Reply to Referee 4

Paper: **Operational and Probabilistic Evaluation of AQMEII-4 Regional Scale Ozone Dry Deposition. Time to Harmonise Our LULC Masks**, by Ioannis Kioutsioukis et al., 2025, ACP

We are grateful to the reviewer for the thorough analysis of the manuscript and the careful reading and suggestions. They all greatly improved its quality.

Reply to the specific comments:

While the manuscript presents a detailed and robust analysis highlighting the inconsistencies in the LULC masks and their implications for ozone dry deposition modelling, it has become extremely long and dense. Several sections, particularly operational evaluation, are notably verbose, with extensive model-by-model commentary that could be streamlined. I recommend that authors condense or relocate some of these detailed discussions to the supplements, especially where the text reiterates statistical patterns already evident in the figures. Additionally, the manuscript would benefit from clearer synthesis or summary paragraphs at the end of each major section to reinforce the key takeaways and guide the reader through the analysis.

We have condensed slightly the section following the reviewer suggestions; however, we consider the details in it important for the scope of the paper as they are there to document the model performances that are needed not only of the current study but also all the others present in the special issue. Following a suggestion of Reviewer 1 the section 'operational evaluation' has been broken up in subsections. Thank you for your comment.

Line 83-88: What are the emission inventories used for lightning NOx, forest fires, biogenic emissions, and other natural sources? Please provide a brief discussion on the internal model processing of these emissions.

As explained in the introduction all the information relating to the case studies and the common input to all models have been summarized in the technical note Galmarini et al. (2021). This editorial choice offers a one stop-shop for all necessary information and avoids repeating it in the various contributions to the special issues, which would take away space for actual research items. Briefly, the lightning NOx as well as forest fire emissions were harmonized across all models with more details provided in Galmarini et al. (2021) while the representation of biogenic and other natural emissions was decided by each modelling group. The following sentences have been added fo further clarity: 'Details on the choice of years, model resolution and domains, model versions and choice of emissions input data may be found in Galmarini et al (2021). Details on the model deposition parameterizations for ozone are found in Clifton et al (2024), and a

discussion on the details of deposition for acidifying species can be found in Makar et al. (2025).'

Line 89-90: Please provide a reasoning behind choosing the spatial and temporal domains for this study. What are the spatial and temporal resolutions used in the model runs?

This information is contained in Galmarini et al. (2021).

Line 136-139: Ozone values in the figures are reported in ppb over North America, while in µg/m3 over Europe. The use of these two different units for ozone concentration complicates the direct comparison. While the authors attempted to align the color scales, I recommend standardizing the units across both regions for the convenience of readers and easier comparisons.

Indeed, we could convert them, at the same time only the macro differences between the two continental air sheds modelled are compared, whilst for the details the two cases stand alone. Furthermore, the measurements are provided with these units. According to us, sticking to the original units is the best way to preserve the integrity of data that are not under our direct control. Finally, the conversion from ppb to ug/m3 for O3 and NO2 is approx. 2 and for NO 1.25 (at 25 deg C) which are manageable conversion factors in case anyone would be interested in a detailed comparison. It should also be considered, as explained in the paper that: ' … since ozone values are reported in ppb over NA and ug/m3 over EU, the range of the colour scales over both continents has been set such that the same colours represent the same absolute errors (note the difference in the numerical values for the colour bars for these figures), to account for unit differences and allow for a visual comparison between continents.' *An explanatory sentence of this choice has been added to the text in section 2.*

Line 161: Figure 4 presents results for Europe, which is incorrectly referred to as showing results for North America. I recommend that authors carefully review the entire manuscript to identify and correct such figure and section reference inconsistencies. While minor, these errors can significantly impact the clarity and interpretation of the results and may confuse readers.

Thank you, we noticed this too and corrected it accordingly.

Line 190-194: The authors hypothesize that the relatively minor difference between WRF-Chem (UPM) and WRF-Chem (UCAR) is primarily due to the difference in gas-phase chemistry mechanisms. However, this claim is made without presenting supporting analysis or citing a reference that explicitly evaluated the gas-phase chemical mechanisms used in these two configurations. I recommend that the authors provide a reference to a past work supporting this hypothesis or rephrase this discussion as a hypothesis.

A good point – we should have included a reference in the revised manuscript (Knote et al., 2015). The impact of the two mechanisms on model performance was evaluated in a previous AQMEII ensemble cycle (AQMEII2) by Knote et al., Atm. Env., 115, 553-568, 2015. Figure 3 of this paper shows that the use of MOZART4 versus CBMZ resulted in opposite signs and magnitudes for O3 biases in North America. We've broken the original sentence into two parts, and have inserted the following sentence between them: "Knote et al. (2015) conducted a comparison of the two gas-phase mechanisms (CBMZ and MOZART4) within the same modelling framework, and showed that two mechanisms to have biases opposing in both magnitude and sign over North America)."

Line 308-311: What are the factors driving the underestimation of wintertime NOx?

While we can't provide a certain answer, we can speculate – we have included the following sentence in the revised manuscript:

"Potential factors which might drive an underestimate of wintertime NOx include underestimates in the emissions of NOx from combustion sources such as wintertime home heating from fossil or wood fuels (van der Gon et al., 2015), underestimates of atmospheric stability (i.e. if the simulated atmosphere is more unstable than the actual atmosphere, NOx emissions may build up to higher concentrations in the model than is observed), and the potential for HONO cycling in the presence of snow on surface leading to longer lifetimes of NOx (Michaud et al., 2015)."

Line 390-393: Are there any implications of combining LCAN and soil for models that distinguish these two terms?

The reviewer has raised a good question. The LCAN term is relatively small - it is sufficiently small that some deposition algorithms leave it out altogether (Clifton et al., 2024) - hence its inclusion with the ground resistance is unlikely to influence O3 deposition significantly, and the ground resistance term is unlikely to be significantly influenced by being combined with the LCAN term.

Line 529-534: The authors identify factors that are expected to be relevant in the determination of ozone concentration variability at the surface but fail to discuss the methodology used in identifying these factors. I recommend that authors briefly discuss the methods used to identify these factors, either in the main text or the supplement.

As a matter of fact, the methodology is fully explained also in mathematical terms in the paper, so it is hard to understand what specifically the reviewer is referring too. The same set of four variables has been selected simply to represent the core mechanisms of O3 fate in the atmosphere (convection, advection, dry deposition, photochemistry) in different domains, months and LULC classes.