

We thank both referees for their comments and suggestions on our original manuscript concerning the validation of the concentrations and wet deposition of secondary inorganic aerosols in the CAMS forecasting system. This has resulted in a major revision of both text and figures in order to address the suggestions and critique from the two anonymous referees. We have ensured that the referencing to all Table and Figures is now correct. We provide a detailed response to both reviews below.

Responses to referee #1:

1- The description of the IFS-COMPO model is very succinct, and details of its main components, particularly aerosol management, would be appreciated. Schematic figures, for both model version, showing the interaction between the various modules (IFS-COMPO, IFS-AER, EQSAM4Clim, etc.) and the changes between the two versions would be relevant.

This paper is a companion paper to that published by Remy et al. (2024) in which a comprehensive description of the updates made between CY48r1 and CY49r1 are given, and a schematic of how all the components in the model interact can be found as Figure 1 in both Remy et al. (2024) and Metzger et al. (2024). In the interests of brevity we refrain from repeating or republishing this schematic. For instance, we make no mention of IFS-AER as this is the aerosol only version of the model which is not used in this study and would add confusion if introduced.

We include the following sentences to point the reader to the other papers: vis : “A more comprehensive description of Cy49r1 updates is provided in Rémy et al. (2024) and of the EQSAM4Clim thermodynamic module in Metzger et al. (2024), along with a schematic showing the interaction of these different models towards providing accurate air quality forecasts.”

2- The observational networks (section 3) are too rapidly described. More information is needed on the different stations in each dataset (number of stations, locations, data available for the period used, etc.). Figures showing the location of the various stations and a discussion of the advantages and weaknesses of each dataset would be useful in this section. Additional information, such as the uncertainties associated with these datasets or references on their reliability, would also be useful. Putting all the necessary station information in this section will also help to lighten the text and figures in the rest of the article.

In order to improve on the ability to directly compare results across different aerosol species we now reduce the number of measurements datasets which we use in the manuscript to three main sources : EMEP, CASTNET (with AMoN for $\text{NH}_3(\text{g})$) and EANET. We introduce a figure in the Appendix which shows the location of all the stations used for the evaluation. The lack of frequent gas-phase measurements for South-East Asia means that the statistics are now computed for the annual means of particle concentrations and wet deposition totals only for this region.

3- All figures in the article need to be greatly improved and standardized. The quality of the figures needs to be improved, with simpler, more legible titles and more

complete captions. Appendix figures should be similar to those in the article (legends, curve colors, titles, etc.) and use the names of the article simulations. Many of the figure numbers in the article are also incorrect. See specific comments for details. Some of these issues should have been fixed before publication in EGU sphere.

We have reviewed the figures upon the request of the referee. We have improved the quality of the figures showing both the surface distributions (original figures A1, A3 and A6) and the evaluations (Figs. 3, 5, 7-10), within the limits placed by the journal on the total figure size. The gas-phase comparisons against both EMEP and CASTNET have been remapped using different software. We have ensured the clarity has been improved such that, upon viewing the manuscript at high resolution, shows the distribution in fine detail.

4- Despite the improvements brought by the new version of the model, and in particular by the implementation of EQSAM4Clim, many biases are still present. Further discussion of ways to improve these biases would be relevant. Finally, this article focuses on 3 specific regions (Europe, the United States and Asia), but some informations on other parts of the world and on the various limitations of this study would also be interesting.

We now add some discussion on the most probable causes of the remaining biases for both the pre-cursors and the associated particles, although without specific sensitivity studies we refrain from putting emphasis on any dominant cause. In that we are unable to validate the surface results for e.g. Africa and South America with the same accuracy as the three selected regions means we refrain from expanding this to global scale. Without a meaningful evaluation of all the components involved in the formation of SIA this would detract from the main message of the paper.

Specific comments

- Page 4, lines 167-168: Please add a reference.

“For convective precipitation, the assumed precipitation fraction has been standardized to 0.05 (whereas in CY48R1, a value of 0.1 was used for chemistry scavenging and 0.05 for aerosol scavenging).”

- Page 5, lines 214, 227: As mentioned above, indicate the number of stations, develop selection criteria, etc.

