for instance *Kaiser et al.* (2012)), and all the other sources (e.g., fossil fuel, natural sources). The inventories used for our simulations and for the MACC Reanalysis performed with the IFS system are presented respectively in sections 2.3.3 and 3.2.1.

~~Finally concerning their physical evolution, large-scale and parameterized transports of the prognostic aerosols, e.g. convective and diffusive transports, are done by the ARPEGE-Climat code in the same way as for any meteorological prognostic field (see paragraph 2.1).~~

A detailed description of the original GEMS/MACC aerosol scheme appears in *Morcrette et* and the list of parameters of the scheme, together with the values used for the MACC Reanalysis (see paragraph 3.2.1), is given in Table 1. These parameters are fully detailed in *Morcrette et al.* (2009) , and for the sake of clarity parameter names in Table 1 correspond to the ones in *Morcrette et al.* (2009) . Other papers related to this GEMS/MACC scheme address improvements of the scheme (*Morcrette et al.*, 2008), the aerosol assimilation system fully integrated into the ECMWF assimilation apparatus (*Benedetti et al.*, 2009), the Global Fire Assimilation System that calculates in real-time aerosol biomass burning emissions by assimilating observations from the MODIS instruments (*Kaiser et al.*, 2012), evaluation of all or individual aerosol distributions (*Morcrette et al.*, 2009, 2011a,b; *Huneeus et al.*, 2011; *Mangold et al.* , 2011), and finally estimations of the GEMS/MACC aerosol radiative forcing (*Bellouin et al.*, 2012).

2.3 Implementation of the aerosol scheme in ARPEGE-Climat

2.3.1 Adaptation of the scheme

Preliminary simulations with the original configuration of the aerosol scheme, with the same static prescribed emissions for BC, OM and SO_4 precursors as those for IFS runs, lead to aerosol concentrations much lower than the ones issued from IFS runs (not shown). As the literature presents a range of values for the various coefficients listed in Table 1, we adopted the values that would maximise the concentrations in ARPEGE-Climat runs. These new values are shown in red in Table 1. The efficiency of scavenging rates corresponds to the lowest values of Table 8 in *Textor et al.* (2006), whereas we got the deposition velocities from *Huneeus* (2007) and *Reddy et al.* (2005) and the settling sedimentation velocities from *Huneeus* (2007). One has to note that in this newer version of the aerosol scheme, the sedimentation process is applied only to the coarser bins of SS and DD, SSbin03 and DDbin03 in Table 1, as suggested in *Huneeus et al.* (2009). Additional information for sulfate and its precursors comes from *Boucher et al.* (2002). Lastly, the hydrophilic/hydrophopicfraction of BC has

hydrophopic/hydrophilic fractions of emitted BC have been corrected from ~~an incorrect value~~incorrect values, we now have ~~a fraction~~ fractions of 0.8/0.2 in place of the original ~~fraction~~ fractions of 0.2/0.8, and the radii of the three dust bins have been modified (P. Nabat personal communication), with 0.32-0.75-9.0 μm and 0.2-1.67-11.6 μm mean bin radii respectively in the GEMS/MACC and ~~our versions.~~ in our version (new bin boundaries of 0.01-1.0 μm, 1.0-2.5 μm, 2.5-20 μm). This size distribution adjustment was based on work done with the regional climate model RegCM (*Zakey et al.*, 2006; *Nabat e* it has been recently validated in a regional version of CNRM-CM by *Nabat et al.* (2014c) .

In addition to the adaptations presented above, developments have been made in the vertical diffusion and mass-flux convection schemes of ARPEGE-Climat (see paragraph 2.1) to account explicitly for the sub-grid transport of tracers.

2.3.2 Inclusion of an additional dust scheme

~~Based on preliminary results using the original GEMS/MACC dust scheme , and~~ Dust aerosols simulated with ARPEGE-Climat and the dust scheme described in Section 2.2 confirmed the underestimation of dust aerosols already outlined by *Melas et al.* (2013) and *Huneeus et al.* (2011) when using a similar dust scheme within the IFS ECMWF model. Therefore, as a more complex scheme could be put into place in view of the detailed ~~parameters on the soil characteristics~~ soil characteristics parameters available in ARPEGE-Climat from the ECOCLIMAP database (*Masson et al.*, 2003), an additional dust emission parameterisation has been included in the aerosol scheme, allowing for comparisons between the two ~~schemes~~parameterisations. This dust emission parameterisation comes from *Marticorena and Bergametti* (1995), which is very common in aerosol global models and takes into account soil information such as the erodible fraction and the fractions of sand and clay. The horizontal saltation flux is calculated as a function of the soil moisture, the surface roughness length and the wind velocity at the model's lowest level. The vertical flux is then inferred from this saltation flux, and the emitted dust size distribution is based on the work of *Kok* (2011). More details about this

dust emission parameterisation can be found in *Nabat et al.* (2012, 2014c). Note that the ~~normalized~~ normalization constant c_α ~~listed in *Nabat et al.* (2012)~~ proportional to the vertical to horizontal flux ratio (*Nabat et al.*, 2012) had to be adjusted to the horizontal resolution of our simulations to a value of $c_\alpha = 5.10^{-7}$. Such adjustment is common in models (*Todd et al.*, 2008) .

2.3.3 ~~Specificities of our "static"~~ Prescribed anthropogenic and natural emissions

The basis for our ~~"static"~~ prescribed emissions is the ACCMIP/AEROCOM emission inventory obtained from ftp://ftp-ipcc.fz-juelich.de/pub/emissions/accmip, fully presented and referred as the A2-ACCMIP data set in *Diehl et al.* (2012), and used in other publications (e.g., *Chin et al.* (2014); *Pan et al.* (2014)).

The A2-ACCMIP emissions are derived, for BC, primary organic carbon (OC), and SO_2, the major sulfate precursor, from the *Lamarque et al.* (2010) inventory developed for the ~~Intergovernmental Panel on Climate Change~~ IPCC Fifth Assessment Report. The original *Lamarque et al.* (2010) 1850-2000 inventory, from land-based anthropogenic sources and ocean-going vessels, in decadal increments, has been interpolated for A2-ACCMIP into yearly increments and extended beyond 2000 with the RCP8.5 (Representative Concentration Pathways) future emission scenario (*Riahi et al.*, 2011).

The A2-ACCMIP biomass burning emissions of BC, OM and SO_2 are those of the ACCMIP/MACCity biomass burning data set, which contains monthly mean emissions with explicit interannual variability and which is the original data set used to construct the decadal mean ACCMIP biomass burning emissions (*Granier et al.*, 2011). ACCMIP/AEROCOM emissions are originally at a $0.5° \times 0.5°$ resolution.

~~Apart from these anthropogenic sources, natural~~ Natural emissions of aerosols include sulfur contributions from volcanoes and oceans (*Boucher et al.*, 2002; *Huneeus*, 2007), and Secondary Organic Aerosols (SOA) formed from natural Volatile Organic Compound (VOC) emissions. We considered the SO_2 from volcanoes described in *An-*

dres and Kasgnoc (1998), which is a yearly climatology of both continuous degassing and explosive volcanoes (1° horizontal resolution). The *Kettle et al.* (1999) dimethylsulfide (DMS) climatology, emitted from the oceans, ~~has the same temporal and spatial characteristics as the *Andres and Kasgnoc* (1998) volcano~~ is a monthly, 1° horizontal data set, and is therefore independent from the surface meteorological conditions in our simulations. A review of DMS inventories, available from http://www.geiacenter.org/access/ge originals, indicates that the *Kettle et al.* (1999) data set served as the basis for other DMS inventories, and is still a valid data set to use. And finally, as our ~~emissions~~ emission scheme does not describe the SOA formation, we prescribed the SOA inventory of *Dentener et al.* (2006), representative of the year 2000. Therefore, all three data sets, SO_2 from volcanoes, DMS and SOA, do not have any interannual variability.

As in *Boucher et al.* (2002); *Huneeus* (2007), we added an H_2S source as an additional sulfate precursor, which we scaled to the SO_2 anthropogenic source (5%), and we ~~considered~~ included a direct emission of sulfate (5% of the emitted SO_2, *Benkovitz et al.* (1996)). In summary, our model adds SO_2, DMS and H_2S emissions in our so-called sulfate precursor.

As preliminary simulations of BC and OM revealed that our modeled related AODs were biased low, and keeping in mind a possible overestimation of our aerosol sinks noting that ~~the option was rejected~~ this option was qualified as "unlikely-but possible-" by *Kaiser et al.* (2012) who also worked with the *Morcrette et al.* (2009) model, we chose to augment our emissions by applying scaling factors to them. This appears to be quite a common practice in the aerosol modelling community, e.g. for BC and OM see *Kaiser et al.* (2012); *Tosca et al.* (2013), and for SOA see *Tsigaridis et al.* (2014). Noting that a factor of 1.5 exists between OC emissions, as provided in the Juelich data set, and OM emissions (see *Kaiser et al.* (2012) or *Chin et al.* (2014) and references therein), we present results in this paper having applied a factor of 2 to the original Juelich BC and OM biomass burning emissions, and to the *Dentener et al.* (2006) SOA inventory. We computed this scaling factor from MISR and MODIS observations over the two major biomass burning regions of South America and Southern Africa to bring

our 2004 AODs into reasonable agreement with the satellite data. Note that, unlike in *Tosca et al.* (2013), we did not apply factors depending on the region. ~~Finally we also rescaled the sulfate precursor emissions, excepted from the biomass burning source, with a factor of 0.7.~~

The emissions are injected into the surface layer of ARPEGE-Climat, which is about 20m thick in our 91 level configuration, and quickly distributed throughout the boundary layer by model processes such as convection and vertical diffusion. We limited the OM surface emissions to 5.10^{-9} kg m^{-2} s^{-1}, and the BC and SO_2 emissions to $5.\ 10^{-10}$ kg m^{-2} s^{-1}, as higher values, reached very occasionally in space and time during very intensive biomass burning events or volcanic eruptions, generated unrealistic high AOD (higher than 10) in the model. ~~This limitation has a small impact~~ The impact of this limitation on the monthly or yearly total emissions, and on most biomass burning events, is very small.

The resulting yearly totals emitted appear in Table 2, distinguishing the biomass burning, the natural and the other sources. Total emissions are higher in our simulations than in the MACC Reanalysis (see further details on the MACC Reanalysis emissions in 3.2.1) for all aerosols, but all our totals are ~~in~~ within the ranges provided by the literature (see also Table 2). ~~Both~~ A significant part of the intra and inter-annual variabilities ~~come~~ comes from the biomass burning emissions (not shown), with the biomass burning sources representing 49%, 54%, and 3% of the total sources for BC, OM and sulfate precursor emissions respectively in 2004, which is the reference year chosen for four of our simulations (see paragraph 3.1).

3 Simulations performed and evaluation data used

3.1 Simulations

The simulations performed (see Table 3 for a summary) include firstly an ARPEGE-Climat simulation with 2004 conditions for all forcing, namely SST, GHG gases and

climatologies of aerosols. This climatology of aerosols is the one that interacts with the radiation scheme of ARPEGE-Climat, as in the CMIP5 simulations (see *Voldoire et al.* (2012); *Szopa et al.* (2012)), and such a configuration allows an evaluation of the prognostic aerosol distribution independently from their possible impact on the meteorology. This simulation, referred as the FreSim simulation, has been repeated over 10 years to account for the internal variability of the climate model. A second simulation consists ~~in~~ of a nudged ARPEGE-Climat simulation, with ~~a~~ spectral nudging (see *Douville* (2009)) of wind, temperature, humidity and surface pressure applied every 6 hours towards the year 2004 of the ERA-Interim Reanalysis (*Dee et al.*, 2011). The motivation for this nudged simulation is twofold: first, ~~as classically in nudged simulations (*Zhang et al.*, 2011) ,~~ the nudging towards a meteorological reanalysis ensures that the simulated large-scale circulation is close to the observations and thus ~~the comparison of modelled aerosols is the most realistic one~~modelled aerosols are most realistic (*Zhang et al.*, 2011) . Second, comparing our free-running and nudged simulations will allow to estimate some possible weaknesses of the free-running simulations. In this simulation, which is called NudSim, nudging is applied to the entire atmosphere and all model levels, with a transition zone from the surface over the last five model levels, the nudging strength being fixed at a 6-hour e-folding time. ~~Nudging, or not , the humidity~~ Whether or not the humidity is nudged, led to quite different aerosol distributions, and we present here results where nudging of the humidity is applied. Two other simulations, i.e., FreSimd2 and NudSimd2 are identical to FreSim and NudSim except for the dust scheme, which is the one described in paragraph 2.3.2. Lastly, two transient simulations, with corresponding transient forcings, FreSimd2_Trans and NudSimd2_Trans, have been performed with the dust scheme of section 2.3.2 over 1993-2012. This period covers the years of the MACC Reanalysis as well as the satellite and AERONET data ~~that are our evaluation sets~~ used in our evaluation (see paragraph 3.2). NudSimd2_Trans has been nudged towards the ERA-Interim Reanalysis of 1993-2012 as with NudSim.

Another difference between the free-running and the nudged ARPEGE-Climat simulations, apart from their specific meteorology, is that release of aerosols in the ~~course~~ case of stratiform precipitation re-evaporation is not applied to the free-running simulations. Such a release led to a limited number of abnormally high AODs, which was sufficient to perturb local AODs during a couple of weeks. This issue is not caused by the wet deposition formulation itself, but appears to be linked to the characteristics of specific meteorological conditions along the vertical axis, which we do not encounter in the nudged simulations.

3.2 Evaluation data

3.2.1 The MACC Reanalysis data

The MACC Reanalysis, as part as the MACC FP-7 project is a 10-year long reanalysis of chemically reactive gases and aerosols using a global model and a data assimilation system based on the ECMWF IFS (see *Inness et al.* (2013)). Its aerosol scheme is that described in *Morcrette et al.* (2009), so it is similar to the scheme evaluated here, and its aerosol assimilation system uses MODIS AOD (*Benedetti et al.*, 2009). Anthropogenic aerosol emissions are described in *Granier et al.* (2011), while the biomass burning emissions take advantage of the Global Fire Assimilation System (GFAS) of MACC that rests upon daily fire radiative power information from the MODIS instruments (*Kaiser et al.*, 2012; *Inness et al.*, 2013). The Reanalysis used, as we did, the SOA climatology of *Dentener et al.* (2006), but did not consider any sulfur emissions from ~~volcanos~~ volcanoes or oceans, and no ~~specific direct H_2S or~~ direct sulfate emissions.

