Review 1 reply

The manuscript by Wilson and Prather describes a quasi-three-dimensional modeling approach and employs the model to quantify the global net O3 production and CH4 loss from South Korean air pollution over 45 days in May-June 2016 during the KORUS-AQ campaign. The model consists of three stages: (1) a boundary layer-residual layer (BL-RL) stage, (2) an isolated pollution plume integration (PL) stage, and (3) a dispersed pollution (DP) stage. Taking a test case with South Korean air pollutant emissions compared to a control case with no anthropogenic and biomass burning emissions, the relative global CH4 loss and O3 production due to South Korean air pollution is diagnosed over the three model stages. Global modeling studies have typically diagnosed O3 production and loss from reaction rates for the odd-oxygen family. Here, the model developed by the authors explicitly resolves O3 perturbation lifetimes to more accurately calculate the O3 production term. This study offers a useful demonstration of this alternative approach to deriving O3 budget diagnostics. Further, the sensitivity of O3 and CH4 reactivity to plume aging times is assessed, providing valuable insight on the potential degree of overestimation of the anthropogenic contribution to global O3 and CH4 budgets resulting from rapid dispersion of air pollution in course resolution models.

I find that the manuscript is well written overall, provides a thorough detailing of the model, and fits well within the scope of ACP. I recommend publication of the manuscript after the authors address the following comments.

Thank you for the time you have invested into reviewing and improving the manuscript on our behalf.

Comments:

Can the manuscript title be made more specific to say “Impacts of South Korean Air Pollution”? Is there a reason why this is unspecified?

This is a good suggestion and will be helpful to readers who are researching the South Korean region. We have also made the title more specific to our work following the advice of Reviewer 2. The new title is:
‘***Life–Cycle Impacts of South Korean Air Pollution on Tropospheric Ozone and Methane: Sensitivity to Dispersion Time***’

While the motivation for studying the KORUS-AQ period is made clear in the introduction, the motivation behind developing the new modeling approach (to explicitly calculate O3 perturbation lifetime to correctly quantify the O3 production term) is not introduced until

Section 3. Overall clarity would be strengthened by moving the background discussed in lines 265-272 to the main introduction, as this is a key motivation and focus of the conclusions.

We agree with this recommendation and have reworked the introduction with the inclusion of this paragraph (lines 28 to 55 in the new revision).

The authors should choose consistent language of either BL-RL/PL/DP "stage" or "model" for improved clarity.

This is an important detail: while the three modelling stages could be considered as separate models, the results are incomplete without all three stages. We now refer to them consistently as 'stages' to reinforce this for the reader.

Can the authors elaborate on the choice to assess the impact of biomass burning emissions and anthropogenic emissions simultaneously? If computationally feasible/reasonable, it would be highly valuable to attribute O3 and CH4 changes to anthropogenic and biomass burning emissions separately (e.g., perform another case with only GFED and MEGAN emissions and difference from full emission test case).

The GFED5 inventories are for all open burning and include anthropogenic emissions from agricultural waste burning. As part of the anthropogenic emissions these must be included to supplement the KORUS v5 inventories that include 'area, point, mobile, and ship emissions' (Park *et al*., 2021). The GFED5 emissions were intermittent throughout the month and contributed relatively little to total emissions. We have included the clarification: 'We consider the GFED5 inventories as part of the anthropogenic perturbation alongside KORUS v5, assuming the emissions are predominantly from agricultural waste burning.' (lines 97 to 98). We have also added a reference to Sindelarova *et al*. (2014), who are credited with creating the MEGAN-MACC inventories.

Line 433 and 532: There are a few places in the manuscript where biomass burning emissions from GFED are implied to be anthropogenic. It is unclear to me if this is a wording issue or if it is the case that most of the biomass burning emissions from South Korea are from anthropogenic sectors. Unless the latter is true, the authors should be careful not to label GFED as anthropogenic.

Research on South Korean biomass burning centers on agricultural waste burning, e.g., Ban *et al*. (2024); Ryu *et al.* (2006). According to Ryu *et al.* (2004), 'Biomass burning is a widespread, annually occurring phenomenon that is used to clear land for cultivation, convert forests to agricultural lands, and remove vegetation to promote agricultural productivity and growth of high-yield grasses.’ Thus, we assume GFED5 emissions in South Korea are predominantly anthropogenic.

