

Answer to reviews of GMD-2019-142 « Description and evaluation of the tropospheric aerosol scheme in the Integrated Forecasting System (IFS-AER, cycle 45R1) of ECMWF “

First, we wish to thank the reviewers for their comments, which raised many good points and helped improve the quality of the paper.

Anonymous Referee #1

General comment

The motivation behind the code revisions/additions that were chosen could be more clearly discussed in the introduction of the paper. As well, the authors should try to identify which modification was responsible for the improvement between cycles 40R2 and 45R1. These changes would make it easier for the reader to follow the logic of the paper.

This is a good point, thank you. The code revisions that are integrated into the operational version of IFS-AER must satisfy the two conditions (one qualitative, one quantitative) that they bring the model closer to “physical” reality (ie that more processes and/or species are represented), and that they improve the skill scores vs observations. A motivation behind several developments is to try to reduce the low bias of the AOD simulations against AERONET and MODIS. This was the main objective of the new SOA source and of the new sea-salt scheme. Debiasing the model vs MODIS allows the assimilation step to be more efficient in reducing error at analysis. A paragraph was added in the introduction to detail this, and in section 8.5 to highlight the developments that were responsible for most of the improvement between cycles 40R2 and 45R1.

Specific comments

P1L13: The code cycle 40R2 was not mentioned before in the abstract, and therefore it is unclear why the version 45R1 is compared to 40R2, and not 32R2.

Because it is a very old version, cycle 32R2 is now quite impossible to use with the ECMWF HPC. 40R2 was used because simulations were available as this cycle is the one used for CAMSiRA. Apart from the changes in the meteorological part of the IFS from cycle 32R2 to 40R2, there has been relatively few changes of IFS-AER from cycle 32R2 to 40R2. This is detailed in the first paragraph of section 8 “Evaluation”.

P5L1: When running coupled with IFS-CB05, SO2 no longer is a prognostic variable in IFS-AER. Why there are more prognostic variables in the coupled version?

This was taking into account nitrates (two variables) and ammonium; the sentence was modified to clarify this.

Why are certain code changes not included in operational cycle? It is clear to me that certain options would be computationally expensive (for example: P4L13-stratospheric chemistry; P4L29 - coupling with IFS-CB05) but I do not understand why others are non-operational (e.g.

P5L17 - height of emissions for biomass burning and SO₂ from volcanoes, especially considering statement on P6L20-24). Perhaps a general statement could be added for why certain code segments are operational or nonoperational.

Biomass burning injection heights are not used operationally in 45R1 (they are now in 46R1, but this is outside of the scope of this paper), in order to keep consistency between the aerosol and trace gases emissions: while the use of injection heights was shown to be mostly positive for aerosol simulations, it was not evaluated yet for simulations of trace gases (CO in particular).

P6L7: "remarkably consistent between the three datasets": I would say two datasets, since only the anthropogenic emission inventories are compared.

This is corrected, thank you.

P6L10-14: In cycle 45R1, do the scaling factors come from Kaiser et al. (2012) or Rémy et al. (2017)?

The scaling factors from Kaiser et al. (2012), ie a constant value of 3.4, is used. This has been clarified in the text.

P7L21 and Figure 1: Are the units correct? Figure caption reads kg m²s⁻¹, and also the there are higher values shown than 0.25.

The units of Figure 1 were wrong and have been corrected, thank you. Also, the cap of 0.25 µg/m²/s was not used in the data used to generate the emissions plot; this has also been corrected.

P10L3: It is mentioned that G14 is closer to the AOD observations, but this is not immediately clear to me from Figure 3, where for some months (e.g. May-June, Oct- Nov) it seems like M86 is closer to the AOD observations. Can you back this statement up with any quantitative measures (e.g overall bias?)

Figure 2 was modified to add the global bias against MODIS/Aqua AOD (collection 6.1), which shows clearly how a negative bias over most of oceans with M86 is reduced with G14. I agree that against AERONET the signal is less clear; the statement was modified.

P13L8: "Maps of probability of occurrence of observed AOD by MODIS dust AOD above different thresholds"- this phrase is not clear to me.

Indeed this was mashed up. This paragraph was rewritten.

P14L1: I do not understand how and why the anthropogenic SO₂ emissions are divided into these categories. Does "high"/"low" refer to altitude or the magnitude of emissions?

This is an option (added in the text). "high/low" refers to the altitude of emissions: this option (not commonly used) is meant to distinguish between SO₂ emissions that are released at the surface and those that are emitted higher up (ship emissions for example). The 80/20 ratio between "high" and "low" emissions is arbitrary and should be refined using emissions by sectors. The text has been modified to clarify this option.