The MACC Reanalysis was performed onto 60 vertical hybrid sigma-pressure levels, with a model top at 0.1 hPa, and a T255 spectral truncation corresponding to a reduced N128 Gaussian grid with a horizontal resolution of approximately 80 km (0.7 °). Analyses of the characteristics of the simulated aerosols during this 10-year Reanalysis appear in various papers including those of ~~*Bellouin et al.* (2012); *Melas et al.* (2013); **?**); *Cesnu*~~

3.2.2 Satellite and ground-based data

We used several observation data sets that complement each other. The satellite data were obtained from the NASA Langley Research Center Atmospheric Science Data Center, and consist firstly of the MODerate resolution Imaging Spectroradiometer (MODIS), on board the Aqua satellite that is ~~largely~~ widely used in the modelling aerosol community. We used the ~~level-3 collection 5.1~~ monthly product of total AOD at 550 nm ~~monthly product~~ over the 10-year period 2003-2012 (see *Tanré et al.* (1997), *Levy et al.* (2007)) at 1° resolution, and the similar product derived from the "Deep-Blue" algorithm developed to get aerosol optical thickness over bright land areas (*Hsu et al.*, 2004). In addition, as there exist a variety of satellite aerosol products that may disagree, as analysed for instance in ~~*Bréon et al.* (2011);~~ **?** *Bréon et al.* (2011); *Nabat et al.* (201 we included in our analysis AOD data from the Multiangle Imaging SpectroRadiometer (MISR) ~~(**??**)~~ (*Kahn et al.*, 2005, 2010)) on board the Terra satellite. The MISR monthly product has the same horizontal resolution as MODIS and covers the period 2001-2012.

The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), on board the Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite, is one of the very few satellite instruments providing vertical information on the aerosol distribution. We used a level-3 global monthly gridded ~~(1)~~ 3D CALIOP product that covers the years 2006-2011, ~~courtesy of B. Koffi,~~ already introduced at the end of the *Koffi et al.* (2012) paper, and under final evaluation (see Koffi in prep and references therein). Extinction coefficients are provided at various wavelengths, under clear sky and all sky conditions, on a 1° resolution grid, every 100 m from the surface up to 10 km, for all aerosols and also distinguishing the dust component. We made analysis with the 532 nm products, in all sky conditions as *Koffi et al.* (2012) indicates that "the climatology of the mean aerosol vertical extinction distribution is not significantly affected by the presence of clouds."

The AErosol RObotic NETwork (AERONET) is a ground-based globally distributed network of automatic sun photometer measurements of aerosol optical properties every 15 min, that is a reference for AOD measurements (see *Holben et al.* (1998)). For the present work, we used AOD monthly average quality-assured data (Level 2.0, see *Holben et al.* (2006)) downloaded from the AERONET website (http://aeronet.gsfc.nasa.gov). Multiannual monthly averages are available from 1993, and we retained in our analysis stations that included five years, or more, of total AOD at various spectral bands, from which we recomputed the total AOD at 550 nm when missing in the original data set, using the Ångström coefficient. AERONET AOD data have a high accuracy of < 0.01 for wavelengths longer than 440 nm and < 0.02 for shorter wavelengths (*Holben et al.*, 1998). We derived monthly time series and a representative station climatology from 166 AERONET stations over the world that represent areas under the influence of various dominant aerosols.

The EBAS is a database infrastructure (see http://ebas.nilu.no) operated by NILU - the Norwegian Institute for Air Research - that handles, stores and disseminates atmospheric composition data generated by international and national frameworks like long-term monitoring programmes, including IMPROVE (United States Interagency Monitoring of Protected Visual Environments) and EMEP (European Monitoring and Evaluation Programme) ones, and research projects. For this article we downloaded and processed surface concentrations of SO_2 and sulfate. These data, depending on the network, include daily, or weekly values, and for the EMEP or IMPROVE networks, which provided most of the data we used, are representative of areas away from the sources. We present in this article annual means (for 2005) from all observations available.

The Max-Planck-Institute Aerosol Climatology (MAC-v1) AEROCOM/AERONET ~~climatolog~~ monthly product of aerosol optical properties takes advantage of developments in aerosol modeling and in aerosol observational capabilities. It relies on information provided by the global network of ground based sun-photometers, mostly from the AERONET network (see above), together with an ensemble of model ~~outputs~~ output

of the AEROCOM experiments. The climatology includes estimates from pre-industrial (1860) to 2100 conditions, and distinguishes between fine and coarse mode aerosols, the former with a radius from 0.05 to 0.5 microns that mostly include particles ~~issued from~~ produced by gas to particle conversion, while the latter, with a radius of up to 15 microns, include essentially sea salt and lifted soil- dust aerosols. It includes monthly ~~timescale~~ data with global coverage at a spatial resolution of 1°. Temporal evolution distinguishes between anthropogenic aerosols that include interannual changes while natural aerosols consider only seasonal variations. For further details see *Kinne et al.* (2013).

4 Results

4.1 Some characteristics of the ARPEGE-Climat simulations

4.1.1 Internal variability

As a preliminary ~~compulsory stepbefore any further analysis of the simulations~~step, we looked at the stability over time of the aerosol scheme. Figure 1 shows time series of ~~mean global monthly~~ global monthly mean concentrations, in the 1000-500 hPa layer, of the 12 prognostic aerosol ~~"bins "~~ bins over a period common to the MACC Reanalysis and our transient simulations (2003-2012). Aside from these multi-year simulations, the diagrams include pseudo time-series of the FreSim simulation that repeated 10 times the 2004 conditions.

Overall, all simulations, both nudged or free-running, show no drift over time of the aerosol concentrations. Starting with an initial state with no prognostic aerosols, equilibrium of aerosol concentrations is reached in ARPEGE-Climat simulations within the period of a month (not shown).

Figure 2 displays the interannual standard deviation (STD) of the AOD (total and five main aerosols) for JJA and the FreSimd2 simulation. This STD is a representation of

~~ARPEGE-Climat internal variability~~ the internal variability in ARPEGE-Climat, and we present this simulation and this season as the STD for the FreSim simulation has similar characteristics to those of the ~~FreSImd2~~ FreSimd2 simulation, and as the ~~response of~~ variability in the model for the DJF season is lower for all aerosols than that for the JJA season.

STD $>$ 0.01 are always under 20 to 30% of the corresponding mean value, for all aerosols (not shown). Standard deviation of the total AOD is rarely higher than 0.05, with the highest values in the biomass burning regions of Central South America (SAM) and Africa (SAF), and ~~over~~ west of India (IND), which corresponds with larger STDs for OM and DD, respectively (see Figure 2). Further insight ~~on~~ into the internal variability of ARPEGE-Climat total AOD is provided with figures of vertical profiles of extinction coefficients for total aerosols (see Figures 16 and 17) and for dust aerosols (see Figure 18). A description and analysis of these figures appear in section 4.2.3, but for the matter of interest in this paragraph we can say that larger STD in the SAF and SAM regions, related to the diverse spread of biomass burning aerosols (i.e., OM and BC), and in the Indian region (IND) in conjunction with variability in wet scavenging, appear to be consigned to altitudes below 3-4 kms. In contrast, STD of extinction coefficients ~~of~~ in the central Atlantic (CAT) region, fully explained by the values and spread in dust extinction coefficients (see Figure 18) is quite large ~~,~~ up to 5 kms. Overall, the interannual STD of the FreSimd2 simulation is lower, for all sub-regions of the globe and for both seasons, than that of the CALIOP extinction profile product.

Overall, we can conclude from this short analysis that the internal variability of ARPEGE-Climat has little impact on the seasonal climatology of the AODs, both considering all or individual aerosols.

4.1.2 The nudged versus free-running simulations

As relative differences in AOD between nudged and free-running simulations appear independent of the dust scheme (not shown), we will discuss results for the simulations with the new scheme only. Figure 1 is a first illustration of the relative behaviour of

the nudged (blue lines) versus free-running (green lines) simulations. Global monthly means of aerosol concentrations from these two types of simulations appear as distinct curves except for 3 ~~"bins"~~bins, namely the hydrophobic OM and BC, and the sulfate precursor. In the FreSimd2_Trans and NudSimd2_Trans simulations, these 3 ~~"bins"~~ bins share several common characteristics of their physical evolution, including no wet scavenging, no sedimentation, a dry deposition independent from the meteorology, and the same ~~static~~ prescribed emissions. The specific meteorologies of these two simulations, that govern sub-grid scale and large-scale transport, appear then to have little impact on the global mean monthly concentrations of these 3 ~~"bins"~~bins. For the other ~~"bins"~~bins, values are in general higher for the nudged simulation, in agreement with lower wet scavenging due to lower precipitation (not shown), and to the release of aerosols in the case of re-evaporation of precipitation ~~, release suppressed for~~ which is suppressed in the free-running simulation (see section 3.1). However, the case of ~~sea-salt~~sea salt, with global means lower for the NudSimd2_Trans simulation, illustrates the relative importance of the various sources and sinks: with both lower dynamical emissions for DD and SS in the nudged simulation (by about ~~15~~8% and 14% respectively, see Table 5), DD concentrations are higher in the nudged simulation while SS concentrations are lower. An explanation for that, in addition to the intrinsic distributions of SS and DD, is the smaller importance of wet scavenging on total losses for SS than for DD, with efficiencies for scavenging of respectively 0.2 and 0.5 (see Table 1).

Figure 3 displays differences in AOD between the NudSimd2 and the FreSimd2 simulations, for DJF and JJA of 2004. Over most of the globe, absolute differences in total AOD (first row of the figure) are lower than ~~0.05~~0.1 in DJF and 0.01 in JJA. However, differences are higher than 0.2 in DJF over central Africa, and in JJA over ~~eastern Africa~~the southern Sahelian region, the Indian Ocean and ~~small~~ spots in biomass burning regions such as Indonesia. For the former these absolute differences come from differences in the OM AOD (see second row) in relation to differences in precipitation patterns (not shown) that impact the wet scavenging in this region and season of large

biomass burning, while for the latter differences in total AOD mimic those in DD AOD (see third row).

Further insight into the behaviour of both types of simulations is provided in Table 4, which shows global annual means of the burden, residence time and ratios of various sinks of the five aerosol types for the FreSimd2, ~~with higher dust emissions in conjunction with higher winds (not shown)~~NudSimd2, and MACC Reanalysis, while an estimation of the modelling range of these quantities is provided by *Textor et al.* (2006); *Huneeus et al.* (20 Burden and residence times are higher for the NudSimd2 than for the FreSimd2 simulation for all aerosol types except SS, which is coherent with the results of Figure 1 analysed above in the same section. Values for both simulations are within the *Textor et al.* (2006); *Hur* range, except in FreSimd2 for SO_4 with too low burden and residence time, and in both simulations for SS with too large burdens. However, *Grythe et al.* (2014) report a spread of more than 70 Pg yr^{-1} in the "best" SS source functions studied, which would generate much higher burdens than those of *Textor et al.* (2006) . While the dry dep./wet dep. ratios are similar to lower for the FreSimd2 simulation than for the NudSimd2 simulation, the conv dep./wet dep. ratios are about 2 to 3 times smaller for FreSimd2, and the wet dep./total sink ratios a little larger for FreSimd2. Finally, the sed dep./dry dep. ratio, not null only for the coarser SS and DD bins, are the same for both simulations as dry deposition and sedimentation of large particles are independent from meteorology. In the end, ~~differences in AODs between a free-running and a nudged ARPEGE-Climat simulation appear relatively small~~more NudSimd2 results than FreSimd2 results shown in this table are closer to the AEROCOM means. Figures computed from the MACC Reanalysis diagnostics are also presented in Table 4 but should be taken as indicative only, as an error has been identified in the wet deposition amounts (up to 50% maximum), leading to an overestimation of the wet deposition diagnostics that results, for instance, in smaller MACC Reanalysis residence times. Apart from that error, Reanalysis burden amounts appear too high for SS and SO_4.

4.1.3 Impact of the dust scheme

Table 5 presents the mean annual dust emissions in various regions of the globe, from our four simulations of the year 2004 (see Table 3), the MACC Reanalysis, and the 15 AEROCOM global models analysed in *Huneeus et al.* (2011). The regions are also those of *Huneeus et al.* (2011). The AEROCOM range for the globe (min and max) is wide (e.g., 487-3943 Tg yr^{-1}), but ~~the~~ while the FreSimd2 and NudSimd2 ~~simulation is the only one that falls~~ simulations fall within that range, the ~~FreSimd2 simulationsmodelling higher emissions, and the other three simulations modelling~~ other two simulations, as well as the MACC Reanalysis, model lower emissions. Totals in the regions may not been consistently high (respectively low) within the same model, and our NudSimd2 simulation shows totals for the Middle East and Australia outside of the AEROCOM ranges, with particularly large emissions in Australia. This suggests that further adjustments of the scheme should be studied, and a simple adjustment could concern, for instance, the threshold of ~~proportion of bare soil~~ bare soil fraction within a grid cell required to trigger DD emissions. Such adjustments would depend on the underlying meteorology; the impact of the lowest level and surface meteorology is clearly seen with global emissions of the NudSimd2 simulation being only about ~~85~~92% of the corresponding simulation with ARPEGE-Climat meteorology (i.e., FreSimd2 simulation).

