

We thank the Topical Editor for her patience with conducting a final review of our manuscript and we provide a detailed response below.

[1] I had indicated: " please indicate whether meteorology can differ across simulations, depending on whether aerosol–radiation interactions are activated or not. You respond:" The meteorological component is the same between simulations and corresponds to Cy48r1." I'm sorry, but your answer is not sufficient: are there or are there not interactions between the gaseous species and/or aerosols with the radiative scheme? If there are, then the meteorology, even if reinitialized every day, is not identical across all simulations, and this should be clearly stated in the text. The meteorological component and the meteorological conditions are two different things.

The source code for the meteorological part of IFS used in the simulations corresponds to cycle 48R1, with atmospheric composition updates intended for cycle 49R1. In the simulations shown in this work, the meteorological initial conditions are taken every day at 0h00 from an independent analysis (ERA5), but the 24-hour forecast can indeed be impacted by interaction between gaseous species and aerosols with the radiation scheme. This means that the meteorological forecasts can indeed differ between the simulations, although the analysis is the same. Due to modifications being limited to the inorganic component the influence on meteorology is considered small. We now add in the manuscript: "Still, marginal differences may occur between simulations due to a coupling of gases and aerosol with radiation, which is active by default. "

[2] I had indicated: line 29: I propose to change: "There is also a shift in the size of particles towards the fine mode nitrate away from the coarse mode. " into "There is also a shift in particle size distribution, with nitrate moving from the coarse mode toward the fine mode." You respond: " The sentence has now been updated." I'm sorry, but the sentence has not been updated

We have now modified the abstract in line with the requests of the editor.

[3] I had indicated: Line 53: change "...and the concentrations" into "and concentrations" You respond: "We have now modified this sentence." I'm sorry, but the sentence has not been updated.

We now remove the word "the".

[4] line 54: no capital letter to sulphur dioxide, ammonia, nitric acid, particulate matter. And add a "," before ammonia. You respond: "We have now removed all capitals before the definition of the chemical species and the grammar has been corrected." Not done in the abstract, and "comma " not added.

Capitals are now removed. "We now write: '... sulphur dioxide (SO₂), ammonia (NH₃) and nitric acid (HNO₃).'"

[5] I had indicated "line 111: please reformulate : 'In that, acidic deposition and N-loading can also be output from the model means such that improving the deposition term via an improved distribution in PM will foster the development of this IFS-COMPO future product." You respond: We now use the sentence: "Acidic deposition and nitrogen loading can also be future products from IFS-COMPO which will benefit from the improved simulation and distribution of PM." OK, but you've added a sentence while keeping the original one, which results in a redundancy.

We apologize for this oversight. The original sentence has now been removed, and we now modify the new sentence to: Reduced and oxidised nitrogen deposition can also be future CAMS products which will benefit from the improved speciation between gaseous and particulate nitrogen.

[6] I had indicated "line 121: please clarify what you mean by 'upgrades to EQSAM4Clim' You respond: "We now modify the sentence to " . . . application of EQSAM4Clim (Metzger et al., 2024) . . ."where the details of the recent modifications can be found in this recent publication. I don't see this sentence in the text

We have changed this sentence towards: "The influence on regional wet and dry deposition terms are subsequently evaluated to assess the application of both EQSAM4Clim (Metzger et al, 2024) and the deposition schemes." The actual changes with respect to the inorganic aerosol modeling are described as part of Sec. 2.1

[7] Referee 2 indicated: "Did you check conservativity in the budget tables?" You respond: "Yes the global budget terms are closed and comparable across runs." So please indicate that in the text of the paper as well.

We now declare this specifically above each of the budget tables.

[8] line 18: add "(IFS-COMPO)" after Integrated Forecast System-COMPOsition

Acronym now added.

[9] line 20: add ', ' after "cycle 49R1"

Comma now added.

[10] lines 165 and 169: change "CY49R1" into "pre-CY49R1"

Now corrected.

[11] line 390: remove ', ' between "particles" and "are"

This comma is now removed.

[12] line 420: I propose to remove ' which is used operationally'.

Due to the rapid progress in IFS cycles and that the submission date of this paper was last year, we agree that this text doesn't necessarily provide the most up-to-date information, with the text now being removed.

[13] line 437: you indicate that "update includes adjustments to the below-cloud scavenging parameters" and then "Additionally, a below-cloud scavenging model has been implemented.". Do you mean that there is now in "pre-CY49R1" a separate routine for below-cloud scavenging? Please clarify

We apologize for this additional this sentence "additionally a below-cloud scavenging model has been implemented" should not have appeared. The changes to the scavenging parameterization as evaluated here are exactly as described just before this sentence. We now remove this sentence.