We have introduced a figure in the Appendix showing the distribution of measurement stations from the EMEP, CASTNET and EANET measurement networks used for the evaluation of the e.g. SO₂(g) surface concentrations. For the other species (i.e. SIA particle distributions and wet deposition totals) the location of the measurement stations are shown on the respective figures (Figs. 3, 6, 9-12). We have improved these figures including the use of black circles to highlight the locations.

- Page 5, line 232: “Acid Deposition Monitoring Network in East Asia” – > “EANET” (acronym already explained line 225).

Second definition now removed.

- Page 6, line 253: “Nitrate#1” – > fine mode nitrate?

Now replaced with the correct naming.

- Page 6, line 255: A map with station locations would be useful here. Refer to section 3 if such a map is added.

We have introduced a figure into the Appendix showing the locations of the measurement stations used for evaluating the gas-phase precursors for Secondary Inorganic Aerosols for evaluating the performance of IFS-COMPO. For the wet deposition evaluating the location of the stations are provided on each of the deposition maps shown with a colour coded circle also giving the observed levels of deposition in ug/m3 (c.f. Figure 11).

- Page 6, line 259: “we show monthly mean regional differences for July and December 2018” – > Why did you choose these two months?

These months are representative of summertime and wintertime performance, where seasonality exists in regional emission estimates related to e.g. regional heating (SO₂) and agriculture (NH₃) and also meteorology.

- Page 6, line 268: Bias already cited in the literature?

See Remy et al. (2024) for further details of such biases. We have now made a direct reference in the text.

- Page 7, Table 2: RMSE line missing. Indicate the relative differences (in %) between the two cycles as done in other Tables. Remove “diagnostic” and add “SO₂” to harmonize with other tables.

This table has now been removed to align the comparisons across species and the remaining tables subsequently renumbered.

- Page 7, Table 3: Harmonize the legend of all tables in the article. “Percentage difference changes are calculated as ...” or “with the associated relative differences provided in parenthese ...”: Choose one sentence and use it for all the Tables.

We now ensure that the table legends are consistent with each other for the three budget tables.

- Page 7, line 312: “Figure 1A” – > Figure A1

This figure has now been removed from the paper.

- Page 7, line 312: “the region with the highest surface SO₂ concentrations is the northeastern U.S.” – > Please rephrase the sentence, not consistent with the figure.

We have now moved this figure into the main text (becoming the new Fig 1) and we now rephrase this sentence to “Figure 1 shows the Eastern U.S. exhibits the highest surface SO₂”

- Page 7, line 313: “There is little seasonality in the weekly observational composites” – > consistent with literature?

We are referring to the observational weekly means derived from the CASTNET stations in the U.S. for this specific year therefore feel we do not need to refer to any previous study. We have significantly expanded on the description of these sites as requested by the referee.

- Page 7, lines 318-319: “SO₂ emissions in the global inventory are significantly overestimated” – > What about the literature? Has this problem ever been reported or documented?

We refer to the specific SO₂ emission inventory as developed in the CAMS consortium which are constantly being modified in response to model performance. The comparisons shown in this paper for both Europe and the US show consistent positive biases of > 100%, where [SO₂(g)] is critically dependent on emission fluxes applied (no chemical production term is included in the chemical scheme for SO₂). The methodology used for deriving the CAMS-GLOB-ANTH inventory is given in Soulie et al., (2024), Earth Syst. Sci. Data, 16, 2261–2279, <https://doi.org/10.5194/essd-16-2261-2024>, 2024.

- Page 8, Figure 1: Please give each figure a simpler, clearer title (ex: SO₂ - Europe, SO₂ - US, SO₂ - China). Also use a vertical title for the two figure lines, to the left of the first figure of each line. Put a title on the y axis of the figures. Complete the legend with all the necessary information, but do not include this information in the figure titles. Also describe the second line of figures in the legend. See also to put only one curve legend for each set of figures to lighten the figures. In the caption, please refer to section 3 for the description of the different observation datasets. The figure is not very sharp, please use a PDF or EPS file.

This figure has now been removed to align the comparisons across species and the remaining tables subsequently renumbered. It has been replaced by evaluation using measurements from the EMEP and CASTNET networks including all stations.

