Response to Topical Editor

We thank the topical editor for conducting a review of the revised manuscript and making some clear suggestions regarding the minor revision that must be made before publication. Here we respond to these points below;

Regarding the title of your article: Even though S. Remy used the terms 'ECMWF IFS-COMPO 49R1' in the title of his 2024 GMD publication, the title of your article should be modified to improve clarity. I have the following suggestion, though you may have another one:
"An evaluation of the regional distribution and wet deposition of secondary inorganic aerosols and their gaseous precursors in IFS-COMPO preparatory to cycle 49R1"

We have now changed the title to that given by the Topical Editor. However, we do note that GMD has a strict policy with respect to model versions and this introduces some inconsistency between studies performed with the same model version.

The following renaming of the simulations should be applied throughout your article, as suggested below: Cy48r1 : unchanged, Cy49r1: Cy49r1compo and Cy49r1 NOE4C: Cy49r1compo NOE4C

We have retained Cy48r1 and changed the nomenclature of the other two simulations to pre-CY49r1 and pre-CY49r1_NOE4C, which we find matches the change made to the title of the manuscript. This allows the manuscript to be found when searching whilst indicating that this is not the final version of Cy49r1, but a version of Cy49r1 in a preparatory phase.

in the abstract: change " The application of the EQSAM4Clim simplified thermodynamic module in IFS-COMPO cycle 49R1" into " The implementation of the EQSAM4Clim simplified thermodynamic module in IFS-COMPO, for use in cycle 49R1"

The abstract has now been changed as requested.

In Section 2, the introductory paragraph has been revised in the updated version of your article, partly reflecting the fact that cycle 49r1 is now operational. However, the text still contains some unclear sections. I recommend rewriting it, particularly to avoid conflating the description of the IFS-COMPO system with that of the specific simulations you per- formed. Also, R´emy et al., 2022 and Williams et al., 2022 analyse Cy47r1.

We now adopt the amended text provided by the topical Editor and remove the references.

Specific comments:

Is it correct that you present results for 2018, while Remy et al. 2024 present results for 2019?
you indicate that in your simulations meteorology is "initialised every 24 hours based on ERA5 reanalysis data". Is it the same protocol in Remy et al. 2024?

Yes this is correct we now include the sentences in Sect. 2.2: "These three simulations use a configuration and emissions as those used for simulations presented in Remy et al., (2024) for

evaluating PM. We select the year 2018 to provide further evaluation which is complimentary to the results presented for 2019 in Remy et al., 2024).

you indicate that in your simulations meteorology is "initialised every 24 hours based on ERA5 reanalysis data". Is it the same protocol in Remy et al. 2024?

The 2018 simulations have exactly the same model set up as in Remy et al. 2024.

what is the reference for the climatology of DMS emissions you used?

We now introduce references for both SO2 outgassing and DMS oceanic emissions in Sect. 2.2.

please detail how biomass burning emissions and SO2 emissions are applied.

We now include the following sentence : "For biomass burning and SO2 emissions vertical profiles are used representing pyrogenic convection or industrial stack heights, with other emissions applied at the lowest model levels. A diurnal cycle is imposed for isoprene and biomass-burning emissions to capture either photolytic activity or the tropical burning cycle".

two additional remarks/questions about your simulations: please indicate whether meteorology can differ across simulations, depending on whether aerosol–radiation interactions are activated or not. And what about the gazeous chemistry scheme, is it identical across all your simulations?

We feel we already address this with the following sentences in Sent 2.2 of the revised manuscript : "The meteorological component is the same between simulations and corresponds to Cy48r1." and "Meteorology is initialized every 24 hours based on ERA-5 reanalysis data i.e. IFS-COMPO is run in cyclic forecast mode". Regarding chemistry : "A 15-minute chemical time-step is used for solving a modified version of CB05 tropospheric chemistry (Williams et al., 2022)."

line 18 : please write in full "IFS"

Now expanded in the abstract.

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."

The sentence has now been updated.

Line 52 : change "The SIA occurs" into "Secondary inorganic aerosols (SIA) are found throughout the troposphere."

Now changed to give the correct definition of the acronym.

Line 53: change "and the concentrations" into "and concentrations"

We have now modified this sentence.

line 54: no capital letter to sulphur dioxide, ammonia, nitric acid, particulate matter . And add a ”,” before ammnonia.

We have now removed all capitals before the definition of the chemical species and the grammar has been corrected.

line 55: please refer to the exact definitions of PMP1.0, etc... particles smaller than ..

We have now changed this sentence to : High concentrations of SIA contribute to total Particulate Matter concentrations that are smaller than various predefined sizes : namely 1.0µm (PM1.0), 2.5µm (PM2.5) and 10µm (PM10).

line 62: in the form ”OF” NO..