[14] line 480: please replace" The details of the sensitivity experiments" by "The details of the experiments"

We have now removed the word 'sensitivity'

[15] line 492: please replace "The emissions adopted" by "Anthropogenic emissions"

We have changed the text as requested. We have slightly updated the specification of the emissions in the model, to improve the readability:

The anthropogenic emissions adopted in these configurations are taken from CAMS_GLOB_ANT v5.3 (Soulie et al., 2024), biogenic emissions taken from the CAMS_GLOB_BIO v3.1 dataset (Sindelarova et al., 2022; <http://eccad.aeris-data.fr/>) and biomass burning emissions taken from GFAS v1.2 (Kaiser et al., 2012). All emissions are provided at 0.1 x 0.1 resolution at a monthly frequency, except for the biomass burning emissions which are prescribed daily. For biomass burning and anthropogenic emissions from specific sectors vertical profiles are used representing pyrogenic convection or industrial stack heights, with other emissions being applied in the lowest model level.

[16] line 493: remove "with"

We have reformulated this sentence and removed the word 'with', see the response to query '15' above.

[17] line 500: remove ',' after << based >>

The comma is now removed.

[18] line 549: you write 'Unfortunately for S. E. Asia measurements of both precursors and SIA at a weekly time frequency ...'. Please rather indicate: "Unfortunately, conducting a similar analysis in Southeast Asia was not possible owing to the unavailability of both precursor and SIA weekly measurements."

We now replace the sentence as requested.

[19] line 549: '.' after study

We have now updated the grammar.

[20] line 578: the sentence "A direct link...": this last sentence of the paragraph does not logically follow from the previous ones

We agree with the topical editor and have now removed this sentence.

[21] line 586: you write: 'The maps for December show higher mixing ratios towards the East, with a significant contribution from shipping.'. Is indeed shipping responsible for these higher mixing ratios?

We agree that this description is not clear enough and now modify the sentence accordingly: The maps for July show higher mixing ratios towards the east, with a significant contribution from shipping in the Mediterranean and the English Channel. For December, the highest mixing ratios occur over the continent, most notably Germany and Poland.

[22] line 678: "by 1.5%" or by "by 1.4%" as indicated in Table 2?

Now corrected towards 1.4% as in the Table, thank you

[23] line 699: please clarify 'The yearly mean bias (MB) value decreases by around 25%, with a moderate correlation.'

We now remove this sentence and limit the discussion related to the evaluation of SO₂(g) to lines 352-405.

[24] line 791: legend of Fig. 2: please specify what the grey band means, and provide details on the variability criteria used (the latter comment applies also to Fig. 5 and Fig. 8)

*Fig. 2, 5, 8 (grey band):
“As requested, we now specify that the grey band denotes the station range (min–max) of observed weekly means at each time. This is applied consistently in Figs. 2, 5, and 8.”*

[25] line 801 : “Although some smaller differences” : smaller than what?

We now modify the sentence: Although some small differences in the SO₂(g) weekly values occur between simulations, there is no improvement in pre-Cy49r1 with respect to the weekly MB. The positive summertime MB for SO₂ implies that there is either an overestimate in the regional source terms or underestimate in the depositional loss term (c.f. SO₄²⁻ below).

[26] line 854: “which would then lower the negative bias” : don’t you mean the positive bias?

We have now modified this sentence into “... which would then lower the positive bias seen for [SO₂(g)].”

[27] line 856: “means of surface [SO₂(g)]”: I don’t see any SO₂(g) presented in Fig. 3; please correct the sentence.

We have now corrected this sentence and removed reference to [SO₂(g)].

[28] line 944: “in Fig 4”: isn’t it rather Fig 3?

Thanks for highlighting this typo. We now correct the table 3 legend to refer to Fig 3.

[29] line 931: legend of Fig. 3, and in the legends of all equivalent Figures in the paper, change: “The site locations used are shown in each pane and taken from the EMEP, CASTNET and EANET networks, respectively.” into “Site locations and corresponding observed values, taken from the EMEP, CASTNET and EANET networks respectively, are shown in circles”.

The figure legend has been replaced, also for Figs. 6, 9, 10, 11 and 12.

[30] all throughout the paper, please remove capital letter for northern, southern, southeast, etc... when they are adjectives, and when they don’t refer to an official region

We homogenised the naming convention (e.g., ‘Southern U.S.’ → ‘southern U.S.’; Northwest → ‘northwest U.S.’), harmonized all occurrences to ‘pre-Cy49r1’ (and ‘Cy48r1’), including section headers and table labels. We retain Southeast Asia as this is a named region in the manuscript.

[31] line 1354: please indicate in a couple of words what an aerosol dynamical model is

“Here, an aerosol dynamical model refers to a microphysical framework that prognoses particle number/size distributions (via condensational growth, coagulation, nucleation and inter-mode transfer), thus enabling kinetically limited gas-to-particle uptake and condensation processes beyond the instantaneous thermodynamic equilibrium assumed by EQSAM4Clim.”