- Page 8, Figure 2: Please give a simpler and clearer title (ex: SO₄ - Europe). Put a title on the y axis. In the caption, please refer to section 3 for the description of the different stations used.

We have remade this figure and removed the previous Fig A2. Now European and US comparisons of SO₂ and SO₄²⁻ are displayed side by side. We now change the figure legend to : “A comparison of weekly mean SO₄²⁻ for Europe (top panel; µg/m³) and the US (bottom panel) simulated in Cy48r1, Cy49r1_NOE4C and Cy49r1 as compared against measurement composites from stations representative of a rural scenario taken from EMEP and CASTNET observational networks, respectively, for 2018. The sampling frequency of the data for South-East Asia does not allow a corresponding weekly plot for this region.”. We also simplify the figures titles as suggested.

- Page 10, Figure 3: Review the titles of the various figures (unclear and illegible). Put a title for each column (ex: CY48R1 and CY49R1) and each row (ex: SO₄ - Europe, SO₄ - US, SO₄ - Asia). Put only one colorbar per line to make it more legible and add

its own unit. In the caption, please refer to section 3 for the description of the different observation datasets.

We have now completely restyled this Figure and changed the titles and provided a higher resolution version, where the location of the observational stations can be seen more clearly by adopting a black surrounding circle. We provide only one colour bar now at the bottom of each column.

- Page 12, Figure 4: Please give a simpler and clearer title for each figure (ex: NH3 – Europe and NH4 -Europe). Put a title on the y axis. In the caption, please refer to section 3 for the description of the different stations used. Harmonize all figure captions of the article. “The corresponding biases are shown in the bottom panel.” is not appropriate here.

We have now moved Fig. 4A into the main text such that the comparison between the performance in Europe and the US can be made more easily. We have also homogenized the figures legends as for the corresponding plots for SO_4^{2-} .

- Page 15, Figure 5: Same as Figure 3.

We have restyled this figure with similar changes as those described for Figure 3.

- Page 16, lines 513-519: You refer twice to Figure A7, but it's Figure 6, isn't it?

Thanks for finding the typo which is now Corrected.

- Page 16, line 520: I think this is Figure A6 and not Figure A7.

This figure has now been moved into the main text.

- Page 17, Figure 6: Same as Figure 4.

We have now moved Fig 7A into the main text such that the comparison between the performance in Europe and the US can be made more easily. We have also homogenized the figures legends between HNO_3 and NO_3^- for both regions.

- Page 19, Figure 7: Same as Figure 3.

In line with Figure 3 we have improved this figure addressing the issues raised on figure legends and clarity.

- Page 20, Table 8: Remove % from this table to harmonize with other tables. Please correct ug – > μg (also for Table 4 and 6).

We have now homogenized the format across all tables.

- Page 20, lines 621-630: Not the same font.

This seems to have been introduced during the typesetting process as in our submitted version everything is formatted as Times New Roman as requested in the submission.

- Page 22, line 664: Here too, I think it's Figure 8 and not Figure 11.

Thanks for finding the typo which is now Corrected.

- Page 23, Figure 8: Same as Figure 3. - Page 25, Figure 9: Same as Figure 3. - Page 27, Figure 10: Same as Figure 3.

These three figures have been improved in line with the comments made related to clarity and the figure legends.

- Page 26, lines 780-781: Figure 13 – > Figure 10. Please check that the text and figure numbers are consistent throughout the article.

We have made major changes to the manuscript where we have moved some of the figures which were in the Appendix into the main body of the text on the request of the reviewers. Therefore all figures have been renumbered throughout the article, with care taken not to reference incorrect figures.

- Page 29, Figure A1: Make a more legible figure. For example, use clear headings for each column (ex: NH3 - CY48R1, Diff A, Diff B, Diff C). Also include Europe, US and Asia, as well as July and December for the corresponding vertical rows on the left of the figure. Delete sub-figure headings for greater legibility.

We have restyled and improved the presentation of this figure following the suggestions of the referee and moved the figure into the main text.

- Page 30, Figure A2: Harmonize the titles (ex: NH3 – US and NH4 – US), axes, axis titles and legend with the figures in the article. Use the same curve colors as the other figures (keep the same color for observations and different versions throughout the article).

We have now moved this figure into the main text and replotted to homogenize the figure legends and colour scheme.

