We thank anonymous referee #2 for the 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 #2

Specific comments:

Concerning the figures, their quality is overall poor, especially in the Appendix. I would recommand to do an effort to increase their quality and readibility. 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 recommand to delete all reference to CY49R1 expect 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-Reporing)

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.µg/m3" → I assume you are talking about Fig. 2 results. Please introduce it and announce it properly or move the sentence in the SO4--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 SO_2 in Figure 2 shows that the rate of oxidation in IFS-COMPO is too fast, where a slower oxidation rate of SO_2 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 SO_2, which quickly converts to SO_4^2 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 HNO3" →Is this a remark true at global scale?

This is a physical limitation to NO_3NH_4 formation and the sensitivity to HNO_3(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 NH3 are too strong in Europe when considering NH3+NH4. 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 NH3(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 SO4= bound the NH4+ is accounted for in EQSAM4Clim therefore the excess in SO4= shown in the new Fig. 2 is the form of non-bound SO4=.

- 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 µg/m3

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, SO_2, NO_2 and VOC as integrated into the anthropogenic global emission inventories, but we do not have direct emission of either $SO_4^=$ or HNO_3 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 SO_2 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.