We have now corrected the grammar.

line 65: change ” SIA being from NH4NO3” in ” SIA consists of NH4NO3”

We have now corrected the text.

line 66: please reformulate ’increased meteorological instability’

We now change to “ … due to the increased instability of NH4NO3 due to variations in RH and temperature … “

line 68: most SIA: consider it as singular

We have now changed the tense and checked the manuscript with a grammar checker

line 68: ”At high RH values”: please specify a threshold

Now changed to “At high RH values of 80-100% … “

line 70: change ”the optical properties” into ”its optical properties”

Now changed as suggested.

line 71: change ’ and interactions’ into ’and its interactions”

The microsoft grammar tool shows the proposed change would not improve the grammer

line 73: change ”-1” into ”1”

This is not the value given in the publication which is referenced, where a range of pH values is given as we have written in the manuscript.

line 83: change ’whose cumulative oxidation rate is dependent on the prescribed pH in solution’ into ’whose overall oxidation rate varies according to the specified pH of the solution.

We have now replaced “cumulative” with “overall”.

line 85: ”for the determination of trends”: please be more precise

We have now expanded this to : “.. determination of long-term trends with respect to resident concentrations … “

line 89: change ’solution pH’ into ’the pH of the solution.

We have now updated this throughout the manuscript.

line 93: please reformulate to clarify the sentence: ’The more buffering of solution by NH3, the faster the conversion rate as dictated by the reaction of HSO3- being less than that for SO3 = ’

We have now rephrased to : “The more buffering of solution due to the scavenging and dissolution of NH3, the faster conversion rate is dictated by the rate of reaction of HSO3- being less than that for SO3=”.

line 104: please move the reference Peuch et al 2022 after ”CAMS”. Similarly, William et al 2022 does not refer to IFS-COMPO. Please reformulate.

We have now moved/ removed these specific references.

line 110: change ”is the reduction of biases and increase in correlation for aerosol products” into ’was the reduction of biases and increase in correlation of aerosol products with observations’

We have now changed this sentence in line with the direction of the Topical Editor.

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. ’

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.”

line 118: please change ’ This work is complementary to the recent evaluation of the performance of IFS-COMPO Cy48r1 and Cy49r1 and of the impact of using EQSAM4Clim with respect to regional PM2.5 distributions and Aerosol Optical Depth presented in R´emy et al (2024) into ’This work is both parallel and complementary to the recent assessment of global and regional PM2.5 and Aerosol Optical Depth (AOD) distributions presented in R´emy et al (2024).’

We have now modified this sentence as suggested.

line 121: please clarify what you mean by ’upgrades to EQSAM4Clim’

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.

line 124 "we provide details of the changes": which changes are you referring to?

The changes on which the manuscript is based i.e. the modifications to CY48r1 -> CY49r1.

line 125: please rewrite: 'in Sect. 5, we compare annual mean wet deposition fluxes over Europe, the U.S., and Southeast Asia using model outputs and observations...'

We have modified the sentence accordingly and changed South East to Southeast throughout the manuscript.

line 129 : change "Model description of IFS-COMPO versions" into "General information on the most recent IFS-COMPO versions"

We now change the title of Sect. 2 as requested.

I will stop here with my suggestions for textual revisions. I note that in general the readability of the article could be greatly improved by enhancing the fluency of the English. Numerous tools are available for this purpose. . . Thank you for making all efforts in that direction. For some parts of the text, please rely on the formulations used by Remy, GMD 2024, which appear very clear.

As suggested by the Topical Editor, we have used Microsoft grammar and fluency checker and corrected whenever there were any issues detected such that we view this as an independent arbitrator which avoids focusing on differences in writing style. We have implemented nearly all editorial comments on wording and phrasing (e.g., lowercase of chemical species, reformulated unclear expressions, redefined regional descriptions and checked the values of the comparisons.
Redundant or ambiguous phrases have been rewritten, including the improved articulation of simulation assumptions (e.g., consistent meteorology, use of aerosol-radiation interactions, and chemical schemes). We ensured that the paper adheres to the requested terminological consistency and enhanced readability. This has resulted in substantial modifications to the text throughout the manuscript. We refrain from the use of e.g. chatGPT for the reformation of our manuscript.

Response to Referee #1

We thank Referee #1 for the positive evaluation of our revised manuscript and for the detailed suggestions to further improve the clarity and quality of the figures and captions. We will submit amended figures for the final version.

Response to Referee #2

We thank Referee #2 for the further review of our manuscript and the constructive suggestions.

The changes of the wet deposition scheme and analysis. You indicate that the companion paper already describe the changes made to the code. Nevertheless I consider this in not precise enough.