- Page 31, Figure A3: Same as Figure A1.

See response related to original Figure A1 which has now been moved into the main text.

- Page 32, Figure A4: Same as Figure A2.- Page 33, Figure A5: Harmonize the figure with the other figures in the article. Put the names of the versions used in the article and add observations in the caption. Use station name as sub-figure title.

This figure has now moved into the main text and replotted to address the issues listed by the referee.

- Page 33, Table A1: Add NH3 to the table to harmonize with the other tables. In the caption, please refer to section 3 for the description of the different stations used here.

We have now changed the title of this table to be more compatible with the other tables presented in the manuscript.

- Page 34, Figure A6: Same as Figure A1.; Page 35, Figure A7: Same as Figure A2.

Both figures have now been improved and moved into the main body of the text.

Responses to Referee #2

Specific comments:

Concerning the figures, their quality is overall poor, especially in the Appendix. I would recommend to do an effort to increase their quality and readability. Also the partitioning between the paper and appendix should be questioned. Figure A1 for example is used several times and is worth to appear in the main part of the paper.

In response to the referee's criticism we have now worked on the quality of the figures throughout the manuscript and enhanced the readability. The three plots concerning regional changes in the gaseous precursors simulated in IFS-COMPO have now been moved into the main text and restructured the manuscript accordingly. We also address the issues raised by referee #1 regarding figure legends.

As you are not using the “real” CY49R1, but modified version of CY48R1, I would recommend to delete all reference to CY49R1 except from the fact that the presented modifications are meant to enter in the latter.

This paper is a companion paper to one already published in GMD (Remy et al, 2024) and we use the same nomenclature as in this publication to avoid confusion. All the updates described and evaluated here are active in Cy49r1 therefore the results presented pertain to this version of the model. Given that Cy49r1 is operational we now change to text to reflect this e.g. by removing the word 'proposed'.

A section is missing to explain how are computed the statistics. Are all the data gathered in one vector and the tables represent spatio-temporal statistics

The statistics are calculated with the in-house statistics package available at ECMWF called which are used for evaluation throughout the CAMS initiative. We now add the following sentence : “All statistical metrics represent spatio-temporal averages unless otherwise noted, combining all station-time pairings into a single evaluation vector per region and species”

- Line 126: “which will be operational in November 2024”: Please update this sentence as the date is now expired.

Thanks for the comment. Now replaced with “ ... to be operational in 2025).

- Table 1: The Experiment IDs are not used in the text. Please remove them or add a sentence to tell that they can be used to retrieve the data on the MARS storage system.

We retain the experimental ID's such that readers have the information to access the original simulations at any time in the future. We add the following sentence in the Table Legend : “The experiment ID's can be used to retrieve the original data from the MARS archiving system hosted at ECMWF.”

- Line 205: Please add a reference for the direct production from hot shipping exhausts.

We now include a peer reviewed reference related to this statement.

- Line 211: AirBase is no longer used since mid 2000's. The new system is called AQeR (AirQuality e-Reporting)

In the new manuscript we remove the use of AirBase data.

- Line 214: How did you select "rural background stations"?

In the new analysis we now use all available stations rather than limiting the analysis to a subset.

- Line 271: Figure A1. Please introduce the figure before referencing it. Also Fig. A1 is used a lot. Maybe it would be interesting to move it to the main part of the paper.

We now move the original Figures A1, A3 and A6 included in the original supplement into the main text, where change the text

- Line 272: Please introduce Table 3 before using it.

We have amended the text to : "The associated annual mean statistics are provided in Table 2. To assess the global integrated impact on SO_4^- formation, the associated global budget terms are provided in Table 3 in Tg S/year. "

- Line 272: "For CY49R1 [...] by approximately 1-0. $\mu\text{g}/\text{m}^3$ " → I assume you are talking about Fig. 2 results. Please introduce it and announce it properly or move the sentence in the SO_4 --paragraph.

We have now re-written this paragraph expanding on the results and to introduce the Figure in the correct place.

- Line 274: Figure A1.

This figure has now been moved into the main text such that this appendix Figure labelling is now obsolete.

- Line 279: You introduce Table 3, but you used it only in the next paragraph.