[32] line 1356 : please clarify: “imposed in pre-CY49R1 occurs”

We have now modified the text to: “For December, the relative differences in pre-Cy49R1 compared to Cy48r1 show increases in HNO₃(g) at latitudes below 60°N, together with decreases of between 25-75% at latitudes higher than 60°N, i.e. at lower temperatures, and under a relatively low NO_x environment.”

[33] line 1363: please clarify:” In contrast to Europe no seasonal decreases occur for any location.”

To better clarify we now change this sentence into: “In contrast to Europe no significant decreases in $[\text{HNO}_3(\text{g})]$ occur for any location or season.”

[34] line 1364: what do you mean by: “are the high across all regions”?

We change the formulation of this sentence into: Finally, in comparison to the other regions the surface $[\text{HNO}_3(\text{g})]$ are highest for Southeast Asia. Maximal values reach the order of 4-5 ppb towards the eastern coast (July) and central regions (c.f. December).

[35] line 1433: change “op” to “top”

We have now corrected this typo.

[36] line 1444: please move the reference (Metzger et al., 2018) when first mentioning “aerosol dynamical model”

We have now moved the reference to the first mention of coupling to an aerosol dynamic model.

[37] line 1752: “(bottom right panel of Fig. 8)”: is this the correct figure?

Figure 8 pertains to the comparison of both HNO_3 and NO_3^- against EMEP and CASTNET weekly values such that we do reference the correct figure here.

[38] line 2321: “towards Iowa are not seen in the measurements”, it’s not clear to me what you are pointing here

We now replace Iowa with central U.S. to indicate the location of maximum concentrations.

[39] line 2324: please reformulate “yearly a result of the large positive MB towards the east being moderated by negative in other parts of the U.S”

We have now reformulated the sentence as requested.

[40] line 2328: “This provides ” : what does “This” refers to?

We now replace the beginning of the sentence: “The yearly deposition values range between ... “

[41] line 2673: “show”

We now modify the sentence to: “The global chemical budget terms provided in Table 6 reveal that there is an increase in the gas-phase production term for HNO_3 , ...”

[42] line 2698 “do reveal”

We now correct the grammar of the sentence.

[43] line 2704: high values in the observations are not due to missing sources. Please reformulate the sentence.

We agree that emphasis should not be put on missing source terms and we now remove the reference to “missing local emission sources”.

[44] line 2724: “and has little effect on the forecasts itself”: please explicit what has little effect

We now shorten the sentence to remove the implied effect on chemical forecasts.

[45] line 2739 “an quantify”?

Now corrected to “... and quantify .. “

[46] line 3033 : please remove 'that have'

We have now reformulated the sentence as follows: The gaseous precursors, SIA surface concentrations and wet deposition totals for the three dominant global source regions (Europe, the U.S., and southeast Asia) have been evaluated against weekly/yearly observational composites for the year 2018."

[47] line 3040: in the sentence "For surface [SO₂(g)], no significant impact has occurred with respect to the MB for Europe or the U.S. and with little correlation. ": please be more precise for "MB" and what you mean by "and with little correlation." More generally, all through the conclusions, precise when you provide numbers what they refer to, that they are for yearly means or for ...

We have now changed the sentence to: "For surface [SO₂(g)], no significant impact has occurred with respect to the mean bias over Europe and the U.S.. Also the correlation in the weekly variability improved only marginally"

[48] line 3044: "For the yearly wet deposition of oxidized S, results are mixed with reductions in the MB for Europe and China but increasing markedly for the U. S.". Please correct the grammar

We have now reformulated the sentence to: Reductions in the MB are found for Europe and China, whilst for the U. S. the MB increases markedly.

[49] line 3052: please reformulate "from a reduction in the efficacy of particle"

We have now reformulated this sentence towards: "For [NH₄⁺], the application of EQSAM4Clim significantly reduces the associated MB against observational yearly means by approximately 45% for all three global regions. This is explained by a reduction in the efficacy of particle formation, especially during summertime."

[50] line 3061: "persistent" in what sense?

Persistent with respect to the length of time for which the bias exists. We have now reformulated the sentence to: "For HNO₃(g) in Europe and the U.S., there is a persistent negative bias during the summertime which is changed to a significant positive bias due to a larger fraction remaining in the gas phase. For the U. S., there is a degradation with a relatively low bias changing to a large positive bias for the entire year, which indicates an incomplete process coupling when considering mineral cations without aerosol dynamics. "

[51] line 3159 : I understood you did not get weekly observations from EANET. Please correct the legend of the figure

We now remove the word weekly from the figure legend.