Response to reviewer comments on "Integration of Airborne and Ground Observations of Nitryl Chloride in the Seoul Metropolitan Area and the Implications on Regional Oxidation Capacity During KORUS-AQ 2016"

We would like to thank the reviewers for their careful consideration of the paper. The comments have been addressed and additional information has been added in the paper for clarification. The comments from the reviewers are in *black*, our responses are in *blue*, and parts that have been added to the manuscript are in ***bold blue***.

Reviewer #1

Jeong and co-authors present an analysis of ClNO2 observations from the 2016 NASA KORUS-AQ campaign. ClNO2, additional trace gases, and measurement of aerosol composition were collected aboard the NASA DC-8 aircraft as well as at two ground-based sites, which were the main focus of this study. The first site was located in the Seoul metropolitan area, while the second was located ~26 km southeast in a forested area. The same observation method was used for all ClNO2 measurements, allowing for direct comparisons between sites. Ground-based observations revealed that ClNO2 was elevated at night at both sites, with higher concentrations at the TRF forested site, which was physically removed from NO emissions, which reduce O3 concentrations and calculated nitrate radical production rates. At both sites, there were periods where ClNO2 persisted at high levels throughout the morning after sunrise. Using additional box-model simulations, the authors found that morning entrainment of ClNO2 from the residual layer, nor morning ClNO2 production could reconcile differences between the simulations and observations. Based on these results and backward air mass trajectories, the authors conclude that horizontal transport is the likely cause of elevated morning ClNO2 concentrations. A final set of 24-hour simulations with and without ClNO2 production, constrained to observations, revealed that chlorine radical initiated chemistry can increase net O3 production in the morning by between 2 and 25%.

The authors have provided a succinct and novel analysis that merits publication after the following comments are addressed. First, this manuscript requires more details in the methods section about the box model set-up and the types of simulations that were conducted. Specific comments are provided below. Without additional details, it is difficult to fully assess the model results presented here. Second, the authors conclude that horizontal transport is the most likely cause of elevated surface-level morning ClNO2, without providing sufficient evidence. The authors present three possible causes based on previous studies and find no evidence for the first two, and therefore conclude that the third, transport, must be the main source. While this may be the actual cause, the authors need to provide additional evidence for this conclusion as discussed in specific comments below. Other comments largely include additional suggested references, requested clarification of the calculated net O3 and NO3 radical production rates and aerosol surface area, and other minor and editorial comments.

Major Comments:

Instrument Section:

Ln 130 – The authors note that ClNO2 is thermally converted to NO at 325 C in a CL instrument. The thermal conversion efficiency of ClNO2, however, is estimated to be between 300 – 500 C, depending on the inlet and heater set-up and flow rate of a particular instrument (e.g. Thaler et al., 2011; Wild et al., 2014; Wooldridge et al., 2010). As the CL instrument was used to calibrate the CIMS ClNO2 measurement, the authors should discuss how they accounted for (or quantified) the thermal conversion efficiency of ClNO2.

The following has been added for clarity:

> (Ln 147-149) **"$ClNO_2$ is detected as NO_y in the CL through conversion to NO on the heated (325 °C) molybdenum catalytic converter (Williams et al., 1998). The efficiency of the conversion was assumed to be unity."**

Model Description Section:

More details are required in this section about the model set-up and the types of simulations that were conducted.

Ln 135 –State which meteorological parameters were used as constraints. Also clarify the type of simulations that were run and how the model was constrained with observations. For example, were simulations run for 24, 48, 76 hours? Were simulations constrained every 10 minutes, 1 hours, 6 hours, etc.? Were different simulations run and constrained to observations from both of the ground sites or was a single simulation run with a combination of the two?

The 2.3 Modeling section was revised as suggested:

> (Ln 155-156) **"Each step of the model was constrained with the averaged meteorology parameters (e.g., pressure, temperature, relative humidity) and trace gases observed…"**

Additional information of the model setup has been included in the supplementary:

> (Supplementary Ln 31-34) **"Daytime steady state Cl_2 simulations (Figure 5) were constrained with meteorology and trace gas observations corresponding to each point of the data shown in Figure 4. The constrained parameters were kept constant throughout the 72 hours of integration time and the end points are shown in Figure 5. Steady state simulations of $ClNO_2$ production in the morning (Figure 8) were ran similarly by holding constant of all the measured parameters throughout the runs, except for $ClNO_2$."**

> (Supplementary Ln 49-55) **"Impact of measured $ClNO_2$ on O_3 production (Figure 10) was explored by constraining the box model with diurnal variation of observations throughout each step. Constraining the model with the diurnal variation of measured $ClNO_2$, allowed the box model to capture its trend throughout the course of the day.**

> **Since our purpose of the simulations were to explore the possible impact of $ClNO_2$ on O_3 production, NO_2 and O_3 were only constrained initially at the first step with observations and then calculated based on the chemistry embedded in the model. More specifically, the initial concentration of each following step was taken from the value in the previous step. The integration time in the model was 100 sec and the model was run for 1 day.”**

Ln 149 – Clarify, did the authors apply a hygroscopic growth factor to the measured aerosol surface area? What was the size range of the aerosol particles that contributed to the measured surface area? Did aircraft vertical profiles show that the aerosol surface area was relatively constant with altitude? As there are no measurements of aerosol surface area on the ground, this source of uncertainty in the model should be discussed. In addition, it would be helpful to put the N2O5 uptake coefficient into context. The authors could cite previous studies that derived uptake coefficients in Asia (e.g. Brown et al., 2016; Tham et al., 2016; Wang, Z. et al., 2017; Wang, X. et al., 2017; Wang, H. et al., 2017). As this manuscript is primarily about ClNO2 production, the authors should also state how the ClNO2 yield was calculated in the model. In the event that the Bertram and Thornton parameterization was used, it is also important to note that this has been shown to be an over- prediction of field-derived yields (McDuffie et al., 2018a; Riedel et al., 2013; Ryder et al., 2015; Tham et al., 2018; Thornton et al., 2010; Wagner et al., 2013; Wang, Z. et al., 2017; Wang, X. et al., 2017).