Total DD emissions are multiplied by a factor of 14 by this change of emission scheme (NudSim versus NudSimd2 simulation), knowing that factors are of 2.8, 2.9 and 20.9 for the DDbin01, DDbin02 and DDbin03 respectively. The corresponding changes in AOD, for the three dust bins and the total dust aerosol are shown in Figure 4. The Figure highlights also that dust AOD pattern obtained with the new emission scheme is much more inhomogeneous than with the old scheme (this Figure) and the MACC reanalysis (Figure 5). This is in better agreement with the satellite MISR and Deep Blue output (Figures 6 and 7), and it reflects the soil characteristics taken into account

in the new dust scheme (see section 2.3.2). In the end, the mean global total DD AOD is enhanced by ~~4.7~~4.8.

4.1.4 ARPEGE-Climat simulations versus the MACC Reanalysis

Evaluations of climate models against reanalysis ~~outputs~~ output are very common practice. The MACC Reanalysis is all the more interesting to us as we make use of a twin ~~brother~~ aerosol scheme, and as we can access in the ECMWF MARS archive diagnoses that are less common than the AODs, such as 3D individual ~~"bin "~~ bin concentrations. Evaluation results about the MACC Reanalysis indicate that the MACC system generally provides a good representation of the AOD on a monthly basis (*Cesnulyte et al.*, 2014). However, a few deficiencies have been underlined (*Melas et al.*, 2013) , such as dust being associated to too small particles, and thus being overly transported to regions very remote from the sources. Another deficiency is that sea salt seems to be overestimated and contributes to a high AOD bias in southern oceanic regions~~(*Melas et al.*, 2013)~~ .

The results of the comparison between our model ~~outputs~~ output and the MACC Reanalysis are the following, noting that for BC comparisons between the MACC Reanalysis and our simulations cannot be ~~made fairly as a wrong~~ fairly made as an unrealistic $\frac{hydrophilic}{hydrophobic}$ fraction ~~is present~~ was assumed in the MACC Reanalysis (see Table 1).

Global means of tropospheric ~~"bin"~~ binned concentrations are shown in Figure 1 for the MACC Reanalysis (red lines) and the NudSimd2_Trans simulation (blue lines). Concentrations of the various bins from our simulations are biased low compared to the MACC Reanalysis, except for the hydrophobic bins, this being possibly linked to the suppression of wet scavenging in our scheme (see Table 1), and, linked to our new dust scheme, for the ~~three~~ two coarser dust bins. Modifications of the constants of the aerosol scheme to trigger higher concentrations (see section 2.3.1), in parallel with enhancement of ~~static~~ prescribed emissions (see emission totals in Table 2), resulted in these very different global monthly means. Differences in ~~sea-salt~~ sea salt concentrations are particularly striking.

Analysis of ~~lat-lon plots~~ global maps of AODs (see Figure 5) reveals that transport away from the sources is more efficient with the MACC Reanalysis meteorology than with the meteorological conditions of our nudged simulation. In the end, lower global mean values of the NudSimd2_Trans simulation in Figure 1 are caused by lower concentrations away from the source regions. This is the case for all smaller aerosols with no or little sedimentation, and is clearly visible for instance for BC, OM and sulfate. In the case of SS, in addition to long-range transport ~~within continents~~ characteristic of the MACC Reanalysis, concentrations or AODs are larger in the MACC Reanalysis even at the source regions with higher emissions (64.2 versus 51.6 Pg year^{-1}). However, as SS ~~of~~ in the MACC Reanalysis seems to be overestimated (see above), we chose to go along in this paper with our modeled SS distributions.

Finally, these results can also be explained by the role of the aerosol assimilation present in the MACC Reanalysis that significantly modifies aerosol concentrations and improves agreement with observations as compared to control runs without aerosol assimilation (*Kaiser et al.*, 2012; *Melas et al.*, 2013).

In summary to conclude this 4.1 section, as we demonstrated that (1) in a climatological perspective ARPEGE-Climat free-running and nudged simulations show little differences, and (2) the new dust scheme performs much better than the original one, we will then go along in the remainder of this paper with analysis of the NudSimd2_Trans simulation only against observations.

4.2 ARPEGE-Climat simulations versus satellite and ground-based data

4.2.1 Total AOD

Figures of total AOD (Figures 6 and following) show DJF and JJA means over 2003-2012 of the three satellite data sets, i.e., MODIS Aqua standard and Deep-Blue products and MISR, of our NudSimd2_Trans simulation, and of the *Kinne et al.* (2013) climatology representative of the year 2000. The main spatial patterns as well as the local seasonal cycles of the total AOD in various regions of the globe, in conjunction

for instance with JJA dust emissions in Northern Africa or the Middle East, or biomass burning in Central Africa, or sea salt production in the southern oceans, are clearly depicted by the model. However, overall model ~~outputs~~ output underestimate satellite observations, noting that the three satellite data sets may greatly disagree over large areas. ~~This~~

In the case of MISR, which has the largest spatial coverage of the satellite data we used, the model underestimation is lower in JJA than in DJF, with a ~~relative mean bias between MISR and the simulation of 47~~mean relative bias of -41% and ~~56~~-52%, respectively (see Figures 8 and 9). This low bias is mainly driven by the oceanic values. In contrast, the model overestimates the observations in DJF in areas such as Central Africa, parts of Saudi Arabia and Northern Africa, or in JJA over the Arabian Sea or large parts of South America. Areas of model overestimation seem to follow the trace of biomass burning in tropical regions, while dust appears overestimated over the Arabian Sea. Over continents in JJA, at mid to northern latitudes, the bias appears quite patchy, with both positive and negative values.

~~Relative biases between model outputs and the other two satellite data sets, i.e. the MODIS Aqua and the Deep Blue products, yielded different results.~~ MISR and MODIS differ by more than ~~than~~ 20% over large parts of the oceans, and they contrast even more over continents (not shown). The same comment applies to MODIS Deep Blue over continents, and is even more true for the *Kinne et al.* (2013) climatology. As a consequence, relative biases between model output and the other two satellite data sets, i.e. the MODIS Aqua and the Deep Blue products, yielded different results, see Figures 8 and 9. This is particularly the case over South America and Australia with large areas of observed low AODs (lower than 0.1). Over mid to high latitude oceans, the bias between *Kinne et al.* (2013) and our simulation is lower (around 10 to 50%~~, not shown~~) than the bias between MISR and our simulation (around 30 to 70%).

4.2.2 Fractional AOD

Figure 10 shows several fractions of the annual mean total AOD, for the *Kinne et al.* (2013) climatology, representative of the year 2000, and the NudSimd2 SUBSCRIPTNBTrans simulation. Fractions are those available in *Kinne et al.* (2013), and we grouped our aerosol scheme ~~"bins "~~ bins to comply to the extent possible to these fractions. Total AOD has been separated in AOD from the coarse mode (the two largest of the three bins of SS and DD in our simulations, not shown), the fine mode, that complements the coarse mode, the anthropogenic sulfate aerosols (in our case sulfate from all sources, including natural sources such as oceans or volcanoes), and the natural aerosols (in our case DD and SS aerosols). This grouping may not appear fully satisfactory, the anthropogenic sulfate aerosols would for instance have been best identified running a supplementary simulation with pre-industrial conditions (*Schulz et al.*, 2006; *Myhre et al.*, 2013) , or applying more complex grouping methodologies such as in *Bellouin et al.* (2012); *Sessions et al.* (2015) , but the comparison detailed below is intended as a first estimation of our model output.

Higher coarse-mode AODs are associated with dust (e.g. Northern Africa) and sea salt (e.g., Southern oceans), whereas higher fine-mode AOD contributions are registered over regions of urban pollution and regions affected by biomass burning. As these two modes complement each other, a model underestimation of the former goes with a model overestimation of the latter, and vice-versa. In general, the model ~~underestimates the coarse~~ overestimates the fine mode fraction over continents ~~(by 40~~and at high latitudes(by 20% or more), except for the very northern part of Africa, the Mongolian desert region, and the tropical Pacific ocean. The comparison is better of oceans, with large areas within 20% of the *Kinne et al.* (2013) climatology, the northern tropical Atlantic excepted.

The sulfate fractions of the total AODs of *Kinne et al.* (2013) and the NudSimd2 SUBSCRIPTNBTrans simulation show similarities in their hemispheric repartition, with fractions lower than 0.3 in most of the Southern hemisphere. Over Europe and the

United States, however, our fractions appear too high (by ~~more than two times~~20% to 80%). This is also the case over regions in pristine air affected only by volcanoes, such as the Hawaiian Islands or the Antarctic continent (Mount Erebus volcano), which is coherent with the *Kinne et al.* (2013) sulfate fraction consisting of anthropogenic sulfate only.

Finally, the fraction of natural aerosols is correctly simulated over the oceans and dust-producing regions. Over the rest of the continents, we underestimate this fraction (by 60% to 90%) as we could not include in this fraction the contribution from second organic aerosols, which ~~are~~ is not a simulation output.

Figure 11, which compares observations and NudSimd2 SUBSCRIPTNBTrans outputs of annual (2005) surface concentrations of SO_2 and sulfate, provides additional information on the modelling of sulfate. Correlation between model outputs and observations is better for the European sites (red dots) than for the US sites (black dots), noting that in all cases it is lower than 0.4. While for sulfate the means of observations and model outputs are very close (~0.7), for SO_2 the mean model value is twice that of the mean observed value, some of this overestimation being related to our sulfate precursor including H_2S and DMS in addition to SO_2.

To go further in the evaluation of the various fractions of the total AOD, Figure 13 presents, for the selection of twelve AERONET stations as in *Cesnulyte et al.* (2014), the monthly climatological AOD at 550 nm, computed over all years of data available at each given AERONET station. The NudSimd2_Trans ~~aerosol "bin"~~ binned AODs, at the locations of the AERONET sites, appear in the same figure grouped into SS, DD, OM, BC and SO_4 AODs, in addition to the AERONET total AOD, and allow then for an evaluation of the various fractions of the total AOD. These AERONET sites cover various parts of the globe (see Figure 12 for their locations), and are categorized in three groups depending on the typically dominating aerosol type: urban/anthropogenic for the Ispra, Kanpur, La Jolla, Thessaloniki and XiangHe sites; biomass burning for the Alta Floresta and Mongu sites; and dust for the Capo Verde, El Arenosillo, Ilorin, La Parguera and Solar Village sites.

The annual cycle of the total AOD is generally well represented by the model, with either a unique narrow peak during the year, such as at the biomass burning site of Alta Floresta in South America, or a peak over several months such as at the dust site of Solar Village in Saudi Arabia, or two peaks as in Kanpur~~North~~ , northern India, which coincide with the pre and post-monsoon seasons. The model is also able to capture the range of AODs covered by this selection of areas, going from total AODs lower than 0.2 all year round at La Jolla or El Arenosillo, to medium AODs (around 0.5 in Capo Verde), and to large AODs ~~of~~ around 1 (Alta Floresta). Another characteristic of the model ~~outputs~~ is that, in almost all cases, ~~the model shows a nul to low biascompared to the observations~~it shows a low bias.

The low bias is particularly important for the Ispra site (mean yearly bias-MB- of ~~0.13~~0.11), with sulfate as the dominant aerosol all year round in observations (*Cesnulyte et al.*, 2014), as it is also the case in the model ~~outputs. However, underestimation of model sulfate here is in disagreement with the overestimation of the sulfate fraction in comparison to the *Kinne et al.* (2013) climatology described earlier in this section.~~ output. This underestimation could be questioned as the data quality score of *Kinne et al.* (2013) is moderate only for this ISPRA site, the remaining of the *Cesnulyte et al.* (2014) sites having an excellent quality score. Furthermore, the two nearby sites at the regional scale, Thessaloniki and El Arenosillo, show much better agreement between the model and the observed climatologies, ~~even though~~ noting however that the dust and sulfate contributions differ for all three sites, as for instance El Arenosillo can be affected by dust storms from Northern Africa.

The two Asian sites of Kanpur and XiangHe are also affected by high pollution, and large observed AODs (larger than 0.4) prevailing all year round are underestimated in our simulation by a factor of ~~2. Underestimation~~ 1.8. The underestimation is even larger at Ilorin (MB=~~0.4~~0.38), located in sub-Saharan Africa, particularly in the dry season months from November to April. This site is obviously under the influence of dust storms, but *Cesnulyte et al.* (2014) indicate that fine aerosol from biomass burning

make a significant contribution during this dry season, which is a contribution that we seem to be underestimating.

At the two shore/ocean sites of La Jolla (Pacific shore) and of La Parguera (Caribbean Islands), with relatively clean air all year round (total AOD lower than 0.25), the model underestimation appears related to an underestimation of the dust AOD, with dust transported from the nearby Mojave or further away Saharan deserts, respectively (*Cesnulyte et al.*, 2014).

Nevertheless, agreement between model and observations is particularly good at the two biomass sites of Alta Floresta in South America and of Mongu in South Africa, which is more of a savannah region. This is also the case at the two dust sites of Solar Village, in the heart of the Arabian Peninsula, with a small negative MB of ~~-0.05~~-0.07, and of Capo Verde located ∼730 km of the Senegal coast. The dust transport seems well represented here, although slightly underestimated (MB=~~0.11~~0.09).

As an overall performance of the NudSimd2_Trans simulation, we present in Figure 14 a Taylor diagram (*Taylor*, 2001) computed from the time series of the 166 AERONET stations we retained in our analysis (see paragraph 3.2.2) and of the corresponding simulation ~~outputs~~ output at the station location. These time series could in principle cover the 1993-2013 period, but the time period covered is shorter in most cases. Stations have been qualified according to the dominant aerosol type~~(~~, ocean, mountain, polar, biomass, coastal, dust, polluted, and land, see *Kinne et al.* (2013)~~), and they are numbered on the diagram within each category~~. The most common locations are land (46 stations), coastal (26), and polluted (25). For graphical purposes, negative correlation coefficients have been set to zero, and normalized standard deviations higher than 1.75 have been set to 1.75. Overall, the model performs rather satisfactorily with regards to the time correlation between observed and modelled values: the majority of series have correlation coefficients higher than 0.5 (118 stations), this coefficient being higher than 0.7 for 64 stations. With regards to the variability of the series, the diagram reports on the ratio between model and observed standard deviations, and indicates that this ratio is below 0.5 for ~~a significant number of stations(47). However, this ratio is~~

higher than 1 for the same number of 29 stations, while it lies between 0.5 and 1.5 for 122 stations.