P14L28: What is the rationale behind the change in SO₂ lifetimes in the later cycles?

Conversion of SO₂ was made faster (and SO₂ dry deposition was introduced) in order to shorten the lifetime of sulphate aerosol and also its burden, which was found to be much too high in CAMSiRA (using data assimilation). Thanks to these changes, sulphate aerosol is no more too dominant in CAMSRA.

P21L7: In Table 6 the deposition velocities of sulfate aerosol also differ between continents/oceans

The text was corrected; thank you for spotting this error.

P23L20-21: This pattern of ZH01 showing lower deposition velocities for fine particles and higher deposition velocities for coarse particles is not clear from Table 7. For example, fine mode sea salt is indeed slower in ZH01, but fine mode dust is faster.

This is correct: with the ZH01 scheme, dry deposition velocity as a function of particle diameter decreases from 0.01 to 1 micron and increases from 1 to 10 micron. The sentence was corrected and a sentence was added to explain this.

Why is the particle radius not considered in the wet deposition scheme? Aerosol radius can play a role in both in-cloud and below-cloud removal processes (e.g. Seinfeld and Pandis, 2006, Chapter 20). I am curious why a radius dependence was included for dry deposition but not for wet deposition.

In Seinfeld and Pandis 2006, the process that is most dependent on particle size is below cloud scavenging, which, in our simulations, is usually dominated by in-cloud scavenging. In contrast to dry deposition, wet deposition has not been updated in cycle 45R1 IFS-AER as compared what was described in Morcrette et al. (2009). This is beyond the scope of this manuscript, but below cloud wet deposition has indeed been updated in cycle 46R1, with a better representation of the impact of particle size.

P28L15: Is there a reference/derivation for the coefficients in the PM formulae? It can play an important role in the model evaluation, as mentioned in P39L3-5.

The coefficients in the PM formulae were computed using the assumed size distribution; this was added in the text.

Table 9: Are these listed size distribution parameters used also for the dry deposition/sedimentation schemes? For sea salt and dust the size bin limits were mentioned previously in the text, but for OM/BC/Sulfate/Nitrate this is the first mention of the assumed size distribution. Perhaps the assumed size distributions for these particulate species can also be mentioned in Section 2 or 3, when the different particle species are introduced.

The sedimentation velocities for super-coarse sea-salt aerosol and dust (the other species are not sedimented) have been computed with these size distribution parameters. For dry deposition with the ZH01 scheme, the Mass Median Diameter (MMD) was used.

Thanks for the useful suggestion: a new table was added in section 2.3 (main characteristics of IFS-AER) with these information. Table 9 was correspondingly reduced to the refractive indexes used.

Section 6 and Table 10 clarified some confusion that I experienced earlier in the paper, since they introduce the discussed code versions and highlight updates in the new CY45R1 code. I wonder whether it would be better to move this section and table earlier in the paper, since the reader would then be able to refer to this table when the code versions are mentioned.

Again, thanks for the suggestion; section 6 has been moved to a new section 3.

P31L10-11: I don't fully understand the logic why this would affect the wet/dry deposition ratio of BC compared to sulfate. It could also be related to the choice of deposition parametrizations and the size distributions of particle types.

Since this possible explanation is a pure guess (as we don't have access to the stratiform/convective distribution of precipitation in the simulations presented in Croft et al. 2014), this part has been removed. The underlying idea was that, in IFS-AER simulations at least, BC is relatively more abundant in regions where convective precipitation are either dominant or important, while sulphate is abundant in regions where stratiform precipitation are usually dominant. Yes, the size distribution certainly plays a role, particularly in GEOS-CHEM wet deposition.

Model evaluation metrics: The RMSE and bias metrics will be biased towards detecting deviations between the model and observations when AOD/PM are high, since AOD and surface PM vary by a few orders of magnitude. The model's skill at matching lower AOD/PM locations could be overlooked using RMSE/bias. Did you consider using normalized metrics for the model evaluation, for example mean fractional bias? I think normalization would anyways help with comparing the bias and RMSE between regions in Table 12, since the mean AOD/PM also varies between regions.

Absolutely; in routine evaluations we use Modified Normalized Mean Bias (MNMB) and Fractional Gross Error (FGE), in which lower AOD/PM locations have a much higher weight than bias and RMSE. There was a lot of hesitation between the two options (bias/RMSE and MNMB/FGE); the bias/RMSE plots have now all been replaced with MNMB/FGE plots, as well as the data in Table 14.