We have essentially re-written the entire content concerning the update of the wet deposition scheme in Remy et al (2024) in the manuscript such that no further content is available as the entire update has been included. Although changes to e.g. the lifetime of BC particles have no relevance to this

study, and we do not wish to give a historical narrative on the description of aerosol processes in previous IFS cycles, we do now expand on the description of changes made to the other aerosol species, vis;

IFS-COMPO Prep-cy49r1 is built on the previous operational cycle (Cy48r1) and contains 8 distinct aerosol types with multiple bins for size segregation, namely sea salt, desert dust, organic carbon, black carbon, $SO_4^{=}$, fine and coarse NO_3^{-}, NH_4^{+} and Secondary Organic Aerosol. For Prep-cy49r1 updates have been made to the aerosol component of the model in the form of modifying both the description and properties of desert dust and sea-salt, the aging of carbonaceous aerosol and an update to the aerosol optics by changing the assumptions used for the $SO_4^{=}$ aerosol when referencing the lookup table for Mie scattering. This impacts the resident lifetimes, radiative properties and the long-range transport component for each of the aerosol species. The modifications to the description of the aerosol optics has also been implemented, which has been shown to improve the simulation with respect to the Aerosol Optical Depth (AOD) and Ångström exponent when compared against regional observations (Rémy et al., 2024). The gas-phase chemistry, photolysis and dry deposition are identical to that used in Rémy et al. (2024) and as described in Williams et al. (2022). For brevity, and that this study is only concerned with soluble aerosols, we refer the reader to Rémy et al. (2024) for more explicit details related to the other aerosol types.

We do not fully understand the comment related to the analysis as the components on the analysis of precursors, particulates and wet deposition in our paper are described in full in e.g. Sect. 3. We do not analyse AOD here and perform a different analysis as that presented in Rémy et al. (2024) . In response to the first round of reviews we have replotted most figures to aid clarify and homogenize the style across species and measurements.

The naming of the code version (cy48 vs cy49). I understand your point, even if you do not treat the possible interactions between the composition compartment and the other changes in the IFS model between CY48 and Cy49. Also, I would suggest to at least modulate the title to suggest the ambiguity. Also I would recommand to be more precise for future publications. Concerning the use of Cy49r1 in the present publication I will leave it to the editor to decide.

We agree that there is no present convention for referring to the version of IFS-COMPO which should be changed for future publications, even though we now change the naming convention in this paper. We also note that we are somewhat going against GMD policy where the naming of model versions should be clear and well defined across publications. Please see the response to the Topical Editor when we have both modified the title and changed the nomenclature of some of the simulations.

Did you check conservativity in the budget tables?

Yes the global budget terms are closed and comparable across runs.

Simulation Cy49r1_NOE4C usage is quite under, it might be useful to exploit it more to be able to make the distinction between different impacts, or to remove it completely.

Regarding the use of Cy49r1_NOE4C, we agree that this simulation could theoretically be used more extensively to dissect the individual contributions from EQSAM4Clim versus other updates. However, as noted, such a configuration will not be used for any forecasting or in a research context. We therefore chose to limit its use to key comparisons that isolate the effect of EQSAM4Clim and

believe this is sufficient for clarity without overcomplicating the narrative. We do however use the comparisons to differentiate that the precursor gases and SIA types are affected differently when using the EQSAM4Clim methodology. For NHx and NOx SIA we have shown strong experimental evidence that without accounting for the chemical content of aerosols, correct particle concentrations are difficult to achieve.

Is there a signification in the colours of the modified manuscript?

There is no specific significance associated with the colour highlighting in the tracked-changes version , where it is purely an artefact of the document editing software used and does not convey any information beyond standard editorial marking.

Line 349: "No filtering has been applied to these measurements" Does it mean you applied filtering to the other datasets?

No filtering has been used for any of the datasets for the comparisons shown. We now include this explicit sentence: "No filtering has been applied to any of the observational data used in this study"

Line 443: "The global budget" You forgot to introduce Table 2.

We now change the sentence to : "Table 2 provides the global budget terms for SO_2(g), which shows that in addition to primary emission, approximately one third of SO_2 in the troposphere comes from the oxidation of DMS by the hydroxyl radical (OH), with DMS originating from biogenic activity in the oceans.

Line 793: "Europe, US and SE Asia"

We now replace "SE Asia" with "southeast Asia" throughout the manuscript as requested by the topical editor.

Lines 995+ and 1080+ are developing the same point.

We have now made significant changes to the text to remove this.

Line 1095: Maybe also say that AMON data are used for top right panel.

We now amend the figure legend to mention the use of AMoN data for the top right panel

Line 1107: significant decreases → Significant decrease

We have now amended this sentence.

Line 1121: "Unfortunately the lack […] can be shown." maybe you should add a paragraph on how selected stations or data, because on the EBAS website when selecting 2018 and ammonium data from the EMEP "Framework" there is a station in South France at Pic du Midi which has data for the aerosol matrix and a few stations including Cyprus and Italy in the pm10 matrix as shown in figure below.

We thank the referee for the link and example. As noted in our first-round responses, we chose to use a consistent set of stations across all species to enable direct interspecies comparison. Adding stations selectively for individual compounds would complicate this consistency. This is now explicitly stated in Section 3, where we have added the sentence : “We chose sites which monitor both pre-cursor gases and associated SIA simultaneously to ensure valid comparisons.”