To further illustrate the behaviour of the model at the monthly time scale, Figure 15 shows monthly times series, with the same representation of the AOD as in Figure 13, over all years of data available at a given AERONET site. Included is a selection of six stations performing particularly "badly" according to the Taylor diagram of Figure 14poorly (CC<0.5 or rVAR<0.5 or 1.5<rVAR), followed by a selection of stations performing "well" well (CC>0.5 0.7 and 0.5<rVAR<1.5). According to *Kinne et al.* (2013), all of these stations have a data quality score of 3 (excellent), and a representativeness score varying between 900 km to 100 km. This selection addresses several dominant aerosol types and locations in the world (see Figure 12).

Out the 166 total AERONET sites none of the 5 sites qualified as ocean sites perform "well". The Bermuda thumbnail illustrates that here again The Tahiti graph illustrates here again the poor performance of the model over oceans: as in the La Parguera case (see above in the same section), the model is all the time two low, and it misses higher levels of AOD. The Dhadnah and Grande SONDA cases (qualified as performing "well"well) confirm the good climatologies seen for the relatively "near-by" nearby stations of Solar Village and Alta Floresta of *Cesnulyte et al.* (2014). In these regions the model appears to perform correctly well over large areas. The same comment can apply to the Similarly, the behaviour of the model is coherent at the Taihu station in China , and the correspondent and at the corresponding station of Xianghe in *Cesnulyte et al.* (2014) , changing performing "well" to "badly"(*Cesnulyte et al.*, 2014) , with the same underestimation of the observations.

In contrast, while the three stations of IMS-METU-ERDEMLI, OHPSUBSCRIPTNBOBSERV and Bersk perform badlyToulon, and Belsk perform poorly, either because of a poor CC, or a poor rVar, the Moldova Villefranche station located in the same region of the world performs well. This underlines the challenge of modelling aerosols in that Euro-Mediterranean region (?*Nabat et al.*, 2014a,b,c) (*Nabat et al.*, 2013, 2014c) . The case of Arica, with a MB of 0.22 and an rVar of 0.08 0.30 requires further investigation

~~(specificities/representativity/~~regarding specific conditions, representativity, and quality of the site~~) that~~ , which goes beyond the scope of this paper. And to finish on this comparison, particularly difficult for a climate model, the two cases of Halifax and Lake Argyle, with very different ~~repartitions of~~ component distributions to the total AOD, but with similarly good results, are encouraging.

4.2.3 ~~Vertical evaluation~~Evaluation of vertical distributions

Figures 16 and 17 display mean vertical profiles of total extinction coefficients (km^{-1}) for DJF and JJA, respectively, averaged for individual years. These years cover the 2006-2011 period for the CALIOP instrument, and are representative of the 2004 year for the FreSimd2 simulation (previously mentioned in paragraph 4.1.1) and the NudSimd2 simulation. We ~~output~~ diagnosed vertical information to compare with the CALIOP data from these two simulations only. Profiles are presented for the 12 regions displayed in *Koffi et al.* (2012), representative of regions with a dominance of marine aerosols (NAT, CAT and NWP regions), of industrial aerosols (EUS, WEU, IND and ECN regions), of dust aerosols (NAF and WCN regions), and of biomass burning aerosols (SAM, CAF, and SAF regions). In addition to these figures, Figure 18 shows vertical profiles of dust extinction coefficients (km^{-1}), for the same simulations/observations as Figures 16 and 17, for DJF and JJA, and for the six *Koffi et al.* (2012) regions with a significant contribution of dust aerosols~~to total aerosols~~.

In general, ~~model outputs are~~ the model is biased low compared to the CALIOP data, except for the North Africa region (NAF), which presents ~~a quasi-nul~~ an insignificant bias in DJF and a positive bias between 0.03 and 0.09 km^{-1} depending on the altitude. The ~~seasonality in the vertical profiles of NAF and CAT appears clearly in the modeland in the observations, with dust at higher levels due to transport from easterly winds reaching up to 6 km, and advection of the Saharan dust to the Atlantic between 2 and 5 km (see Figure 17).~~

~~The model~~model's low bias is particularly marked for the CAT, WCN, SAF and IND regions. For CAT, the marine boundary layer aerosol load is clearly underestimated

in both seasons. This is also the case for the marine NWP region in ~~JJA~~DJF, but this marine aerosol ~~load~~ extinction is correctly simulated in the North Atlantic (NAT) region. For the dust area of Mongolia (WCN), *Koffi et al.* (2012) indicate that significant CALIOP versus MODIS AOD discrepancies are obtained e.g., for the WCN West China dust region DJF bias = +128% and SON bias = +74%. Particularly high inter-annual variability observed for this WCN region could be due both to its reduced size and to the high variability of the processes responsible for the uplift of the dust particles. *Koffi et al.* (2012) report a particularly large inter-model (12 model analysed) range for this region of WCN in DJF, probably linked to unresolved processes such as wind gusts, which are not taken into account in our dust emission schemes. The Southern Hemisphere biomass burning South Africa (SAF) low extinction profiles ~~result probably in JJA from an underestimation of the fires, although vertical transport of the fire aerosols appears clearly in~~ seem clearly related to the meteorology, including vertical transport and loss by precipitations, as the nudged and free running profiles differ quite a lot. Such a difference appears in JJA over most domains we show. Finally, with regards to profile shapes, the model depicts rather well the convex character of the SAF profiles in JJA, although we do not represent in our model the ~~characteristic seasonal shape of the profiles. *Koffi et al.* (2012) indicate that a potential factor contributing to aerosol at high altitudes is the~~ formation of secondary aerosols from the biomass burning gaseous products during plume aging ~~. This process is not represented in our aerosol scheme.~~

~~The convex character of the mean JJA profiles in SAF for example~~ that contributes to aerosol at high altitudes (*Koffi et al.*, 2012) . The seasonality in the vertical profiles of NAF and CAT appears clearly in the model and in the observations, with dust at higher levels due to transport from easterly winds reaching up to 6 km, and advection of the Saharan dust to the Atlantic between 2 and 5 km (see Figure 17)~~is, however, rather well depicted by the model. Finally~~. And lastly, for the Indian industrial region (IND) ~~,~~ the NudSimd2 simulation generates an S curve shape in JJA that appears quite unique and could be related to an overly large wet deposition sink.

Figure 18, which depicts dust only extinction profiles, provides further insight into the model behaviour: the North Africa (NAF) profiles in Figure 18, when compared to the profiles of Figures 16 and 17 confirm that dust is the predominant aerosol in that entire region. This also appears to be the case, although to a lesser extent, in the boundary layer for the Western China (WCN) region in DJF, but is not at all the case for the other regions and/or seasons. Agreement between model and observations is ~~correct~~ good for WEU, with very low extinction coefficients, ~~for~~ and for instance for CAF in DJF ~~and finally~~ or for CAT in the 2-4 km layer in JJA. Agreement is poor for other regions/layer depths such as the DJF CAT 0-2km range.

5 Conclusions

We have introduced a prognostic aerosol scheme (v1) within the atmospheric component ARPEGE-Climat of the ~~climate model of CNRM/GAME - CERFACS climate model~~ CNRM-CM6 climate model (*Voldoire et al.*, 2012). Until now, ~~aerosol AODs~~ aerosols were prescribed to the model as monthly AODs.

This scheme is based on the GEMS/MACC aerosol module included in the ARPEGE/IFS ECMWF operational forecast model ~~from~~ since 2005 (*Morcrette et al.*, 2009), which describes the physical evolution of the five main types of aerosols, BC, OM, DD, SS and sulfate. A total of 12 ~~bins~~ tracers are distinguished in the parameterisations of the physical evolution of the aerosols, which include dry and wet deposition, sedimentation, hygroscopic growth, conversion for sulfate precursors into sulfate, and dynamical emissions of dust and sea salt. ~~Large~~ Large-scale (advection) and sub-grid scale (i.e., diffusion and convection) ~~transports~~ transport of these additional prognostic fields of the atmospheric model are also considered.

We implemented a number of changes in the original scheme, such as modifications of the constants involved in the various parameterisations and addition of a new dust emission scheme based on ~~*Marticorena and Bergametti* (1995); *Kok* (2011)~~ *Marticorena and Kok* (2011) . Furthermore, biomass burning emissions of OM and BC and emissions of

SOA have been rescaled (*Kaiser et al.*, 2012; *Tsigaridis et al.*, 2014), as ~~a~~ is common practice in aerosol modelling, by a factor of 2. These changes were aimed at enhancing preliminary low concentrations from our simulations.

We performed a number of simulations to evaluate different aspects of our modelling of aerosols such as the internal variability of the climate model, the behaviour of free-running simulations versus nudged simulations, and the sensitivity to the dust emission scheme. Then, transient (1993-2012) simulations were aimed at validating the model, in a climatological way, from the seasonal to the monthly time scale, against satellite observations, available over the entire or part of the 2003-2012 period, against in-situ AERONET measurements, available, depending on the site~~as~~ , from 1993, and against the *Kinne et al.* (2013) global climatology that relies on information from the AERONET stations.

The internal variability of the model has little impact on the seasonal climatology of the AODs of the various aerosols. Differences in AODs between a free-running and a nudged ~~simulations~~ simulation, linked to different meteorologies and to the suppression, in free-running simulations, of the release of aerosols when re-evaporation of stratiform precipitation, appear lower than 0.05 over most of the globe. Higher differences (> 0.2) exist in conjunction with large AODs of biomass burning emitted OM in DJF or of dust in JJA. In the end, the performance of a nudged simulation is comparable to that of a free-running simulation.

Analysis of simulations differing by the dust emission scheme alone revealed large differences in both emission fluxes and dust AODs. For the former, global dust emissions are multiplied by 14 using the new scheme, realising that this factor is ~~dependant~~ dependent on the region. This factor varies also according to the dust bin size, and to this end global mean dust AOD is enhanced by a factor of 4.7.

~~Spatial distribution~~ The spatial distributions of aerosol concentrations and resulting AODs of, on the one hand, the MACC Reanalysis of reactive gases and aerosols and, on the other hand, our simulations are quite dissimilar, even though the two underlying GCMs share very close aerosol modules. Higher emissions, both dynamic and

~~"static"~~prescribed, and parameters of the aerosol scheme tuned to reduce aerosol sinks resulted in much lower aerosol concentrations (AODs) away from the source regions in our simulations.

Overall patterns and seasonal cycles of the total AOD are well depicted by our nudged transient simulation when compared to the satellite AOD. Over oceans, however, the ~~simulation~~ model has a systematic low bias, of varying importance depending on the observational data set. Over continents, differences are more diverse with patches of low and high biases.

We compared portions of the total simulated AOD with the fractions described in the *Kinne et al.* (2013) climatology. In general, the model underestimates both the coarse and the natural fractions over continents, except over dust-emitting areas. For the natural fraction, this could reflect different aerosols types being considered within the category. On parallel, it appears to overestimate the sulfate fraction over industrialised countries of the Northern Hemisphere.

Evaluation of the various aerosol types has also been performed against AERONET observations of total AOD at 550nm. Monthly climatologies computed over all years of data available at a given site have been examined at the 12 sites of *Cesnulyte et al.* (2014). The very diverse annual cycles of the total AOD, with varying dominant aerosol types, are well represented by the model. However, the model shows ~~systematically a~~ a systematic low to null bias compared to AERONET observations. ~~Underestimation~~ This seems to be linked to missing local sources such as biomass burning, or missing more distant sources such as dust transported over the entire Atlantic ocean. Biases are ~~close to 0~~ small at true biomass burning or dust sites.

To go further in qualifying/quantifying the aerosol prognostic scheme, monthly time series of the 166 AERONET sites that add up to 5 years or more of measurements have been compared to model ~~outputs~~ output at the corresponding grid cells. The majority of series have correlation coefficients higher than 0.5, but generally lower variance for the model. Selected time series confirm the difficulty in modelling aerosol at the local scale, but outline also the good performance of the model in certain cases.

Finally, an evaluation of the vertical profile has been performed comparing ~~seasonal,~~ for summer and winter total and dust ~~,~~ extinction coefficients from the CALIOP instrument (2006-2011) and from the model, over the regions analysed in *Koffi et al.* (2012). The model generally has a low bias, except for the North Africa region where the bias is high. The distinct shape and seasonality of the profiles are rather well represented by the model. A ~~couple of regions appear really hard to simulate~~ number of regions where the CALIOP interannual variability is very large (e.g., the Western China WCN region) ~~, but there the CALIOP interannual variability is very large~~appear really hard to simulate.

The evaluation described here indicates that this prognostic aerosol scheme is ~~suitable~~ promising for aerosol-climate studies. We suggest that remaining issues could be addressed by improving aerosol distributions over oceans. This could result from a different ~~sea-salt~~ sea salt emission scheme, or by considering a parameterisation of DMS emissions. Over ~~continents, apart that~~ the continents, there is room for improvement in the modelling of SOA, and the inclusion of a simple sulfur cycle, considering prescribed monthly distributions of chemical constituents (e.g., OH, or O_3), could ~~yield to better concentrations~~ improve the description of sulfate, which is of primary interest to climate~~.~~, as processes linked to the seasonal or day/night dependence of the chemical reactions that produce sulfate, or linked to the presence/absence of clouds involved in the sulfur aqueous chemistry would then be considered. Implementing a more realistic description of dry deposition velocities by including the effect of the meteorology through the aerodynamic resistance should also be a step forward. Finally, for longer term simulations, nitrate, expected to be of growing importance in the future, should also be considered.

Code availability: A number of model codes developed at CNRM, or in collaboration with CNRM scientists, is available as Open Source code (see https://opensource.cnrm-game- However, this is not the case for the aerosol code presented in this paper. This code is nevertheless available upon request from the authors of the paper.