We have now introduced and defined Table 3 immediately after the introduction/definition of Table 2 to allow it to be used in later discussion.

- Line 306: "Statistics relate to seasonal means" → Please be more precise about this sentence. Maybe in the section 3 when introducing observational data.

We have now provided details regarding the statistics used.

- Table 2: For Europe, according to the section 3, data must be AQeR (replacing AirBase). For US it is AirNow, and China CNEMC.

Thank you for catching this error. We have now re-labelled the columns.

- Table 2: RMSE are missing in the table.

Table 2 has now been removed.

- Table 2: Why would you omit CY49R1_NOEAC in the Table, as you put the results on Fig. 1?

Table 2 has now been removed.

- Table3: (also valid for other budget table): Maybe you cloud add a minus sign in front of figure representing a loss for the considered species.

Thank you for the suggestion. However we do not provide the differences in terms of Tg species and cannot use colours (red for increase, blue for decrease) due to the journal protocol.

- Line 312: Figure A1 ; Line 316: Weekly or seasonal?; - Figure 1: For European figures SO₂_surf → SO₂ and EBAS → AQer. For US figures, AirBase → AirNow. ; - Line 322: For China, is it weekly or seasonal? ; - Line 326: “along with associated biases”. Add “not shown” or modify Fig. 2., - Figure 2: The format of Fig. 1 is very interesting, why not use the same for Fig. 2 and have the three area and the biases on the same figure?; Figure 2: Change EBAS to EMEP in the title.

This figure has now been removed

- Line 341: “The low MB for SO₂ [...] is too fast”. I don’t understand this sentence. What rate of oxidation for which species? Please be more precise.

Comparing the associated negative bias in SO₂ and the high positive bias of SO₄= for summertime (with higher OH) suggests that the lifetime (chemical oxidation rate) is too low, where less SO₄= would equal more SO₂ improving both comparisons. We have now rewritten the sentence to ; ‘Considering the corresponding low summertime MB for SO₂ in Figure 2 shows that the rate of oxidation in IFS-COMPO is too fast, where a slower oxidation rate of SO₂ by OH and/or aqueous phase processing would be required to improve the performance of IFS-COMPO.’

- Line 354: Are you referring to direct production from hot shipping exhausts?

We now extend the sentence with “...here related to missing shipping emissions of SO₂, which quickly converts to SO₄² in the plume (Celik, et al., 2020).”

- Line 370: Please add that you plotted surface concentrations.

The figure legend has now been amended.

- Figure 3: Why not plotting also CY49R1_NOEAC

The clarity and quality of the original figure has been significantly improved., as requested by anonymous referee #1. We feel that by including additional panels for the CY49R1_NOE4C simulation would introduce potential confusion due to the size of the measurement stations being so small

- Line 388: I don’t understand the reference to Fig. 8-10.

Thank you for finding this error now changed to the new Fig.4.

- Line 392: EBAS → EMEP

We now correct this typo.

- Line 401: Figure A3.

We have now moved this figure into the main text.

- Line 401: “The corresponding [...] in Table 5.”. This sentence is not properly places as you don’t use Table 5 for a while.

We have now moved this sentence as requested by the referee.

- Line 403: “Tich?” → “Tichy”

This seems to have been related to a typesetting issue rather than a typo from our side with the representation of y with a hyphen missing.

- Figure 4: EBAS → EMEP in the title

We now move the name of the dataset into the figure legend.

- Line 423: Introduce Table 5 here.

We now introduce Table 5 (new Table 4).

- Line 434: “Although maximum [...] during winter.” Do you have an explanation for this phenomena?

This is due to the low volatility of NO_3NH_4 during the colder temperatures during wintertime, resulting in a longer tropospheric lifetime than in the summertime. The impact of EQSAM4Clim only becomes apparent after April due to the HNO_3 limiting particle formation as described in Tang et al, ACP, Atmos. Chem. Phys., 21, 875–914, <https://doi.org/10.5194/acp-21-875-2021> 2021. We now include extra discussion on this issue.

- Line 439: Figure A5 → Fig. A4

This figure has now been moved into the main section of the text and the numbering of figures updated accordingly.

- Line 441: “This suggests [...] of HNO_3 ” → Is this a remark true at global scale?