Section 2.1: Have the authors quantified the total GFED and MEGAN emissions that are cut off at the coastlines? How sensitive are the results to the inclusion of the full emissions from those grid cells?

GFED5 emissions were not cut off. MEGAN emissions were cut-off, and we lost about 7% of total isoprene and 10% of total ethene emissions. The largest losses were in some East coast cells (up to 20% loss). The major coastal cities such as Seoul, Busan, and Ulsan were fortuitously placed within uncurtailed MEGAN-MACC grid-cells. Most of the missing isoprene were in remote cells without large anthropogenic emissions. In hindsight, we could have found a way to include all biogenics, but believe this would have negligible impact on our life-cycle study.

Section 2.2: Since Section 2 is said to describe the “data and tools” (line 46) used in the study, the authors might consider titling Section 2.2 as “In-situ observations” rather than “Atmospheric composition”. Then, the subheadings 2.2.1-2.2.3 could be labeled more broadly as e.g. “KORUS-AQ airborne observations”, “ATom airborne observations”, and “NIER surface pollution maps”. This would provide a centric section to list all variables and their uses from each in-situ dataset (not just those used for deriving atmospheric composition). For example, J-values and LAS measurements are used from the KORUS-AQ mission for F0AM but are not mentioned until sections 2.3.2 and 2.3.3. The authors could provide a brief paragraph or sentence describing these variables in Section 2.2.1 and cite the respective photochemical box model sections where more detail will be provided.

We thank the reviewer for these logical suggestions, and they have been implemented. Our new headings indicate what the tools are, then we detail how we use them, as per your recommendations. We include the note: ‘We also use KORUS-AQ observations of photolysis frequencies (J-values) and aerosol surface area densities in our chemistry modelling and describe these data and their usage in Sects. 2.3.2 and 2.3.3.’ (Lines 121 to 122).

The Section 3 title “Models” is quite vague and is not specifically representative of its subsections.

We have amended the section title to ‘Modelling system’

Figure S8: Would the authors consider moving this figure to the main text? It is a very useful visualization of the modeling process and timeline. There are currently many tables in the main text, so I think this could be swapped with one if concerned for space.

We have promoted the timeline to Figure 9 in the main text.

Technical corrections:

Line 126: The section titled “Surface pollution maps” is labeled 2.2.2 but should be section 2.2.3.
Line 141: In the title of Section 2.3.1, the “M” in mechanism should be lower case.
Line 160: “For” should be “far”.
Thank you for finding these; we have made the corrections.

Figure 4: First, the panels are listed as (a-c) in the figure caption, but the figure panels are not labeled with letters. Second, the order that the NOx and O3 panels are presented is flipped in the figure caption. There is an inconsistent use of labeling multi-panel figures alphabetically across the manuscript.

We agree and have simplified the panel designations to be positional labels (left, middle, right).

Figure 1 and Figure 4: Can the panel labels (NOx, CO, Cover class, NO2, and O3) either be made bigger or placed as titles?

We have made the title fonts larger.

Review 2 reply

The study frames an important question in respect of chemistry vs dilution in ozone production and methane loss on country scale outflows. The modelling approach which is explained in detail, allows separation of emissions, chemical transport and dilution and looks like a powerful tool to assess country-scale export. The study is clearly set-out and rigorous.

We appreciate the time you have taken to review and improve our manuscript on our behalf.

If I where to be critical, I would have loved to see the counterfactual and what would have happened if the more usual approach was applied.

Comparing our budgets to the more standard approach with a regular global chemistry-transport model is an important, but next, step. With the CTM we have to assess how the horizontal resolution would alter the life-cycle calculations and we would further like to design a test with variable horizontal resolution, although we could probably do no better than 100 km *vs*. the 10 km here. The other elephant in the room is how to implement full MCM v3.3.1 chemistry (5,000 reactions and 1,610 species) in a CTM without crippling the computation. Even the most extensive CTMs do not implement the full chemistry. Thus, the comparison with a CTM would need to use both models under different parametric circumstances. Consequently, we feel a CTM study would provide more value as a separate paper.

Probably worth in introduction presenting the difference to the usual way models of this type would represent the dynamics and chemistry. This occurs later on (Section 3 – models) but we would benefit from a better framing in the introduction.