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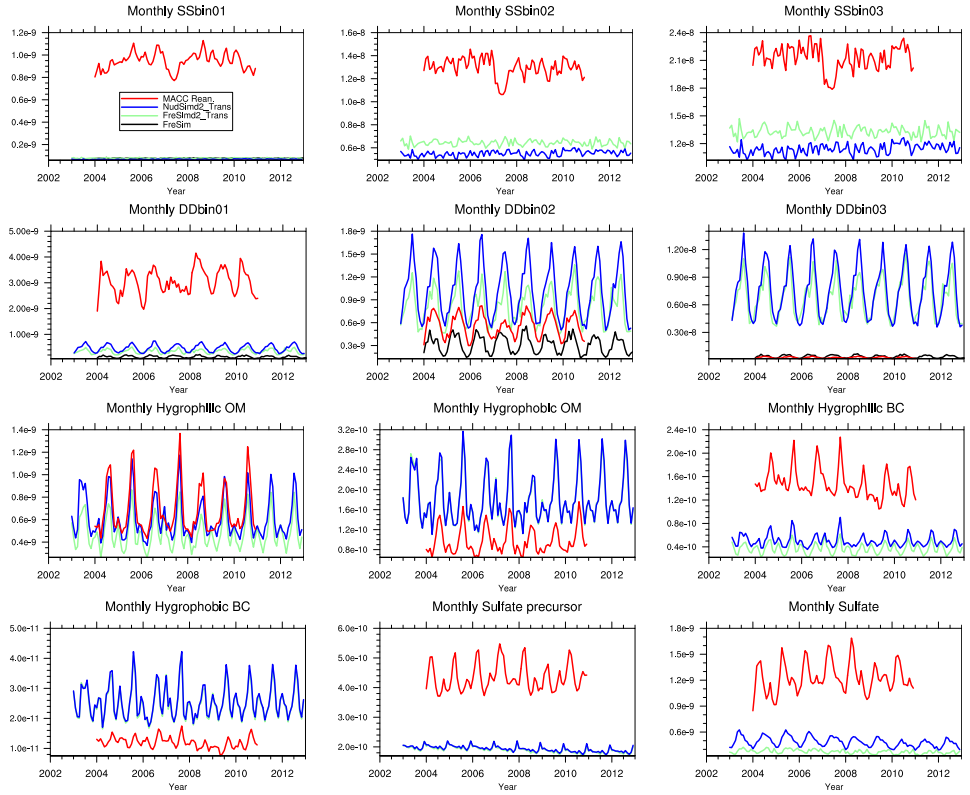

Fig. 1. Time series of monthly mean global bin concentrations (kg kg^{-1}) in the lower troposphere (1000 to 500 hPa layer) for the FreSimd2_Trans (green line), NudSimd2_Trans (blue line), and MACC Reanalysis (red line). In addition, dust bin concentrations are added for the FreSim simulation (black line, 2004 repeated 10 times). The 12 "bins" of the aerosol scheme are shown.

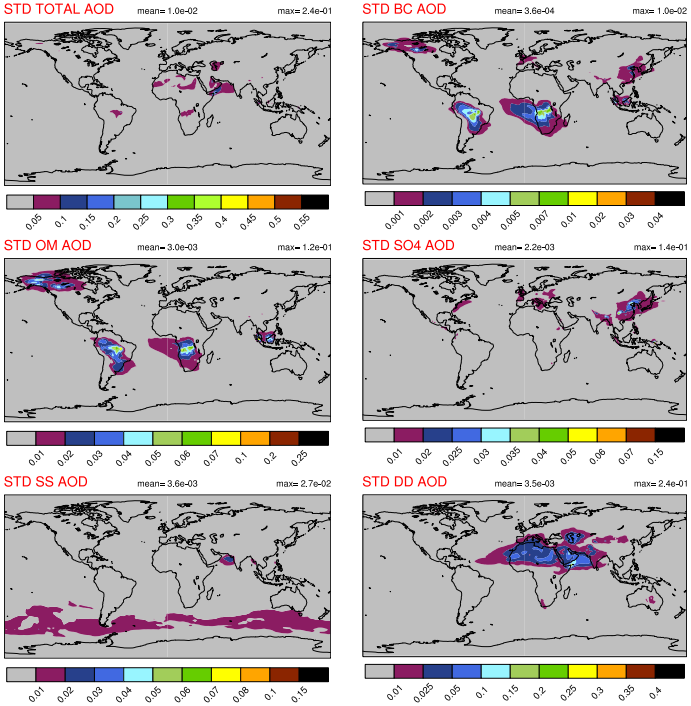

Fig. 2. Mean standard deviation for JJA for the FreSimd2 simulation, as a representation of the ARPEGE-Climat internal variability, of the total, BC, OM, sulfate, SS, and DD AODs. Color scales are the same as in Figure 5 and 7.

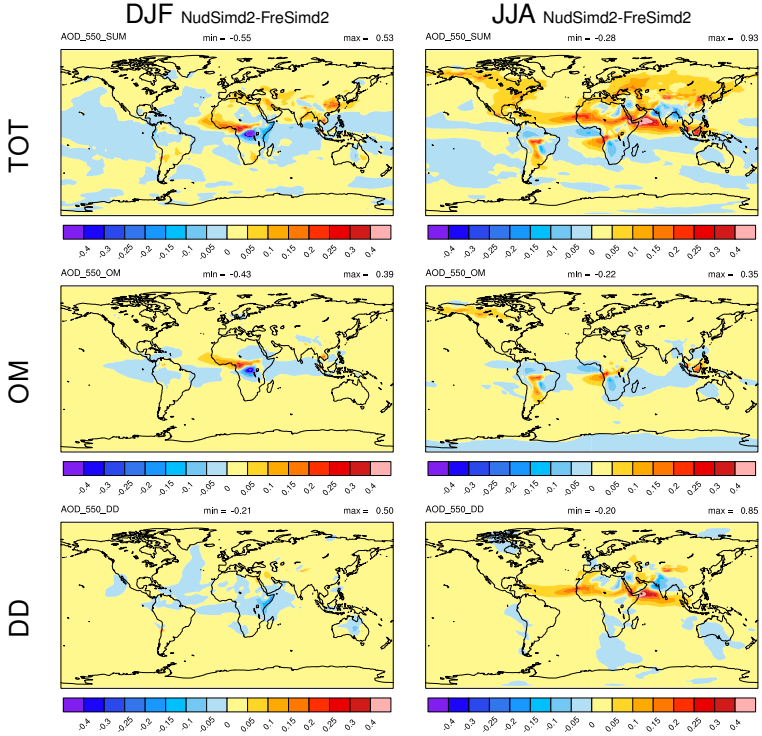

Fig. 3. Differences in AOD between the NudSimd2 and the FreSimd2 simulations, for DJF (left column) and JJA (right column), and for total AOD (first row), OM AOD (second row) and DD AOD (last row).

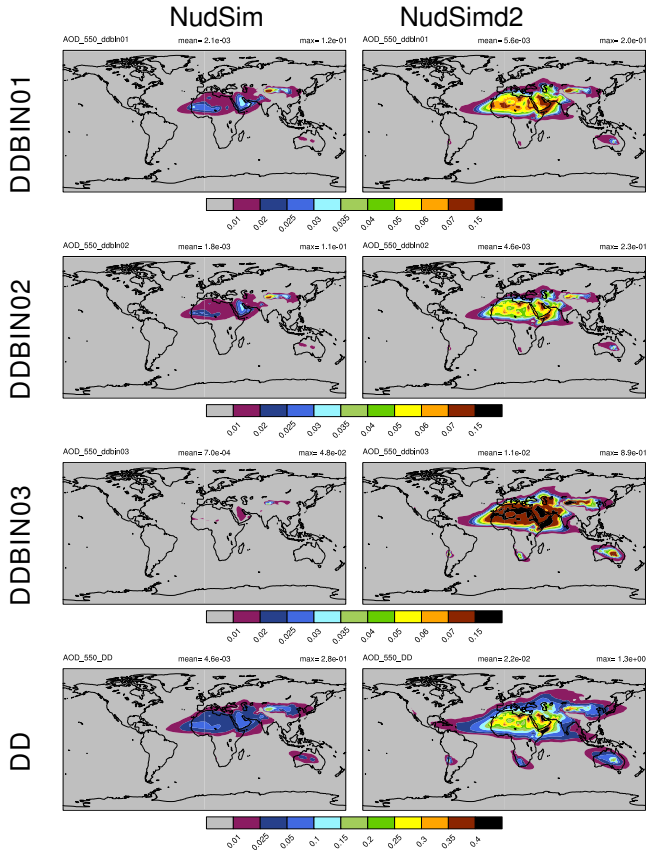

Fig. 4. Mean 2004 dust AOD for the NudSim (first column), and the NudSimd2 (second column) simulations, for the three dust bins, from the smallest (first row) to the largest (third row), and total DD AOD in fourth row.

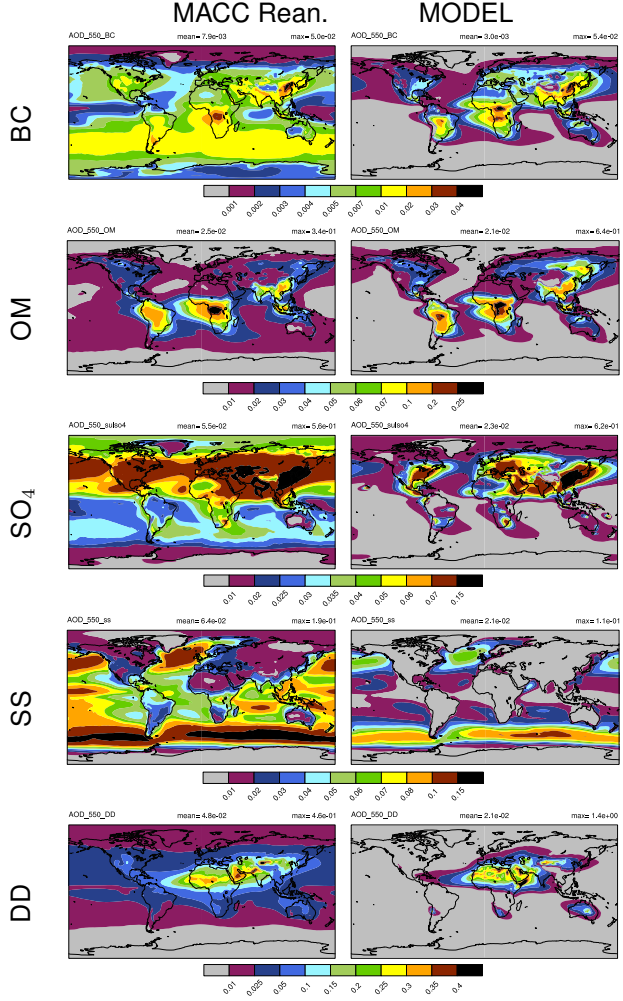

Fig. 5. Mean AOD (2003-2012) for the MACC Reanalysis (first column), and the NudSimd2_Trans simulation (second column), for BC, OM, sulfate, SS and DD.

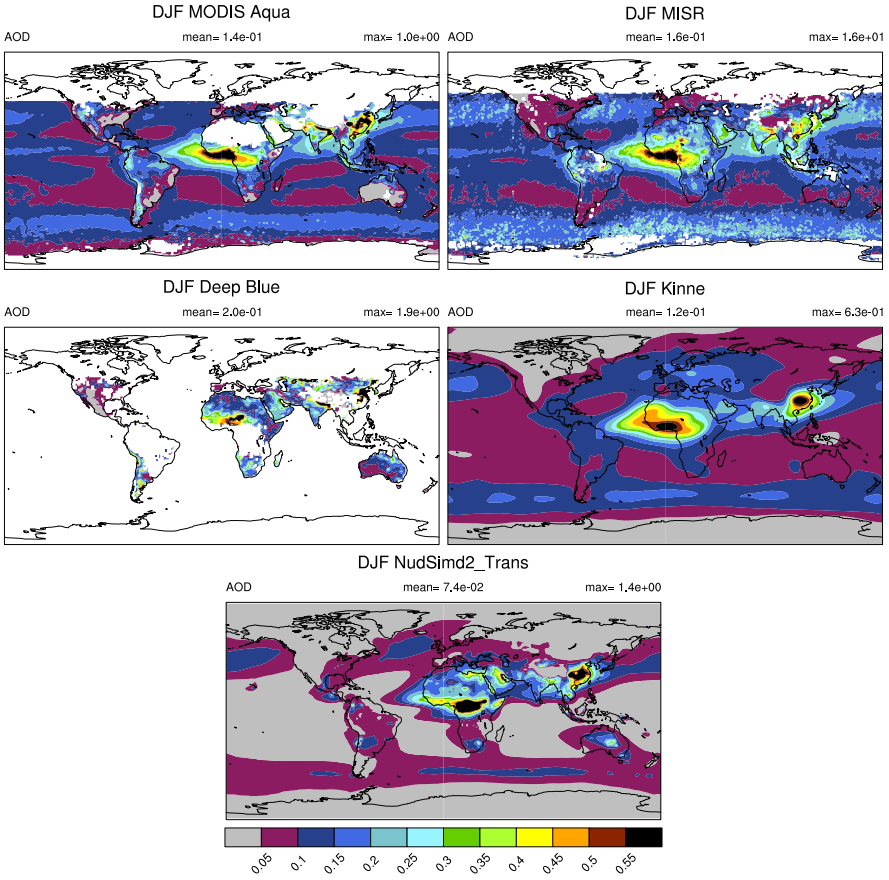

Fig. 6. Mean DJF 2003-2012 total AOD for the MODIS Aqua, MISR, MODIS Deep-Blue and *Kinne et al.* (2013) data sets (from the top in the direction of reading), and from the NudSimd2_Trans simulation (third row).