This is a physical limitation to NO_3NH_4 formation and the sensitivity to $\text{HNO}_3(\text{g})$ has been determined by comparing multiple species associated with SIA formation (Tang et al, ACP, 2021). For the tropics the high temperatures mean that the lifetime of NO_3NH_4 is very short and somewhat limits maximal concentrations.

- Figure 4 also seems to show that the emissions of NH_3 are too strong in Europe when considering NH_3+NH_4 . What can you say about this? Also, in the US what are the implication of the ammonium aerosol formation together with sulfate that are overestimated according to Fig. A2?

Independent studies have shown that the emission estimates for $\text{NH}_3(\text{g})$ are realistic where we include the following sentence : “The CAMS_GLOB_ANT v5.3 (Soulie et al, 2024) emission inventory has recently been validated for NH_3 against top-down estimates providing confidence in the quality of the estimates for Europe (Ding et al., 2024). “. The fraction of $\text{SO}_4=$ bound the NH_4+ is accounted for in EQSAM4Clim therefore the excess in $\text{SO}_4=$ shown in the new Fig. 2 is the form of non-bound $\text{SO}_4=$.

- Line 455: Table A1 - Line 456: Figure A3?

Table A1 has now been removed and we have also moved the Figure from the Appendix into the main text.

- Line 456: “Differences between [...] burden in Table 4.” You are comparing surface field with 3D diagnostic. This is not necessarily relevant, especially when it comes to SIA.

The vertical profile of NH_3 means that most of the particle formation occurs in the lower troposphere for NH_4^+ therefore we can use these changes in a qualitative sense.

- Line 459: -0.26 $\mu\text{g}/\text{m}^3$

Now corrected in the text.

- Line 479: *I didn't see a proper discussion section.*

We now modify this text to remove the suggestion of further discussion.

- Line 484: *Table 5*

We now correct this typo.

- Line 486: *Figure A3* - Line 488: *Figure A5*

Both figures have now been moved into the main text and the numbering changed in the text.

- Line 503: *Table 6*

We have now changed the reference to the correct table in the figure legend.

- Line 514: *Figure A6?* - Line 519: *Figure A6* - Line 520: *You introduce Fig. A6 now, after referencing it twice.*

This figure has now been moved into the main text and the Figure renumbered as Figure 8.

- Line 521: *I supposed you mean relative difference in percentage.*

We have changed the text to relative differences.

- Line 523: *Please add a reference for the sentence about direct HNO₃ emissions.*

We now add : Vinken, G. C. M., Boersma, K. F., Jacob, D. J., & Meijer, E. W.: Accounting for non-linear chemistry of ship plumes in the GEOS-Chem global chemistry transport model. Atmospheric Chemistry and Physics, 11(22), 11707-11722. <https://doi.org/10.5194/acp-11-11707-2011>, 2011.

- Line 536: *I thought you didn't have direct shipping emissions?*

We have emission of CO, SO₂, NO₂ and VOC as integrated into the anthropogenic global emission inventories, but we do not have direct emission of either SO₄⁼ or HNO₃ as described in the text. We now clarify this in the text.

- Line 545: *please keep using ppb all along the paragraph.*

We have now changed ppt to ppb throughout.

- Figure 6: *EBAS* → *EMEP* in the title.

We now move the name of the dataset into the figure legend.

- Line 557: *"The evolution [...] bottom panel" I don't see any bias plotted.*

We now improve these figures by including corresponding bias plots for these type of comparisons in our amended manuscript.

- Line 559: *Fig. A7*

This figure has now been moved into the main section of the text and the numbering of figures updated accordingly.

- Line 569: *Table 8 is not introduced*

Table 8 should have been Table 7 (new Table 6). This typo is now corrected.

- Line 578: Fig. A7

This figure has now been moved into the main section of the text and the numbering of figures updated accordingly.

- Table 7: relative difference for coarse NO₃- in CY49R1 is false.

We apologize for the error and have now corrected the change in the respective budget term.

- Line 597: I would rather write 0.2-2µg/m³

Now changed in the text

- Line 612: please add “surface” concentrations

Now added in the text.

- Line 645: “corresponding” → “comparison with observations”, or something approaching.

This sentence has now been changed.

