Review of “Description and evaluation of the tropospheric aerosol scheme in the Integrated Forecasting System (IFS-AER, cycle 47R1) of ECMWF” by Remy et al.

This manuscript describes the latest version (denoted cycle 47R1) of the aerosol scheme used in the IFS along with a description of the model updates implemented in this scheme since cycle 45R1 documented by Remy et al. 2019. A wide range of aerosol developments have been implemented including online coupling of the aerosol scheme to the chemistry scheme for the sulfur cycle, updates to the emission sources of sea salt and mineral dust as well as wet and dry removal processes. An evaluation is provided of the impact of the new model updates. Most of the updates are implemented in the operational configuration of cycle47r1 but not all. An evaluation of 1 year of a free-running simulation (without assimilation of aerosol information) is given.

Overall, this is an interesting and useful manuscript describing an operational aerosol configuration of the CAMS aerosol system. Aerosol forecast products produced by this system are widely used and so a detailed description and evaluation is warranted. It also describes differences between this system and the aerosol scheme used in the also widely used CAMS reanalysis. Understanding these differences as well as establishing the baseline performance of the latest model will be of use to many users. As this is largely a technical description and evaluation paper it is highly suitable for publication in GMD.

I have a number of recommendations which should be addressed before publication.

Major remarks:

- I would recommend that the authors consider making some fundamental changes to the manuscript layout. In my view it currently doesn’t flow well and this makes it quite hard to read and follow. The model updates sections (Section 3) includes some quite detailed evaluation of the specific updates documented but this is then followed by a further general evaluation in Section 7. Some of the latter still compares Cycle 45r1 with Cycle 47r1 so why not just put all the evaluation aspects together? Could the early sections, detailing model updates not just focus on the difference between the old and new model and then combine all the evaluation together under the Evaluation section.
would also recommend having a Results section which includes current Section 6 and Section 7 as subsections. To me the current layout is a bit disjointed and unclear.
- The model updates are not sufficiently motivated in my view and in many cases the model "improvements" or developments are not reflected in the skill scores. Can the authors motivate the changes in more detail, clearly outlining what the key drivers of the updates were. Are process-based improvements in one part of the model uncovering compensating biases elsewhere within the aerosol scheme? This also should be discussed. Has the original scheme been tuned in any way for instance to give the correct AOD values?
- The description of sulphur cycle in Section 2 would benefit from being described in more detail. The coupling of the aerosol and chemistry schemes is a significant step-change in the complexity of the IFS aerosol scheme and warrants full description. What sources of SO2 are represented and what chemical reactions (gas phase and aqueous phase processes) are represented in CB05, does it include a representation of DMS chemistry for instance. In order to understand the key drivers of the improved evaluation of surface SO2/so4 concentrations is it important to know what processes are represented or not and a reader should not have to go to another reference to get the information needed to understand the results presented here.
- Tables 2 and 3 are very confusing and I’m afraid I don’t understand them at all. This could be due in part to the Captions perhaps not being complete enough and incorrect labelling used (what config does “IFS” refer to?). I have read and reread the relevant sections but still do not understand why both cycle45r1 and cycle47r1 are included in both tables. I thought coupling to chemistry is included in the latter but not the former so why do we have 4 different simulations of the sulfur cycle? If it is to separate out the change in sulphur cycle from coupling to the CB05 alone from the other model upgrades included in cycle 47r1 then this is not at all clear in the text and both main text and table captions need to be improved. Could the 2 tables be merged perhaps to facilitate comparison? The whole section on the coupling and consistent and clear labelling really needs to be improved.
- It would also be nice given the significant impact of the deposition improvements in the cycle47r1 to discuss this generally overlooked part of aerosol modelling, with much focus often being placed on emissions and chemical production etc. The results here highlight the large and important role of more tightly constraining deposition processes in models more generally. It would be nice for the authors to place this work a bit more in the context of the current state of wider aerosol modelling/literature and not just the ECMWF models.
- While some areas of the evaluation quantifies the impact of the improvement on model skill scores in others it is more qualitative, and the authors use language such as “x is slightly better than y” or “the skill seems to improve”. This I feel detracts from the significance of their findings and from the benefits attributed to these model developments. An attempt should be made to be more quantitative in their language.
- In parts I find the text a little sloppy and so there are a lot of typographical corrections listed below. Taking a bit more care with the writing would aid both clarity and make the paper easier to follow in places. Figure labelling, I find to be incomplete and doesn’t include information on the temporal sampling of model or observed data in many places. For example, in Figure 1 I presume the model data is an annual mean but this isn’t clear from the caption. The captions need to be self-explanatory in their own right.
- There is insufficient description of the observations used, what time periods do they represent? What temporal sampling was used? This is very important in terms of interpreting the results to understand how representative the comparison is and are you comparing apples with apples! This likely could be better achieved via the restructure of the manuscript recommended above.

Minor comments:
P1 L14/15: concentration of sulphate ... is improved à concentrations of .... are improved

P1 L15: imbalances à biases

P2 L20: Cycling forecasts without data assimilation – presumably it is just the DA of aerosol information that is excluded and the data assimilation of meteorological variables is retained to constrain the simulated meteorology? Please make this clearer in the text.

P2 L22-26 : The Section labelling is all incorrect here, as the Introduction is Section 1, needs correcting.