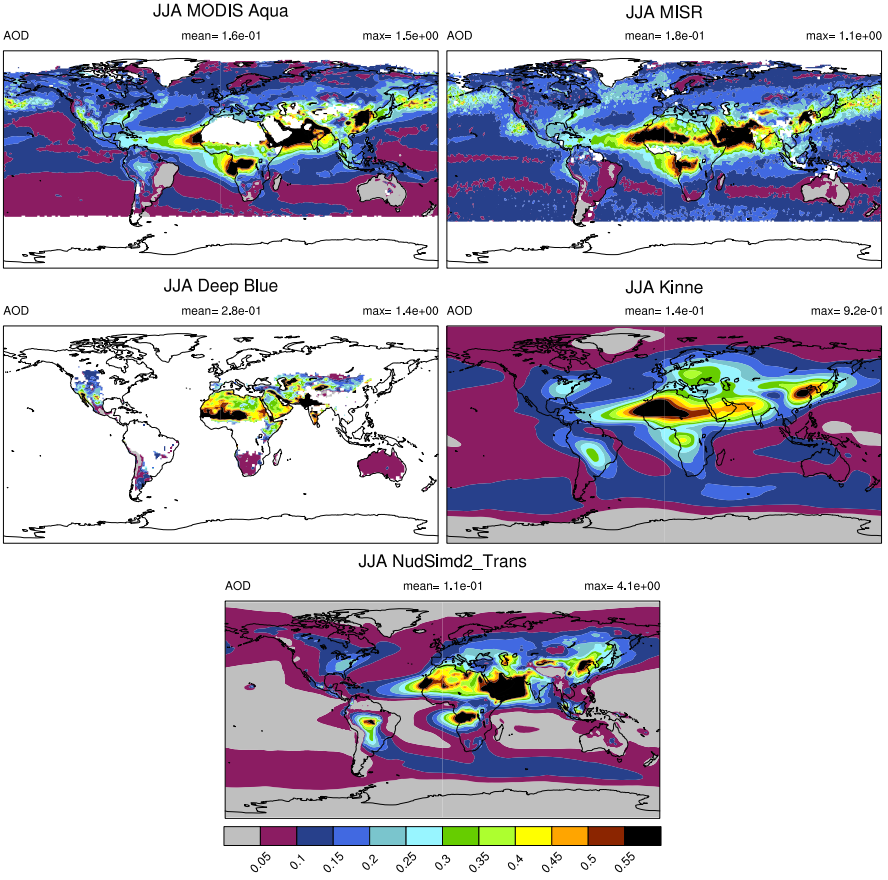

Fig. 7. Same as Figure 6, for JJA.

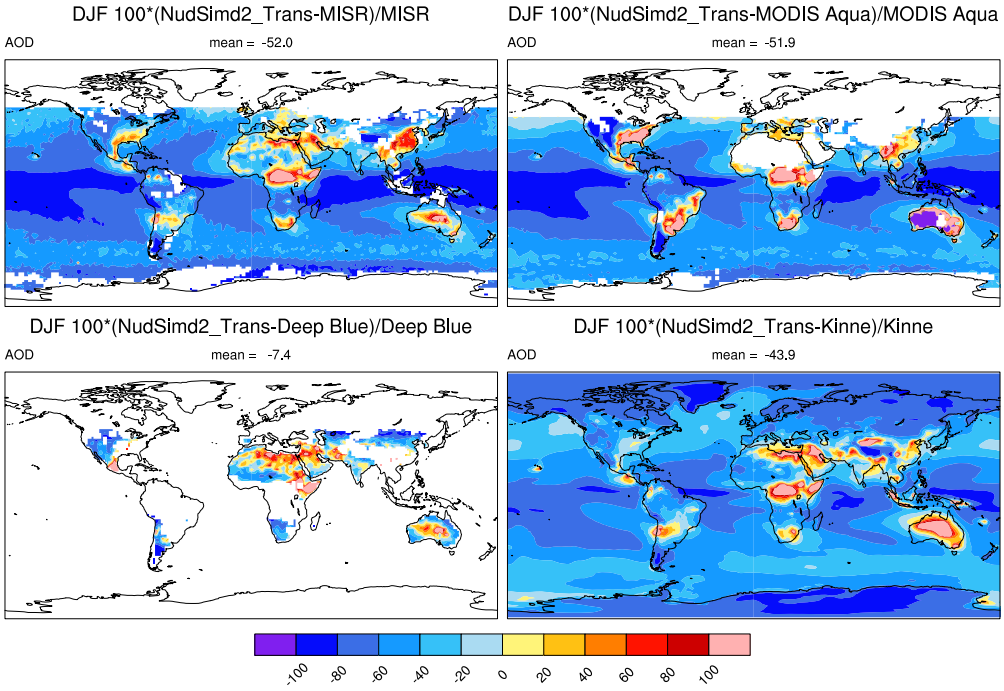

Fig. 8. DJF total AOD mean relative differences (2003-2012): 100(~~MISR-x)/MISR, with x=~~NudSimd2_T~~rans~~ rans-x)/x, with x=MISR first row/column, and x=Modis Aqua or x=MODIS Deep Blue or x=*Kinne et al.* (2013) in the direction of reading.

6

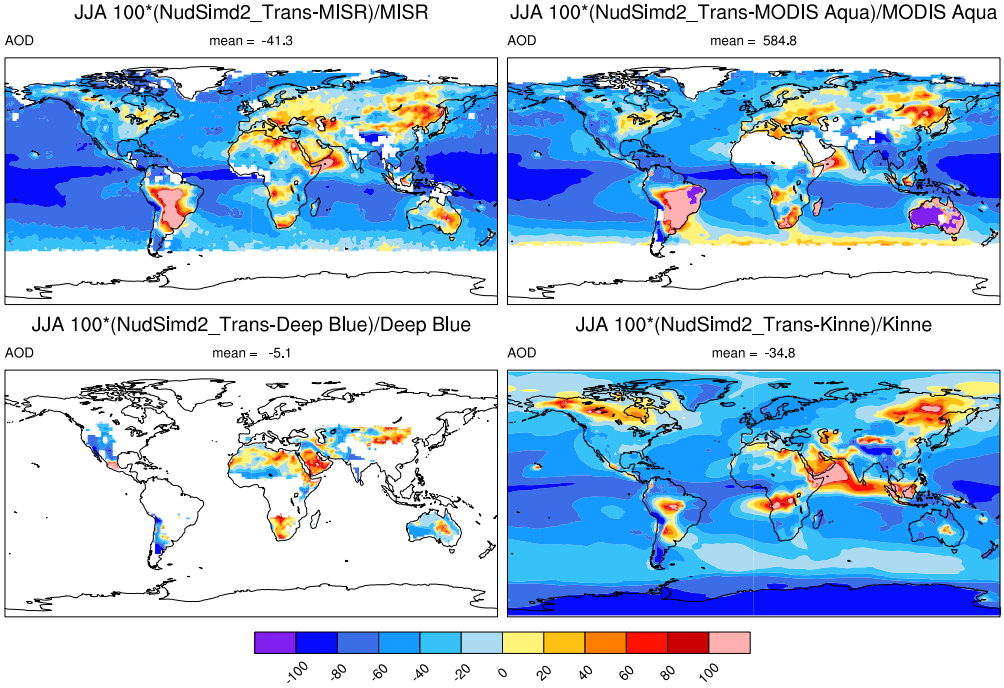

Fig. 9. Same as Figure 8, for JJA.

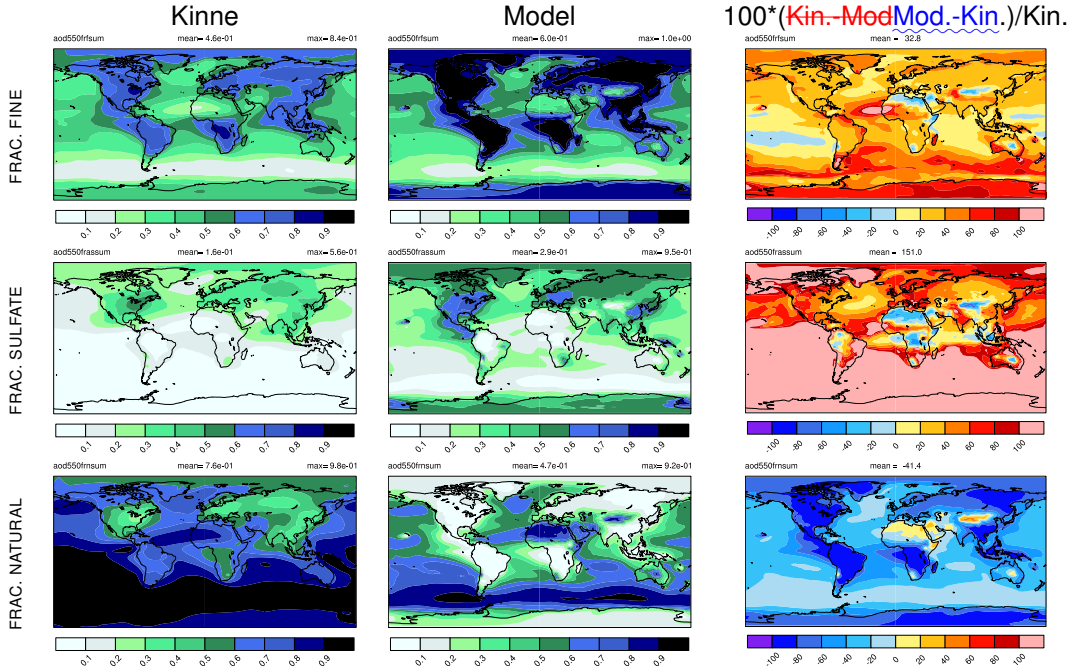

Fig. 10. Mean annual fractional AOD from the *Kinne et al.* (2013) climatology (first colunm), NudSimd2
SUBSCRIPTNBTrans simulation (1996-2005) (second column) and relative difference between the two data sets: fraction of ~~coarse~~ fine mode (first row), of ~~fine mode (second row), of~~ sulfate (~~third~~ sulfate row), and of natural aerosols (~~fourth~~ thirs row) (see text for details).

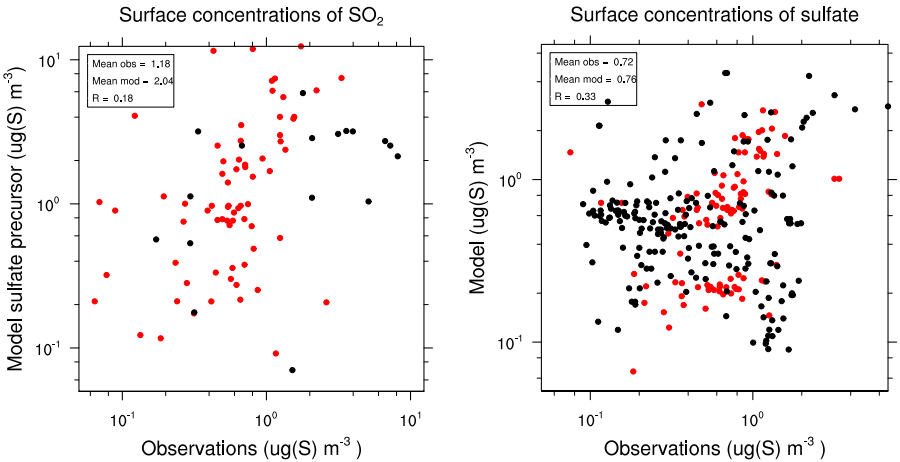

Fig. 11. Scatter plot of observations (EBAS database, see text) and corresponding NudSimd2 SUBSCRIPTNBTrans output: mean annual surface concentrations (2005) of (left) observed SO_2 (μg(S) m^{-3}) and modelled sulfate precursor, (right) sulfate (μg(S) m^{-3}). Red dots are mostly for European sites, while black dots are for US sites. Means of all observations, all model output and correlation coefficients (R) are shown.

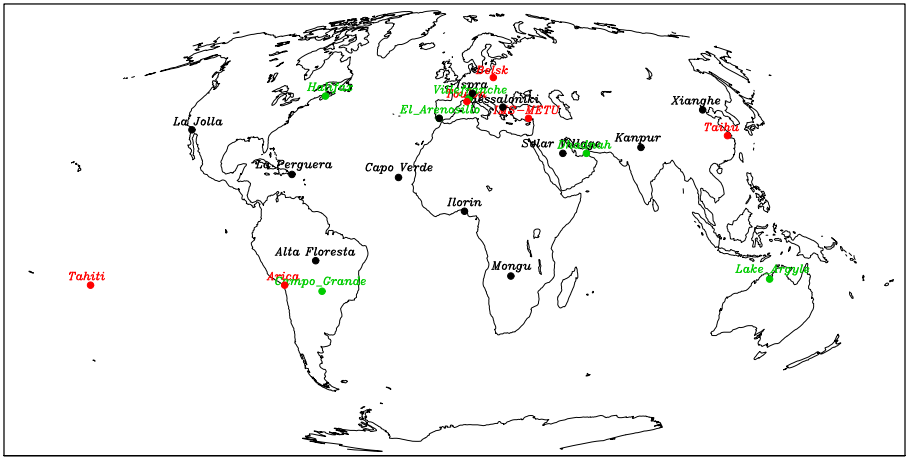

Fig. 12. Location of the AERONET stations presented in Figure 13, names in black, and in Figure 15, names in red for poor performance, in green for good performance.

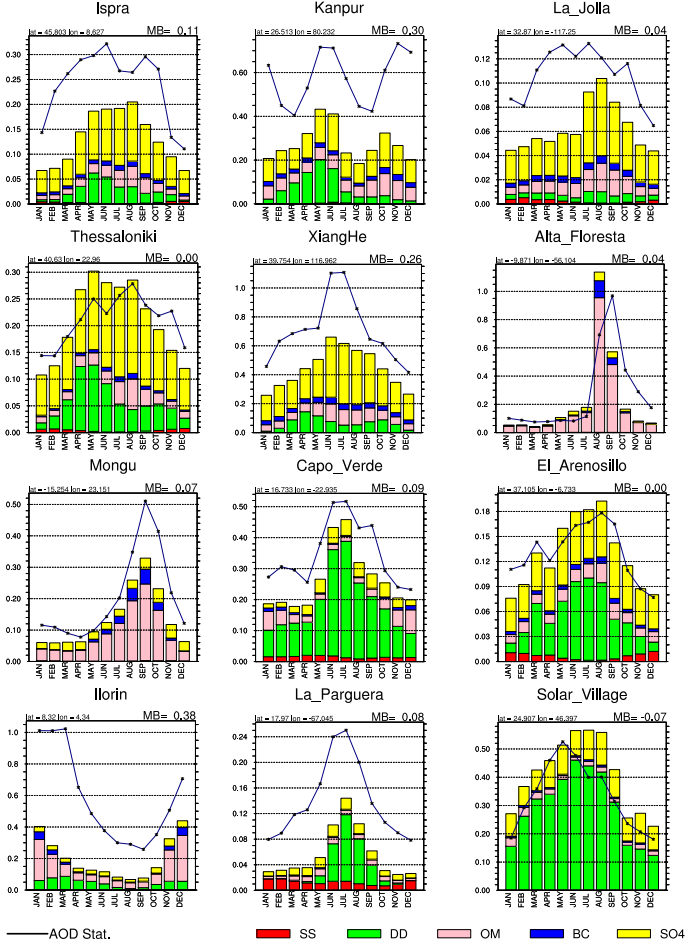

Fig. 13. Monthly climatology of AOD, computed from all years of available data, for the AERONET stations of *Cesnulyte et al.* (2014). Total observed AOD, and SO_4, BC, OM, DD and SS AODs from the NudSimd2 simulation are displayed.