P40L1-5: What is the reason for the improvement in model skill between CY45R1 and CY40R2? Can you point to a change in Table 10 that is responsible?

A paragraph was added in this subsection: The development that had the most impact on skill scores against AERONET is the implementation of a new SOA source in cycle 43R1, which led to a significant improvement of both bias and RMSE. Using the ZH01 dry deposition scheme also improved scores. The new G14 sea-salt aerosol scheme had little impact on AOD scores against AERONET or on PM skill scores, but improved notably the bias and RMSE versus MODIS AOD

P40L6-8: Does the seasonal cycle of the North American bias in PM_{2.5} provide evidence for the hypothesis that the SOA emissions are responsible? Since this bias is larger for May-Nov?

This is a problem the whole year long. The evidence comes from looking at PM diurnal cycle (not shown), which shows very high values at night when SOA is emitted and not vertically diffused, and from comparisons with simulations without the new SOA source.

P43L1: This statement could be made more accurate, to say that the coupling improves the error of the forecasts, especially over Europe. The bias seems to be worse everywhere except Europe and Africa in the coupled version.

This is a good suggestion, thank you.

P43L8-9: The final two concluding sentences could be made stronger, since these ideas were not discussed very much in the model evaluation. Perhaps these future upgrades could be linked to deficiencies that were found in the results?

Yes; actually since cycle 46R1 is now out, the new upgrades are actually known and consist in a upgrades of the dust emission scheme, of wet deposition and the activation of the use of biomass burning injection height. IFS-AER now runs coupled with the chemistry in the operational context. The conclusion was modified to reflect this.

Technical corrections

P2L10: space missing before (NCEP)

P4L18: "the" missing before prognostic variable

P4L26: Two components are considered of organic matter and black carbon, hydrophilic and hydrophobic fractions, with : : :

P7L29: fluxes of sensible

P9L19: two schemes

P9L20-21: is also shown to compare

Figure 5: Caption should specify that super-coarse dust is shown.

P14L23: CY is used before the cycle name for the first time. This should be made consistent throughout the paper.

P15L1: a diurnal cycle and a simple dependency on temperature

P15L23: "important decrease"- significant decrease -> but also could specify where - in North America and Europe SO2 emissions have decreased strongly, in East Asia they have increased since 2000 (e.g. Hoesly et al., 2018)

This sentence just means to compare sulphate burden (and AOD) for the same period between the CAMS interim reanalysis and the CAMS reanalysis: for the latter sulphate burden is much lower, which corrected a clear positive bias in CAMSiRA. This was detailed in an additional paragraph in the same section.

P16L11: the equation numbering restarts from 1 here. Reactions should be numbered R1, R2, etc.

P17L24: fraction of airborne calcite in coarse

P23L10: the first comma should be a period?

P23L19: provides

P23L30: ZH01, notably

P23L31: will be addressed

P25Eq38: $j-1$ should be in superscript in numerator

Table 11 caption: in parentheses, not in hypens

P31L9: with

P32L5: combines

Figure 10 caption: should read (top) / (bottom) not (left) / (right)

P33L5: acronym RMSE should be defined the first time it is mentioned

Figure 12: titles of plot refer to global sites, but the caption and text refer to North American sites

P38L4: by a higher contribution from nitrate and ammonium

All corrected, many thanks for the careful checking!

Anonymous Referee #2

General comment:

“In the introduction and perspective, I am surprised that the added value of implementing coupled aerosol model to improve the weather forecasts is not mentioned in the context of ECMWF activities. This may be due to the low priority given to aerosol-cloud interactions (P3L29), but more explanation should be given in the text on this choice to understand if the impact is thought to be marginal, or if the work is planned in the future.”

Two paragraphs were added on this subject in the introduction, mentioning in particular how IFS-AER was used in the context of sub-seasonal forecasts at ECMWF (Benedetti et al. 2018).

“My only general concern regards the lack of context about state of the art in global aerosol modelling, as it would be useful to explain in more details where the AER-IFS lies in comparison with other major similar models.”

A paragraph was added in the introduction on the subject. Also, the budgets of IFS-AER are compared, as much as possible, with those of GEOS-CHEM as reported in Croft et al. (2014).

Specific comments:

P1L17: these mortality numbers are only for ambient air pollution (excluding indoor air).

Absolutely. This has been precised in this sentence.