P2 L24 : aerosol sources à primary aerosol sources

P3 L4: All of sea-salt à All of the sea-salt

P3 L6: The use of “etc.” isn’t satisfactory here. You should state clearly what model variables are divided by 4.3 or if all, say “all sea salt properties”

Note there is inconsistent use of “sea salt” and “sea-salt” throughout the manuscript. Please make consistent.

P3 Subsection title 2.1.1 : sulfur à Sulfur

P3 L27: Use of the CAMS_GLOB_ANT emissions versus MACCity, can you more accurately quantify the impact of the different emissions dataset on the subsequent simulated sulfur cycle. Emissions of SO2 are a big uncertainty in modelled S cycle budget generally and so could play a not insignificant role here? Also in Tables 2 and 3 I presume from the values these are annual mean fluxes but its not clear from the captions.

P3 L29 the chemical conversion rates are globally of the same order of magnitude – please see my comment above on how more detail on the simulation of the S cycle is warranted. Chemical conversion of what to what? Also if the chemical conversion rates are the key drivers of the increase in S lifetime in cycle 47r1 (presumably in Table 2) why is the lifetime of cycle 45r1 in Table 3 similar (~3 days).

Table 4: Caption is incomplete and I do not see any comparison or mention of the AeroCom Phase 3 comparison mentioned in the text.

P6 L5: Put reference in brackets.

Table 5: This table is quite informative but is barely mentioned in the text. Inclusion of the appropriate reference for each cycle would also be good to be included. M86, N12, G14, A16 are undefined. What is meant by Mass Fixer? This also should be explained in the text.

P9 L2: Monahan86 and Grythe14, why not just label them as M86 and G14 as you do in Table 5. Inconsistent labelling is confusing.

P9 L5: ofMonahan à of Monahan.

P9 L5-9 How globally representative are the ocean surface brightness retrievals?

P9 L24: Similarly to à Similar to

P9 L29: How was the evaluation carried out? Temporal frequency of observations and model (again not stated in caption of Figure 5 but it looks to be weekly?)? How
representative is this comparison? The sea salt contribution to the total AOD will be maximised in local wintertime, and so exhibits a clear seasonal cycle, has this been assessed? The MAN network could otherwise contain contributions from secondary sources of sulfate aerosol from DMS and other biogenic sources, looking at the seasonal cycle could help discriminate between the various sources.

P10 L2 AEROCE/SEAREX programme – include appropriate citation

P10 L8: is slightly improved -> can you be more quantitative

Table 6 and 7: It would be good to include the diameter ranges below the bin labels.

P14 L8 and Figure 6: is this an annual total?

P14 L14: The skill of the simulated dust seems to improve -> please be more quantitative

P14 L21: IFS-AER – which version? Please cf with reference to IFS-AER on L22 (same page)

P14 L21 producign à producing

Figure 7: It would be nice to see some uncertainty bounds or even a standard deviation of the observations (dust being highly variable in space and time) on these plots.

P18 L3: have been brought to à have been added?

P19 L14/15: why bold? Also I don't really understand how something can be implemented in a cycle but is not operational? Do the cycle numbers and revisions not refer to an operational configuration?

P20 L14: This sulphur à The sulphur . Also sulphur and sulfur are both used in the text.

P21 L2: CASTNET – include appropriate reference?

P23 L1: Where is the budget for Cycle 45r1 presented?

P23 L9/L18: “20” missing unit

P23 L29: Are the AeroCom values for the year 2017, if not this would easily explain differences in emissions?

P25 L6: deserts a à deserts

Figure 12: Highest and lowest values use the same colour which is a bit confusing.

P27 L14/15: what drives the simulated peaks in AOD? Are SO2 emissions from fire included?

P27 L29 : positiv à positive

P29 L6: significantly over à significantly improved

Figure 14 caption: regionallevel à regional level

P30 L8: probably don’t always hold true à isn’t true in all instances
Figure 16: While the evaluation of dust deposition is qualitative at best, it does look like the model deposits most of its dust too close to the African coastline with not enough extending westward over the Atlantic.

P30 L24: 2-3 ug/m3 than à 2-3 ug/m3 more than

P31 L2: biomassburning à biomass burning

P31 L4/5 : Again the improvements associated with the NEWDEP changes are very interesting. Are the new deposition changes offsetting the increase in AOD and PM2.5 associated with the biomass burning emission height change? Presumably if more particles are emitted higher up away from BL processes and sedimentation processes. Do you see a shift from dry to wet deposition between 45R1 and 47R1? Also the NEWDEP changes seem to impact some aerosol species (eg biomass burning) more than others (eg nitrate), can the authors offer a suggestion as to why this is the case?

P31 L7-10: Improvements in PM biases over China, can this be linked back to improved S cycle via coupling of aerosol scheme to CB05 chemistry?

P31 L25: obsreved à observed

P31 L27: in which the same nitrate scheme as IFS-AER has been adaptedà which has implemented an adapted version of the IFS-AER nitrate scheme

Figure 15: is only half the possible range of FGE covered by the colorbar used here? There seems to be a large underestimation of AOD over the Maritime Continent that persists through all model versions.

Figure 18 caption : Please note the different scale between the two panels – but the scales appear to be the same?

Figure 19: I can’t make out the observations in black circles.

Figure 20: what time period is used for the observations?

Figure 20 caption: should this be OM in PM2.5 and not surface ammonium concentration?