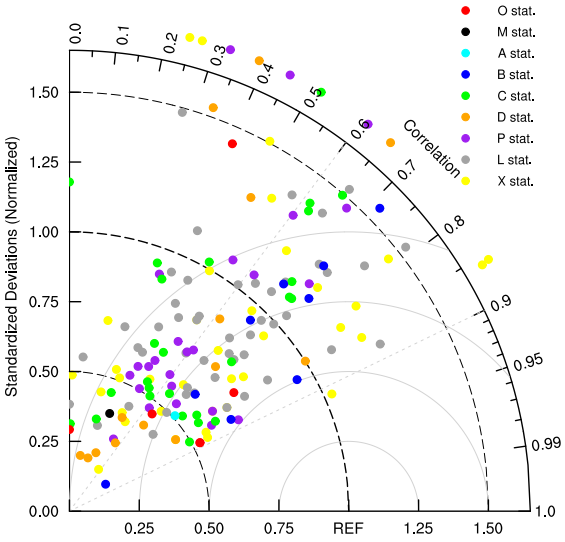

Fig. 14. Taylor diagram (*Taylor*, 2001) for the AOD monthly time series of 166 AERONET stations and ouputs from the NudSimd2_Trans simulation (see text for details). The qualification of the stations is that of *Kinne et al.* (2013) indicating the site dominant aerosol category (O, ocean; M, mountain; A, polar; B, biomass; C, coastal; D, dust; P, polluted, L, land), and X, no qualification.

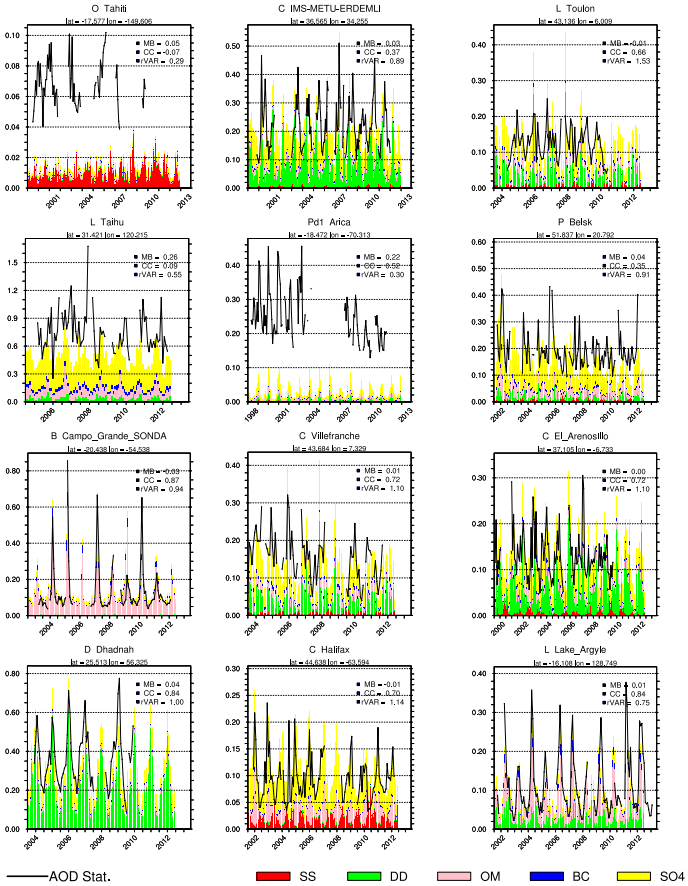

Fig. 15. Times series of monthly AODs, for a selection of poorly performing AERONET stations, first six images, and of ~~good~~ well performing AERONET stations, last six images, according to the Taylor diagram of Figure 14. The same AODs as in Figure 13 are shown. rVar: ratio of observed versus modelled ~~variances~~standard deviations, CC: correlation coefficient between observed and modelled time series, and MB: mean bias.

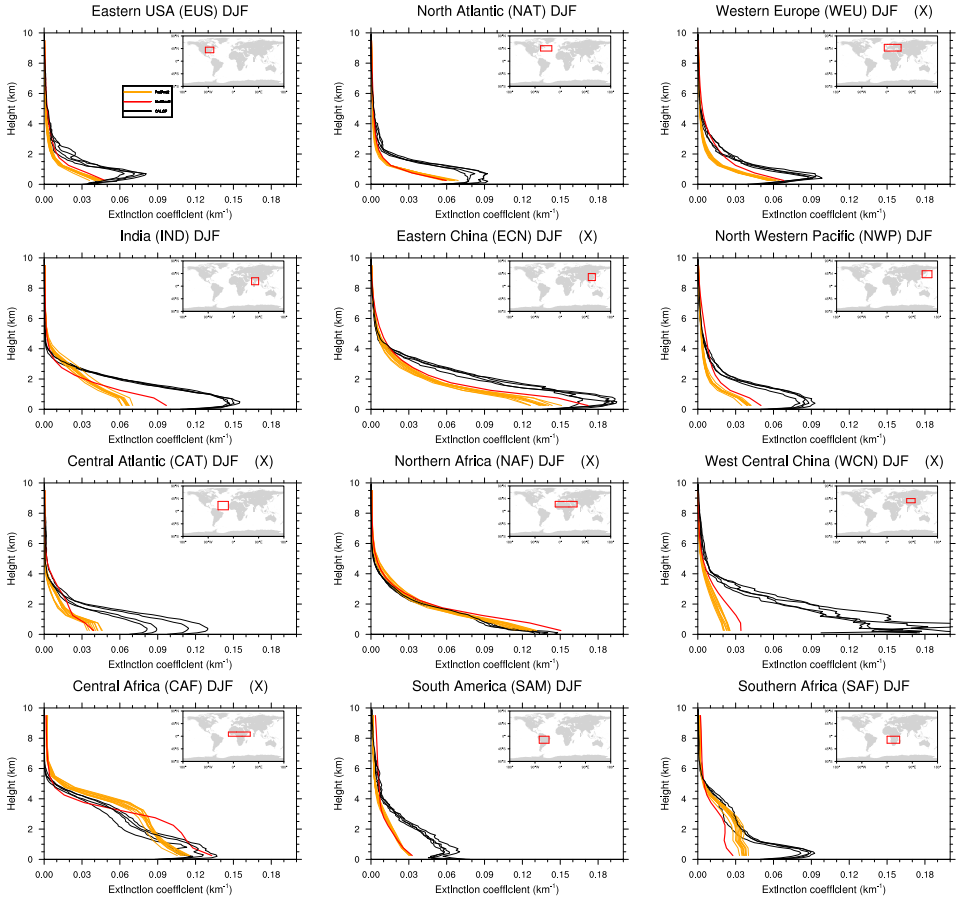

Fig. 16. Mean DJF vertical profiles of extinction coefficients (km^{-1}) for total aerosols, for the FreSimd2 simulation (orange lines) for 2004, repeated 10 times, the NudSimd2 simulation (red line), and for individual years of the CALIOP 3D product (black lines), over 12 regions of the globe, as in *Koffi et al.* (2012) (see in top right corners of individual figures).(X): regions also presented in Figure 18.

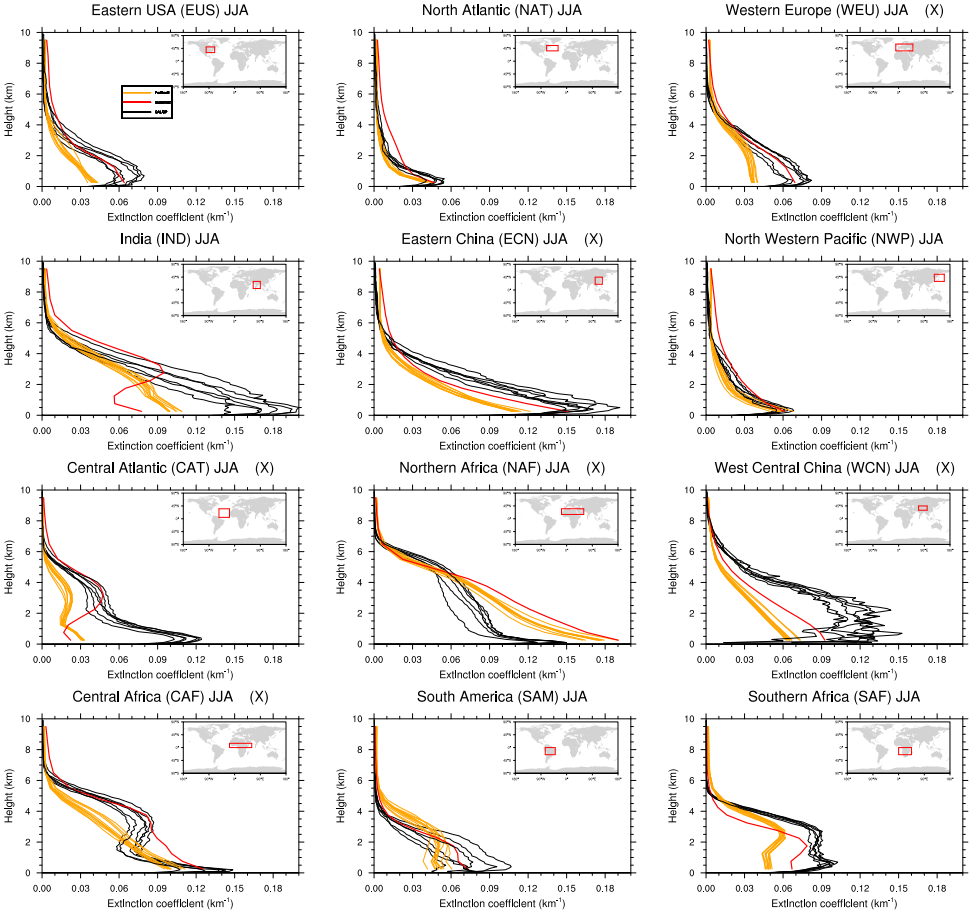

Fig. 17. Same as Figure16, for the JJA season.

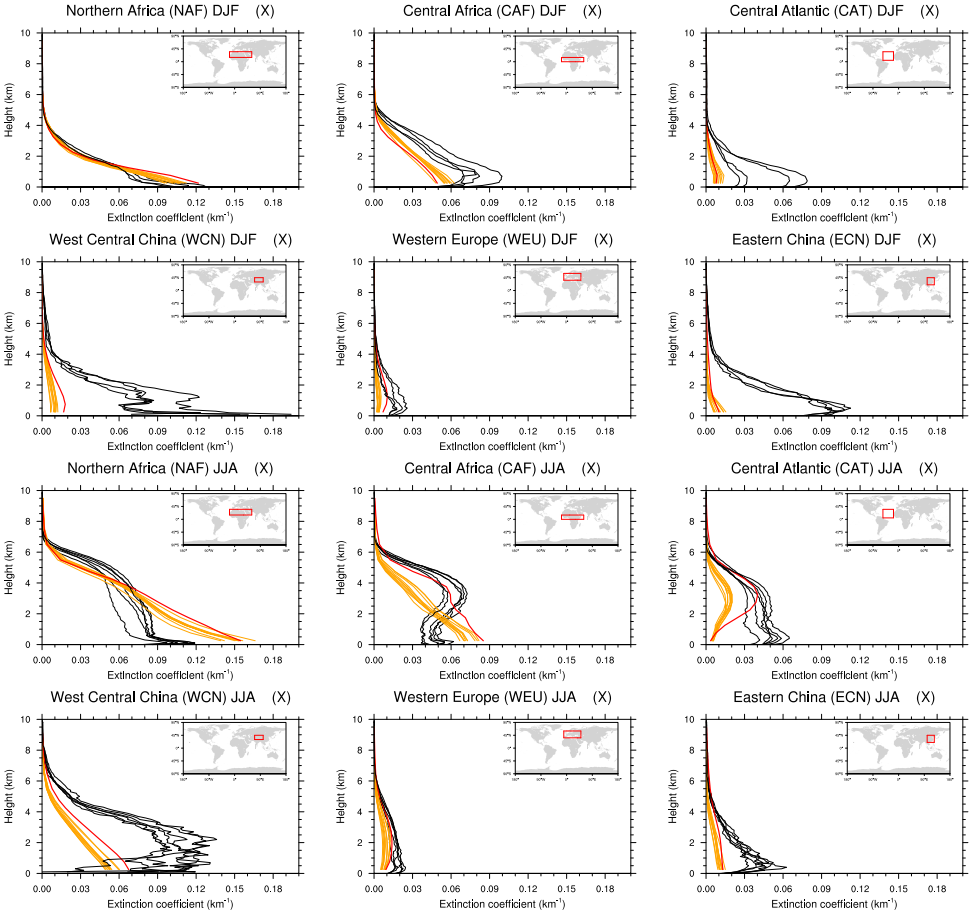

Fig. 18. Mean vertical profiles of extinction coefficients (km^{-1}) for the dust aerosol, for the FreSimd2 simulation (orange lines) for 2004, repeated 10 times, the NudSimd2 simulation (red line), and for individual years of the CALIOP 3D product (black line), over 6 regions of the globe with dust aerosols, as in *Koffi et al.* (2012), for DJF (rows 1 and 2) and JJA (rows 3 and 4).

