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 NH_3(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 EGUsphere.

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. SO_2(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 (SO_2) and agriculture (NH_3) 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 SO_2 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 SO_2"

- 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: "SO_2 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 SO_2 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 [SO_2(g)] is critically dependent on emission fluxes applied (no chemical production term is included in the chemical scheme for SO_2). 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 SO_2 and SO_4^{2-} are displayed side by side. We now change the figure legend to : "A comparison of weekly mean SO_4^{2-} for Europe (top panel; μg/m^3) 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.