P2L14: a link to the ICAP-MME service should be provided as a web search did not allow me to reach those alternative global aerosol products.

A link to the ICAP-MME download ftp have been added

P3L29: why mentioning only the impact of aerosols on surface temperature as photolysis rates are also sensitive as acknowledged P5L5.

This paragraph refers only to the impact of using interactive aerosols in the radiation scheme, while the first paragraph of page 5 describes the coupling of the aerosol and chemistry schemes. Photolysis rates depend on temperature and thus can be impacted by the use of interactive aerosols in the radiation; a sentence was added in the same paragraph.

P4L13: what would be the implication of including stratospheric chemistry for AER-IFS given the importance of heterogeneous processes in the stratosphere?

The computational cost would be significant, and IFS-AER deals only with tropospheric aerosols so the potential benefits of including stratospheric chemistry appear to be not so great. However, work is ongoing (outside of the scope of this manuscript) to build a full tropospheric-stratospheric aerosol chemistry system using a modal aerosol model, IFS-GLOMAP.

P4L30: is SO₂ or rather sulphate aerosol production rate (as stated P5L9) provided by IFS to AER-IFS when run in coupled mode?

When running in coupled mode, the sulphate production rate is provided by the chemical model integrated in the IFS, CB05.

P5L1-8: why not mentioning here the new CAMS-GLO-AP emissions that should be included in the global forecast soon (if not already)?

Thank you for the suggestion, indeed the new CAMS_GLOB emissions are used operationally since July 2019. This was added

P6L8-19: the scaling factor for biomass burning emissions seems very ad-hoc. How is it handled for instance by regional models? What are the perspectives to represent explicitly condensation to cope with this issue?

Indeed, but as explained in the manuscript this is a common problem when using fire emission datasets in global model. The exact cause of the discrepancy between simulated and observed AOD when not using scaling factors for biomass burning emissions is not known yet. There are plans to better represent the ageing of biomass burning aerosols but it is far from ensures that this will bring a solution to this problem.

P5L19: suggest adding SOA in the title of the section. Given the importance of SOA acknowledged P7L16. More information on the state of the art on SOA modelling in global aerosol models should be provided.

Since SOA is not yet a species of its own (in CY45R1 it is treated as a component of the OM species), we prefer not to include SOA in the title so as not to confuse readers. A paragraph on SOA modelling in global models have been included.

P9L26: typo: M86 instead of G86

Corrected, thank you.

P10L3: quantitative scores (RMSE, correlation and bias) should also be provided as it seems that the new formulation sometimes leads to high overestimations of low AOD levels.

Thanks for the suggestion; global RMSE and Bias vs MODIS/Aqua have been included; the new sea-salt scheme generally improves this skill scores: sea-salt AOD was significantly underestimated with the M86 scheme.

P13L5: the figure legend says total emission are presented whereas only super-coarse are mentioned in the text.

Thanks; the text has been corrected.

P21L3: where was it shown that differences where small? Is it only true locally or just on average?

It is true both locally and on average; experiments were ran (not shown) with the two approaches for dry deposition: adding it to surface fluxes, or passed through the turbulent

diffusion scheme that gave these results. Since the sentence is confusing it has been reworded.

P24: use the same unit in the table & figure

Corrected, thank you.

P26L13: I find it confusing to present here ageing, while hygroscopic growth is presented in 3.6

The content of the ageing section was moved in section 3.6

P28L7: what is referred to as CAMS mineral dust model? AER-IFS?

Yes, this refers to the dust scheme of IFS-AER; this has been corrected for more clarity.

P28L15-24: the notations are not defined (SS1, SS2, etc.)

A definition was added for the notations of the PM formulae.

P29 Table 9: what is referred to as CAMS model? AER-IFS?

Yes, this refers to IFS-AER; this has been corrected for more clarity.

P29L11: confirm if the upgrade to 137 levels has been completed by now

It has been a part of the upgrade to cycle 46R1 on 9th of July 2019. This has been added in the text.

P30 Table 10: what is referred to as IFS versions? AER-IFS?

Yes; the caption of table 10 was reworded for more clarity

P31L9: typo "wityh"

Corrected, thank you.

P32L24: typo: "Unites"

Corrected, thank you.

P37L10: the sentence is inconsistent. Fire and dusts are mixed, and my understanding is that Europe was discussed in that paragraph.

This paragraph and the previous one were rewritten as they were indeed confusing.

P43L1: whereas coupling improves AOD over Europe, it is not the case for PM and should be reminded in the conclusion.