Table 1. ~~Summary~~ Constants used in the aerosol scheme, in black values of ~~ARPEGE-Climat~~ the MACC reanalysis, in red values changed in our simulations~~performed~~: Eff. for scav.: efficiency for incloud scavenging - Eff. bc. r.: Efficiency for below-cloud scavenging by rain - Eff. bc. s.: Efficiency for below-cloud scavenging by snow - Reev. const.: reevaporation constant - Dry dep. vel.: dry deposition velocity (m s^{-1}), ocean, land, ice - Eff. for sedim.: efficiency for sedimendation (m s^{-1}) - Frac. emit.: fraction of emissions - Rate phob/phil: transformation rate from hydrophopic to hydrophilic (s^{-1}) - DD emis pot.: dust emission potential (kg s^2m^{-5}), bin radius (μm).

~~Name~~ Constant	~~Forcing~~ BCphil	~~Duration~~ BCphob	~~Dust scheme~~ OMphil	OMphob	DDbin01	DDbin02	DDbin03	SSbin01	SSbin02
Eff. for scav. D	0.8/0.1	~~(years)~~ 0.5/0	0.8/0.1	0.5/0		0.5			0.5/0.2
~~FreSim~~ Eff. bc. r. ~~NudSim~~ α_r ~~FreSimd2~~ height Eff. bc. r.	~~2004~~ ~~2004~~ ~~2004~~	~~10~~ 0.001 ~~10~~	~~(Ginoux et al., 2001)~~ ~~+~~ ~~(Marticorena and Bergametti, 1995)~~	0.001	~~(Ginoux et al., 2001)~~	0.001	0		
α_s ~~NudSimd2~~ height Reev. const.	~~2004~~	~~+~~	0.01 ~~(Marticorena and Bergametti, 1995)~~			0.01		~~(Kok, 2011)~~	0.01
RFRAER ~~FreSimd2~~ height Rain radius (m)			0.5			0.5		~~(Kok, 2011)~~	0.5
R SUBSCRIPTNB~~TR rans~~	~~1993-2012~~	0.001	~~20~~	0.001	~~(Marticorena and Bergametti, 1995)~~	0.001	0.001	-	
Snow radius (m) ~~NudSimd2~~ R SUBSCRIPTNB~~TS rans~~	~~1993-2012~~	0.001	~~(Kok, 2011)~~ ~~20~~	0.001	~~(Marticorena and Bergametti, 1995)~~	0.001	0.001	-	
Dry dep. vel. v_{docean} v_{dland} v_{dice}			~~(Kok, 2011)~~ 0.28E-02/0.1E-02 0.14E-02/0.1E-02 0.17E-02/0.1E-02		0.1E-02/0.15E-02 0.1E-02/0.15E-02 0.1E-02/0.15E-02	0.11E-01/0.07E-01 0.11E-01/0.07E-01 0.11E-01/0.07E-01	0.145E-01 0.145E-01 0.145E-01	0.1E-02/0.15E-02 0.1E-02/0.15E-02 0.1E-02/0.15E-02	0.11E-01/0.07E-01 0.11E-01/0.07E-01 0.11E-01/0.07E-01
Eff. for sedim. v_s			0.10E-02/0		0.6904E-04/0	0.1982E-03/0	0.1962E-02	0.24E-04/0	0.195E-02/0
Frac. emit. Rxpppp	0.8/0.2	0.2/0.8	0.5	0.5	-	-	-	-	-
Rate phob/phil RGRATE			7.1E-06		-	-	-	-	-
DD emis pot. S	-	-	-	-		2.E-11/1.E-11		-	-
Bin radius ZMMD	-	-	-	-	0.32/0.2	0.75/1.67	9.0/11.6	0.30	3.00

Table 2. ~~Totals emitted for static emissions~~Prescribed emission totals, including those used for the 2004 simulations, the 2003-2012 transient simulations, the MACC Reanalysis, and totals reported in the literature.[a] AeroCom mean $\pm\sigma$ (intermodel), *Textor et al.* (2006) Tab. 10. [b] mean $\pm\sigma$ (intermodel), *Huneeus et al.* (2012) Tab. 5. [c] *Tsigaridis et al.* (2014) mean and range from models. BC, OM and SOA in Tg yr^{-1}, ~~all~~ [d] *Boucher et al.* (2013) Tab 7.1 range, [e] *Dentener et al.* (2006) . All sulfur species in Tg(SO_2) yr^{-1}.

Sim./Litt.		Sim. 2004	Sim. 1993-2012 ~~Range~~	MACC Rean. 2004	~~Litterature~~ Literature
Species	Source				
BC	Tot. Sour.	10.3	~~9.1-11.8~~ 9.3-11.6	6.2	12±3[a], 15±14[b]
	Bio. Burn.	5.0	~~4.1~~4.0-6.5		
	Oth. Sour.	5.3	5.0-5.3		3.6-6.0[d]
OM	Tot. Sour.	117.3	~~105.4-139.4~~ 106.0-138.8	48.5	97±25[a], 119±111[b]
	Bio. Burn.	63.2	52.4-85.2		
	SOA	34.7	34.7		19 (13-121)[c]
	Oth. Sour.	19.4	18.3-19.5		9.5-23.0[d]
SO_2	~~Tot. Sour. 90.6 82.4-96.1101.5~~Bio. Burn.	3.3	2.4-4.4		
	Volcan.	~~12.0~~ 14.7	~~12.0~~ 14.7	0	29.2[e]
	Oth. Sour.	~~75.6~~ 105.9	~~68.0-79.7~~95.4-111.2		86.6-175.8[d]
DMS	Oceans.	~~27.9~~ 39.8	~~27.9~~ 39.8	0	20-80[d]
H_2S	Tot. Sour.	~~3.8~~ 5.3	~~3.4-4.0~~ 4.8-5.6	0	
SO_4	Tot. Sour.	~~6.0~~ 8.3	~~5.6-6.2~~ 7.7-8.6	0	
All SO_4 prec.	Tot. Sour.	~~128.0~~ 177.2	~~119.3-133.6~~ 166.0-182.9	101.5	119±26[a]

Table 3. ~~Upper part~~ Summary of ~~the Table: dust emissions (Tg yr^{-1}) over regions defined in *Huneeus et al.* (2011) , for the FreSim and FreSimd2~~ ARPEGE-Climat simulations ~~(mean over the 10 repeated 2004 years), the NudSim and NudSimd2 simulations (year 2004), the MACC Reanalysis (2003-2012 mean), and results from 15 AEROCOM models analysed in *Huneeus et al.* (2011) , median, min, and max values~~performed. ~~In blue, totals lower than the AEROCOM min, in red, totals higher than the AEROCOM max. Lower part of the Table: global sea-salt emissions (Pg yr^{-1}), with a range from *Grythe et al.* (2014) .~~

~~Tg yr^{-1}~~ Name ~~Region~~	~~FreSim/FreSimd2~~ Forcing	~~NudSim/NudSimd2~~Duration (years)	~~MACC Rean. AEROCOM Median~~ Dust emission scheme ~~(min–max)~~
FreSim	2004	10	(*Ginoux et al.*, 2001)
~~Global~~ NudSim	~~379 / 4236~~ 2004	~~258 / 3618~~ 1	~~313~~1586 ~~(487–3943)~~ (*Ginoux et al.*, 2001)
~~North Africa~~ FreSimd2	~~112/ 1298~~ 2004	~~66 / 1039~~ 10	~~88~~ 792 ~~(204–2888)~~ (*Marticorena and Bergametti*, 1995)
~~Middle East~~	~~66 / 659~~	~~51 / 579~~	~~37~~ 128 ~~(26–531)~~ (*Kok*, 2011)
~~Asia~~ NudSimd2	~~76 / 468~~ 2004	~~62 / 407~~ 1	~~75~~ 137 ~~(27–873)~~ (*Marticorena and Bergametti*, 1995)
~~South America~~	~~0 / 48~~	~~0 / 46~~	~~2~~ 10 ~~(0–186)~~ (*Kok*, 2011)
~~South Africa~~ FreSimd2			
SUBSCRIPTNBTrans	~~5 / 77~~ 1993-2012	~~3 / 51~~ 20	~~12~~ (*Marticorena and Bergametti*, 1995)
	~~12 (3–57)~~		(*Kok*, 2011)
~~Australia~~ NudSimd2			
SUBSCRIPTNBTrans	~~37 / 290~~ 1993-2012	20 ~~/ 175~~	~~47~~ 31 ~~(9–90)~~ (*Marticorena and Bergametti*, 1995)
~~North America~~	~~1 / 11~~	~~1 / 13~~	~~16~~ 2 ~~(2–286)~~ (*Kok*, 2011)

Table 4. Burden, residence time and ratios for various sinks from the FreSimd2 simulation (mean over the 10 repeated 2004 years), the NudSimd2 simulation (year 2004), the MACC Reanalysis (2003-2012 mean), and the AEROCOM models reported in *Textor et al.* (2006) (mean $\pm\ \sigma$, see table 10); [a] *Huneeus et al.* (2012) values. DD, SS, BC, OM and SO_4 aerosols are presented.

Param.	DD			
~~$Pg\ yr^{-1}$~~ Simul.	~~FreSim~~ FreSimd2	~~NudSim~~ NudSimd2	MACC Rean.	~~Range (*Grythe et al.*, 2014)~~ AE
				~~Global~~
				~~Constants used in the aerosol~~
burden (Tg)	~~DDbin01~~ 17.99	~~DDbin02~~ 23.30	~~DDbin03~~ 11.00	~~SSbin01~~ 19.2 ±40%
~~Eff. for scav.~~ residence time (days)	1.56	2.18	3.35	4.14 ±43%
dry dep/wet dep (%)	220.16	259.81	30.82	~~D~~ 148± 95%[a]
sed dep/~~0~~ dry dep (%)	13	13	~~0.5/0.2~~ 7	~~0 Eff. bc. r.~~ 46 ± 66%
conv. wet dep/wet dep (%)	12	42	28	44 ± 51%
~~cr~~ wet dep/total sink (%)	29	25	75	~~0~~ 33 ± 54%
~~Eff. bc. r.~~ Param.	BC			
Model	FreSimd2	NudSimd2	MACC Rean.	AEROCOM
~~Reev. const.~~				
burden (Tg)	0.13	0.18	0.43	~~RFRAER~~ 0.24 ±42%
~~Rain radius (m~~residence time (days)	4.68	6.28	2.44	7.12 ±33%
dry dep/wet dep (%)	14.84	17.60	12.62	~~RSUBSCRIPTNBR~~na
~~Snow radius (m~~sed dep/dry dep (%)	0	0	53	0 ± 251%
conv. wet dep/wet dep (%)	27	53	21	~~RSUBSCRIPTNBS~~46 ± 52%
~~Dry dep. vel.~~ wet dep/total sink (%)	87	85	84	79 ± 17%
~~V_{docean}~~		~~0.1E-02/0.15E-02~~	~~0.11E-01/0.7E-01~~	~~0.145E-01~~
~~V_{dland}~~ heightParam.	SO_4	~~0.1E-02/0.15E-02~~	~~0.11E-01/0.07E-01~~	~~0.145E-01~~
~~V_{dice}~~ Model	FreSimd2	~~0.1E-02/0.15E-02~~ NudSimd2	~~0.11E-01/0.07E-01~~ MACC Rean.	~~0.145E-01~~ AEROCOM
~~Eff. for sedim.~~				
burden (Tg)	0.92	1.30	3.35	~~v_s~~ 1.99± 25%
residence time (days)	~~0.24E-04/0~~ 2.25	~~0.195E-02/0~~ 3.18	~~0.180E-01~~ 2.27	~~0.05E-02/0~~ 4.12± 18%
~~Frac. phil~~dry dep/~~phob~~ wet dep (%)	15.46	18.70	7.95	na
sed dep/dry dep (%)	0	0	73	~~Rxpppp~~ 7± 202%
conv. wet dep/wet dep (%)	~~-~~27	~~-~~47	~~-~~22	~~-~~40 ± 54%
~~Rate phob~~wet dep/~~phil~~ total sink (%)	87	84	88	89± 8%

Table 5. Upper part of the Table: dust emissions (Tg yr^{-1}) over regions defined in *Huneeus et al.* (2011) , for the FreSim and FreSimd2 simulations , the NudSim and NudSimd2 simulations (year 2004), the MACC Reanalysis (2003-2012 mean), and results from 15 AEROCOM models analysed in *Huneeus et al.* (2011) , median, min, and max values. In blue, totals lower than the AEROCOM min, in red, totals higher than the AEROCOM max. Lower part of the Table: global sea salt emissions (Pg yr^{-1}), with a range from *Grythe et al.* (2014) .

Dust Tg yr^{-1} ~~RGRATE~~ Region	FreSim/FreSimd2	NudSim/NudSimd2	MACC Rean.	AEROCOM Median (min–max)
Global	330 / 3916	256 / 3597	313	1123 (514:4313)
~~DD emis pot.~~ North Africa	98 / 1226	66 / 1034	88	792 (204:2888)
Middle East	59 / 621	51 / 572	37	128 (26:531)
Asia	75 / 455	61 / 405	75	137 (27:873)
~~S~~ South America	0 / 47	0 / 48	2	10 (0:186)
South Africa	5 / 72	3 / 51	12	12 (3:57)
Australia	31 / 257	20 / 174	47	31 (9:90)
~~Bin radius~~ North America	1 / 11	1 / 13	16	2 (2:286)

Sea Salt Pg yr^{-1} ~~ZMMD~~ height	FreSim	NudSim	MACC Rean.	Range (*Grythe et al.*, 2014)
Global	~~0.32/0.2~~ 59.9 ~~3.00~~	~~0.75/1.67~~51.6 ~~10.00~~	~~9.0/11.6~~ 64.2	~~0.30~~ 1.8 to 605.0