

Review of Jeong et al.,

Original comments are in *gray italics*, author responses are in *blue italics*, additional comments are in black.

Comment #1

Supp. Line 38 – Please clarify why dry surface area was used to calculate $\gamma(\text{N}_2\text{O}_5)$. This assumption will artificially increase the N_2O_5 rate constant as $\gamma(\text{N}_2\text{O}_5)$ is typically calculated using the wet aerosol surface area.

In our study, dry surface area from observations were not used in deriving the $\gamma(\text{N}_2\text{O}_5)$. As described in the paper, it was derived based on the Bertram and Thornton (2009) study where they used an empirical pre-factor in the equation. Other parameters used in deriving γ are described in the Supporting Information section 3. The estimation by Bertram and Thornton (2009) can be an overestimation as mentioned in the first round of reviewer comments. However, the purpose of the box model simulations was to make sense whether the observed ClNO_2 levels were in the range that could be explained by in-situ chemistry. The box model results significantly underestimated the observations (~50 times), therefore leading to a conclusion that the second ClNO_2 peak in the morning is likely not from in-situ production.

I misspoke in my original question, I meant to ask why the authors had used dry aerosol SA in the calculation of the 1st order loss rate coefficient of N_2O_5 , not $\gamma(\text{N}_2\text{O}_5)$. The authors state on line 39 in the supplement that they do not consider aerosol hygroscopic growth when they calculate the 1st order loss rate coefficient for N_2O_5 ($k = 0.25 \cdot c \cdot \text{SA} \cdot \gamma$). Using dry SA will result in a smaller loss rate constant and could lead to an under-prediction in ClNO_2 production (as is observed by the model). Please clarify why you did not use hygroscopic growth in the calculation of $k_{\text{N}_2\text{O}_5}$ as is stated on line 39 in the supplemental and what implications this may have on your results.

Comment #2

Section 2.3 – While the authors have provided many details in their update, it remains unclear exactly how the simulations were setup, how many different types of simulations were run (and what the similarities and differences were), as well as important missing details about the model treatment of boundary layer dynamics and deposition.

First, the authors need to state explicitly in the Section 2.3 of the main text what the duration of each model simulation was (i.e., 72 hours?) and the frequency at which the model was constrained with observations (i.e. every 100 minutes?). Currently, the supplement indicates a discrepancy where it states that the model duration is 1 day. However, it becomes apparent later in the main text that (at least) two different types of simulations were conducted (both 72 hour and 1 day simulations). The setup details (e.g., duration, initialization time, constraint frequency, any chemical/physical differences, number of simulations, etc.) are needed for each type of simulation in order to avoid confusion later. For instance, on Line 219, it is unclear what the 'end points' of 72-hour simulations are. Are these final mixing ratios at the end of different simulations? What time of day are the simulations set to end? How many individual simulations were run and what were the differences between them? At the moment, Section 2.3 reads as if there was only one type of simulation used.

Second, the authors need to include additional information about how boundary layer dynamics are treated in both the 72 and 24-hour simulations. For instance, how does the model treat the separation of the surface and residual layer at night and the entrainment of O₃ in the morning? Does the model include deposition? These details are missing from both the main text and the supplement and are important for the accurate simulation of O₃ production.

Third, on line 34 in the supplement, it is unclear what the authors mean by 'all the measured parameters'. Does this mean that all concentrations are held constant? All rate constants? All photolysis frequencies? Please clarify.

As stated in the first sentence of the 2.3 modeling section, there were 3 types of simulations carried out each represented in Figure 5, 8, and 10.

We revised it as below for clarification:

(Ln 158-160) "We used Framework for 0-D Atmospheric Modeling (FOAM v3.1) for simulating three types of simulations: 1) daytime Cl₂ production (Figure 5), 2) in-situ ClNO₂ production in the morning (Figure 8), and 3) testing the impact of measured ClNO₂ on the regional tropospheric chemistry (Figure 10)."

Thank you, it is now clear that the authors conducted 3 different types of simulations. I will reiterate, however, that it is important to move some of the simulation setup details from the supplement to the main text here. The reader should be able to understand the basic differences between these three simulations without having to refer to the supplement, which is not currently the case.

For instance, the authors added: ***(Line 165-167) "For simulations presented in Figures 5 and 8, a constant meteorology and trace gas observation set, collected at the corresponding time point, were constrained throughout the model run. For Figure 10, the model was constrained with a diurnal variation of the parameters."*** and

(Ln 182-183) "More details on the setup of the box model and the main differences between the three types of simulations are in the supplement material (S3)."

Please add a few more basic simulation details to lines 165-167. For example, change to something like... "For simulations presented in Figures 5, a constant meteorology and trace gas observation set, collected at the corresponding time point, were constrained throughout the 72-hour model simulation. The Cl₂ concentrations at the end of the 72-hour simulation are compared to simultaneously observed mixing ratios of ClNO₂ in Figure 5. Simulations in Figure 8 were similarly constrained as those in Figure 5 but allow ClNO₂ concentrations to vary with time in order to assess ClNO₂ production predicted by the model. For Figure 10, the model was constrained with a diurnal variation of the parameters for the [72-hour?] model simulation. To assess the impact of ClNO₂ chemistry on net O₃ production, all species were constrained except for NO₂ and O₃, which were initialized with observed values and allowed to vary in time"

These are just suggestions, but information like this would give the reader enough to be able to understand how and why the different simulations were conducted. The reader can then go to the supplement if they want more information.

The duration of each of the model simulations are in the supplementary. The supplementary does not indicate that the model duration was 1 day.

I'm still confused about the duration of the 3rd set of simulations (shown in Figure 10). The authors state on line 55 in the supplemental: "The integration time in the model was 100 sec and the model was run for 1 day." Please clarify.

Additional Minor Comments:

Line 71 – The authors use CMAQ here but don't define it until later on line 80.

Line 89 – Remove '...due to prolonged nighttime'.

Line 255 – remove the word 'these'. This makes it sound as though Bassandorj (2018) was studying the same stagnation events in Korea, which is not the case.

Line 256 – It looks like there is an extra part of a sentence included here. Remove the text between 'NO₂' on line 255 and 'However' on line 257.

Line 364 – The authors clearly show in Section 3.1 that removing HCl production reduces Cl₂ production. However, in order to conclude here that ClONO₂ and HOCl uptake are responsible for the positive Cl₂/ClONO₂ correlations and dependency of this correlation on O₃, the authors should show how changes in uptake, not HCl, impact the correlation. While it may be the case that ClONO₂ and HOCl uptake are responsible for this the result, I suggest either a sensitivity test with the ClONO₂ and HOCl uptake coefficients set to 0 or changing this concluding sentence to state that HCl production from VOCs may be responsible for the observed trends (which is the test that was actually conducted).

Line 372 – Change 'it is clear' to 'back-trajectories suggest that air masses were mostly transported from the west ...'.