This was mentioned in the conclusion.

Anonymous Referee #3

General comments:

I have two substantive comments on the paper. The first is that many of the figures are not really publication quality in that they are titled/labeled with a lot of jargon that means something to the writers but not to anyone else. I noted those occasions below and suggest redoing those figures to remove the title text (or else writing it in a way that is sensible to the general reader).

Thanks for the suggestion; these titles and labels have been removed from the plots.

My second substantive comment is that there is a great deal of text written that does not pertain specifically to the IFS-AER operational configuration. I understand this from the point of view of explaining the possible permutations of the system, but it is hard in the end to reconstruct in my head the actual configuration used in the operational system, which seems to be the point. Table 10 is not sufficiently comprehensive in this sense. I suggest to please add a table, and maybe do it up front, that writes down the details of the IFS-AER operational configuration as a reference (chemistry or lack of, BB emissions altitude, and so forth, sea salt emission scheme, etc.). Otherwise I was getting lost in the details.

This is a good point, and was the subject of discussion between the authors as to exact scope of the paper (operational 45R1 only, or to present also options not used operationally). In order to clarify the operational configuration, the “operational configuration section” has been moved into a new section 3, before the detailed description of the IFS-AER components. More details have been included in this new section in order to complement table 3 (ex table 10).

Specific comments :

Page 3, line 28: implication of using online ARI

The impact is generally small on aerosol burden itself; locally and for extreme dust events, the impact on temperature and winds can create a feedback on aerosols themselves. This paragraph has been completed to mention this.

Page 4, line 1: A point of clarification: is IFS “the” ECMWF forecasting system? And if so, isn’t it run at a considerably higher than global 40 km resolution? So is IFS-AER run at the same resolution as the operational NWP system or not?

Yes, IFS is the ECMWF forecasting system; in the CAMS project IFS with aerosol (IFS-AER) and chemistry (IFS-CB05) extensions is used. A lower resolution is used as compared to the

operational high-res NWP because of the computational cost: CB05 adds 56 prognostic variables and AER 12 or 14 depending on the configuration. In an operational context there are tight constraints on time for the model to run, which effectively limits the horizontal and vertical resolution used with IFS-AER. A sentence was added to this paragraph to explain this.

Page 4, line 32: I don't follow the counting of species, please clarify. 12 species when running standalone I guess excludes the "optional" nitrate. When running coupled do you move the SO₂ to the chemistry but still count the sulfate species? Are you now counting the nitrate? Did you not note an ammonium tracer, implied in section 2.4? Assuming all of that my count is 3 dust + 3 sea salt + 4 carbon + 1 sulfate + 2 nitrate + 1 ammonium = 14. Please clarify. And what is the condition for running the optional nitrate? Here and elsewhere in the paper it is useful to state explicitly what is in IFS-AER cycle 45R1 which is the main point of the paper. The operational configuration would seem to be IFS-AER standalone.

Yes, when running coupled to the chemistry, SO₂ is not anymore a species used in IFS-AER, but sulfate aerosols are still simulated (using oxidation rates provided by IFS-CB05). The total does come to 12 in standalone mode, and 14 when running coupled. Nitrates need the gaseous precursors provided by IFS-CB05, so can be used only when running in coupled configuration. In the new section 3 "operational configuration", it is stated that the operational configuration for cycle 45R1 is standalone, and coupled for operational cycle 46R1.

Page 6, line 1: Any citation for the partitioning of hydrophobic and hydrophilic carbon?

Yes, a reference was added.

Page 6, line 18: I think the reason given here for the scaling factors is not credible. What you are really doing here is using a scaling factor to tune the emissions to give best agreement with simulated AOD, and that in fact implicates all of the model physics (especially sink processes) as well as whatever optical parameters are used to go from dry aerosol mass to hydrated extinction efficiency. So what you are doing here is finding the model "perceived emissions" needed to converge on observed AOD. If it were simply a gas to particle factor it is not clear why this would be model dependent since you are not doing that chemistry. I suggest rewriting this sentence in the spirit of the "perceived" emissions as I describe above.

This is exactly that, as outline in Kaiser et al. (2012): tuning/scaling the emissions so as to match MODIS AOD. The exact reason why this is needed is not yet clear, and a variety of causes can participate to this: purely model factors such as optical parameters and dry/hydrated extinction efficiency as you mention, model sinks/transport etc. The different resolution between MODIS native FRP, GFAS gridded products and the model resolution certainly plays a role. The fact is that scaling factors are applied in several global atmospheric composition models. In any case, the concept of "perceived emission" certainly fits here. This paragraph was modified accordingly.