This is an excellent point, and we have reworked the introduction to include this discussion (lines 28 to 55).

In the chemical modelling, it would be good to know the nature of the spurious results, that required the 1st order loss of the peroxide and HNO3. Where they any measurements of NOy or H2O2 to support these losses?

A first-order loss is required to avoid unrealistic accumulation of gas-phase HNO_3 and H_2O_2, since these are out of balance in the free troposphere without these processes. Likewise, the emissions of NO_x and other species are necessary to maintain the free tropospheric composition seen in ATom. We do not see how any observations support the losses, except by the imbalance found in the ATom chemistry (*i.e.*, some species accumulate over 24 hours and others are depleted). Real wet deposition of HNO_3 and H_2O_2 (and injection of NO_x) would have spatial and temporal variability that we cannot model here.

Did the approach lead to any mass conservation issues with the ghost cells?

We were concerned with this potential problem and accounted for every mole of air departing from a given terrestrial BL (RL) and placed them into either another terrestrial BL (RL) or a BL (RL) ghost-cell.

Is it possible to make an assessment of the uncertainties of the modelling approach? The overall budget figures lack uncertainties.

We could propagate the chemical parameter uncertainties (rate coefficients, cross sections, …) but this would be a separate paper, and it would probably not address the primary structural uncertainty – the model itself. We do not know how to address this without more extensive parallel comparisons with CTMs for example. This is beyond the scope of this paper.

We did however address the primary uncertainty in our new model, *viz.*, how the budget numbers change with the lifetime of the pollution plumes in the free troposphere. This we believe is a new result here.

Given the nature of the conclusions, how should these results/methods be incorporated into the more classical approach outlined in e.g. Griffiths et al (2021).

The question raised here is how would *e.g.* the next-gen Griffiths' CMIP paper on O_3 use or refer to this work. Our model cannot be used directly for tropospheric ozone trends as it would be nigh impossible to simply scale the result to multi-year global emissions. Our results could, however, provide an alternative calculation for the O_3 and CH_4 budgets if the atmospheric chemistry MIP set up a KORUS-like experiment for air quality. The MIP could also specify CTM diagnostics to calculate the pollution-driven budgets for O_3 and CH_4. Defining how to calculate such budgets with current CTMs could be a useful joint project, or one that we could pursue with joint hybrid and CTM modelling.

We have added to the conclusion: 'Atmospheric chemistry MIPs could incorporate our results by performing a regional air quality CTM study, e.g. for South Korea, and comparing the resulting CH_4-OH loss and O_3 mass perturbations. Diagnosing O_3 production from the mass perturbations, as done in this work using lifetimes, remains a challenge.'

The authors have produced an intriguing piece of work, where all the assumptions and the model are well laid out. The authors should be complimented on a nice piece of work that has a clear impact on our understanding of tropospheric ozone and methane.

Thank you once again for your kind words and for helping us improve the paper.

Minor comments

Title is too general and needs to tell the reader why they should read this.

We have changed the title to: '***Life–Cycle Impacts of South Korean Air Pollution on Tropospheric Ozone and Methane: Sensitivity to Dispersion Time***'.

Abstract – given the nature of the scaling (discussed in conclusion) worth adding "Simplistic scaling …. "

The sentence now begins with 'A simplistic scaling of…'.

Intro – Need to give a better explanation of Zhao et al (2025), not clear the point you are making.

We realized that the Zhao work (global CTM perturbations) does not really align with the model experiments here and is not comparable, so the reference/discussion has been dropped.

Section 3 – needs a better title.

We have changed the section title to ‘Modelling system’.

Line 285 – is the assumption first-order or pseudo first-order?

Calling the assumption ‘pseudo first-order’ might mislead the reader into expecting a single first-order mode of decay from a given pollutant perturbation, and thus we have written: ‘we assume that pollutants are dilute enough that the chemical response is first-order with respect to the number of moles of dispersing pollutant…’ (lines 281)

Line 309 – what does “A careful budget is kept of chemical species entering the model domain via the FT” mean?

We agree this sentence adds unnecessary confusion. The intent was that the pollutant moles entering the model from the background FT (boundary condition) are tracked and diagnosed. Since this contribution is identical in the standard model run and the control run, the diagnostic is not relevant to the budget calculation, so the sentence has been removed.