- Line 649: “following figures” → what figures?

This pertains to the regional comparisons of the wet deposition totals which follow the tables. We now specify which figures.

- Line 664: Figure 8 should be introduced in the next paragraph as you use it.

We have now renumbered the figures and introduce the comparison of total S wet deposition in the correct place.

- Line 665: “To allow [...] 1000 mgS/m²/year” You should remove this sentence as it doesn’t reflect the figures content.

We have now changed the text for 1000 mgS/m²/year to 500 mgS/m²/year so as not to be misleading.

- Line 682: “by around 10%”

Now changed in the text.

- Line 694: I suppose it is Table 4.

The tables have been renumbered and we ensure that we refer to the correct table in the text.

- Line 699: temporal → spatial

Now changed in the text.

- Line 702: Do you have volcanic passive degassing emissions in IFS-COMPO? Please discuss also this aspect.

Volcanic outgassing is included in the simulations as detailed here <https://atmosphere.copernicus.eu/eruptive-emissions>, which does contribute to the background SO₂ concentrations in specific (typically remote) locations. Given the lack of measurement stations in Italy we do not think that this impacts our results in any way for the chosen simulation year. We now mention this in the description of the simulations.

- Line 705: As Fig. A1 is quite difficult to analyse due to its poor quality, we have to believe you.

This figure has now been provided in more high resolution as .eps format and amended in accordance with the requests of referee #1.

- Line 744: *I don't see a measuring point in Iowa.*

There is a measurement station situated in the bottom left corner of the state but we agree that this is difficult to see on the figure. We have improved the visibility of the location of the measurement stations on the new version of the figure.

- Line 747: *0.77 → 0.72, or a high correlation is achieved in CY48R1.*

We have now corrected the value of the correlation coefficient for CY49r1 in the text.

- Figure 8, 9 and 10: *Labels are unreadable for Asia*

These figures have now been replotted and provided at a higher resolution, where modifications have been made according to the suggestions of referee #1. The location of the stations used for validation have also been made clearer.

- Line 715, 760 and 799: *Table 10*

All tables have been renumbered and we ensure that we reference the correct table in the new figure legends.

- Line 780: *Figure 13?*

We have now renumbered most of the figures due to shifting those in the Appendix into the main body of the text.

- Line 788: *I don't understand the sentence about the fully coupled forecasting system. Please explain.*

This refers to the lack of data assimilation in the IFS-COMPO simulations used in the manuscript. We now provide more clarity in the text.

- Line 790: *"values range from 50-800 mgN/m2/year with the highest value (>2000 mgN/m2/year)". If the values range from 50 to >2000, please write 50 to 2xxx.*

We have now modified the text.

- Line 806: *"with the next operational IFS version (CY49R1)". No you didn't use CY49R, you used CY48R1 with aerosol evolution submitted for CY49R1.*

We refer the referee to our answer given above regarding the nomenclature of model versions.

- Line 816: *I would not be so direct. Indeed some aspects are better represented using EQSAM4Clim, but other don't. Please be more specific about this part of the conclusion.*

We have now removed this sentence and present the conclusions in a less generic manner.

- Line 854: *Please modify seasonal with monthly*

We have now modified the figure legend.

- Figure A4: *how many stations were used?*

We have now moved this figure into the main text such that direct comparisons can be made against the EMEP results. The figure has been replotted, formatting homogenised and clarity improved.

- Figure A5: The observations are missing in the legend. Also use CY4xxx as reference instead of Experiment IDs.

In the updated manuscript we have reflected on the inclusion of these comparisons and have decided to remove it as we only presented this for NH3 and, although they are included in the regional statistics for the US, feel they do not contribute to the main flow of the manuscript.

- Table A1: relative difference for CY49R1 bias and RMSE should be negative, - Line 896: As for figure A1

This table has been removed and the figure moved into the main text.

Author contribution: I don't understand VH contribution as you did not use the "complete" CY49R1 cycle, but only a modified version of the CY48R1.

VH was a PI of the CAMS35_2 project which funded this work and was involved in updating the infrastructure of the code as used here. This is stated clearly in the author contribution section.

Please modify AirBase with AQeR and add CNEMC.

We have removed this analysis from the manuscript therefore we no longer use the datasets.