Page 7, line 21: Figure 1 does not show anything about seasonality.

Yes, this was an error, now corrected.

Page 8: there is something missing (like a "x") before the 10^{-6} in equation 3.

Added, thank you.

Page 9, line 19: do you mean "two schemes"?

Corrected, thank you.

Page 9, line 20: "mode" instead of "model"

This part of the sentence was deleted as it was not really useful.

Page 9, line 21: two schemes? Why do you write "three"?

This is indeed two schemes, corrected.

Section 3.2: The presentation suggests there are no global tuning factors. Experience tells me this is usually not the case, and that models at least need a scaling factor as a function of spatial resolution. I'm also skeptical of the utility of comparing the predicted emissions to the G14 estimate. I won't dismiss G14's estimate of emissions, I haven't read the paper, but generally my sense is that non-inventory emissions are not usually observationally constrained sufficiently and so (like for biomass burning and dust later) some tuning is done to get emissions that in your model improve agreement with AOD, which is observed.

No global tuning factors were used, but the G14 formula is itself the result of a fit to observations (same for the SST dependency), which might explain why no tuning was needed there.

Figure 3: The title is best cropped from this figure as it uses jargon not discussed in the paper ("gzis", etc.)

Absolutely; all such titles have been cropped.

Page 12, line 8: Please explain better the dynamic nature of the lifting threshold velocity. What does "emission capacity" mean? Can you please provide an equation?

This paragraph has been expanded and a new equation inserted to explain the simple way lifting threshold velocity is estimated in IFS-AER.

page 12, line 12: I think you mean equations 10/11 and not 13.

Corrected, thank you.

*Page 21, line 3: Please explain the statement about the connection to the vertical turbulent flux scheme. Are emissions just added to the model grid boxes (flux*time = mass changed to cell)? What does it mean surface fluxes are unchanged here?*

This part has been clarified: the vertical diffusion scheme updates the surface concentration using the deposition velocity. Alternatively, before cycle 45R1 the deposition velocity can be used to update the surface fluxes. The impact of choosing either approach is very small.

Page 23, line 31: How have these been addressed?

Since this is part of cycle 46R1 it is a bit out of scope for this article. The median mass diameter of some aerosol species used in the ZH01 was too high and was reduced, which greatly reduced dry deposition of these species over mountainous areas.

Page 25, line 15: Do you mean the settling velocity is horizontal invariant? Constant in space is too broad, because you do have vertical variability (don't you?)

Not at all, the same settling velocity is used everywhere and at every model level. This applies only to super coarse dust and sea-salt, which don't usually get transported too high in the free troposphere, but still, updating this could be done in next cycles.

Page 26, line 1: What is the origin of the numbers in Table 8 for D_i ?

They come from Reddy et al. (2005). This has been specified.

Page 26, line 10: What are the "R" values in equation 42?

It is the assumed radius of rain drops/snow flakes, set to 1mm. This has been inserted in the text.

Page 27, line 24: Please define OPAC and provide citation. This is first time you use the term OPAC.

OPAC has been defined, with the appropriate citation.

Page 31, Table 11: You mean parentheses and not hyphen at the end of the caption.

Yes, this has been corrected, thank you.

Page 23, figure 10: you mean top and bottom in the caption

Corrected, thank you.

Page 32, Section 8: Is aerosol data assimilation invoked in this analysis?

No aerosol data assimilation was used in the simulations that have been evaluated. This has been explicated at the head of the section.

Page 35, Figure 11: suggest again removing the title from the figure as it is inside jargon

The title has been removed

Page 36, figure 12: suggest again removing the title from the figure as it is inside jargon

The title has been removed

Page 36, line 12: Observed values of PM_{2.5} seem to reach 20 $\mu\text{g m}^{-3}$, not 30.

Page 37, Figure 13: suggest again removing the title from the figure as it is inside jargon

The title has been removed

Page 37, line 5: I am confused about the text here. i don't see a persistent low bias in the model throughout the year (red line = model higher than blue line = observations generally May - November)

It was meant only for January and February 2017: it is true that for other months the bias is more mixed. This whole section has been rewritten using MNMB and FGE instead of bias and RMSE but for MNMB the signal is the same.

Page 38, Figure 14: same comment about figure title

The title has been removed

Page 39, Figure 15: same Page 40, Figure 16: same Page 41, Figure 17: same Page 42,

The title has been removed

Figure 18: same

The title has been removed