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Geoscientific Model Development Discussions

Interactive comment on "Development and basic evaluation of a prognostic aerosol scheme in the CNRM Climate Model" *by* M. Michou et al.

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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 re-analysis, 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 me-*

teorological 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 emis-

sions 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

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