The following has been added in the supplementary for clarity:

> (Supplementary Ln 34 - 48) “**Heterogeneous reactions of gas-phase N_2O_5 (i.e., $N_2O_{5(g)} + Cl^-_{(aq)} \rightarrow ClNO_{2(g)}$), $ClONO_2$ (i.e., $ClONO_{2(g)} + Cl^-_{(aq)} + H^+_{(aq)} \rightarrow Cl_{2(g)} + HNO_3$), and HOCl (i.e., $HOCl_{(g)} + Cl^-_{(aq)} + H^+_{(aq)} \rightarrow Cl_{2(g)} + H_2O$) were included in the model. For these heterogeneous reactions, a simple first-order reaction was assumed by accounting for γ, Φ, molecular speed of the gases, and surface area of aerosols. Hygroscopic growth factor was not considered in the model. γ_{N2O5} was calculated from the Bertram and Thornton (2009) study using measured inorganic aerosol composition, temperature, and relative humidity and water content derived from the thermodynamic model Extended Aerosol Inorganics Model (E-AIMS, (Clegg et al., 1998; Friese and Ebel, 2010)). The average and median γ_{N2O5} values during the whole campaign were both 0.017. This is in the lower range of what has been derived from previous field observations in Asia that ranges from a campaign average of 0.004 to 0.072 (Yun et al., 2018; Brown et al., 2016; Tham et al., 2016; Wang et al., 2017b, d, a, c). γ values of $ClONO_2$ and HOCl were set to 0.06 (Deiber et al., 2004; Hanson et al., 1994; Hanson and Ravishankara, 1994). The yields (Φ) of the three heterogeneous reactions were assumed to be 1, therefore the steady state simulations would be an upper-limit of Cl_2 or $ClNO_2$ production. Aerosol surface area was taken from airborne measurements of particle size distributions. An averaged value was used from data retrieved below 1 km over the SMA. The airborne data did not show a significant**

> **vertical dependence within the daytime boundary layer. Based on this, an average of 78 ± 41 $\mu m^2 cm^{-3}$ were estimated for particle sizes between 10 nm and 5 μm."**

Ln 150 – Add more details about the FLEXPART simulations. For example, add more details such as those in the Figure 3 caption.

The following has been added for clarity:

> (Ln 173 - 180) **"The FLEXible PARTi-cle dispersion model (FLEXPART v9.1, https://www.flexpart.eu) was used for the air mass source contribution (Figure 3) and backward trajectory analysis (Figure 9). The backward trajectories reported in our study were initialized 9:00 LST at TRF, following it 24 hours back in time. The trajectories were driven by the National Centers for Environmental Prediction (NCEP) Global Forecast System (GFS) with a 0.25 degree resolution. Influence of air mass originating from the ocean at TRF and OP was calculated every 6 hours following an air mass 5 days back in time. Meteorology was driven by WRF with a 5 km horizontal resolution. Since emissions of CO are very low in the ocean, and assumed to be inert in the model, it was used as a tracer for contribution of air originating from the ocean within a given air mass at each ground site."**

Results and Discussion Section:

Ln 161 – The statement that ClNO2 rapidly photolyzes near sunrise contradicts the authors later statements on line 213 that ClNO2 persists after sunrise.

Line 265 has been re-written as below:

> **"At both sites, $ClNO_2$ levels started to increase or sustained after the first 2-3 hours of rapid net loss upon sunrise.. "**

Paragraph starting on line 243 –Is there further information in the flight data to support the hypothesis that boundary layer transport is the main source of elevated surface-level ClNO2? For example, were NOx and O3 observations to the west of the observation sites elevated relative to the east? My concern is that this section reads as though the third proposed possibility must be correct since there was no evidence for the first two possibilities. As written, there is not enough evidence in this section to support the third possibility that transport is the main source of surface-level ClNO2.

Following has been included in the discussion

> (Ln 299-305) **"During KORUS, the DC-8 did not fly to the west of the SMA in the early morning. However, there are large point sources, such as petrochemical facilities and industries, and vehicular emissions to the west and south west of the SMA region. Sullivan et al. (2019) reported that this resulted in enhanced levels of O_3 in**

receptor regions (i.e., Taehwa Research Forest) downwind when westerlies were prevalent. Therefore, favorable conditions such as high chloride content in aerosols from both anthropogenic and natural sources and high levels of NO_x-O_3 could have lead to significant levels of $ClNO_2$ to build up and transported to TRF before being completely photolyzed. During the campaign, influence of large biomass burning was negligible as reported in Tang et al. (2018, 2019).”

Section 3.3.

It is unclear how ClNO2 was used to constrain the model simulations. For example, was the model only initialized with observed ClNO2 mixing ratios, or was the model constrained to observations throughout the morning? As the authors spend time in the previous section discussing the elevated morning ClNO2, it seems important that model simulations emulate that observed behavior.

A more detailed description of the box model runs have been added in the methods and supplementary. The following has been added for clarity:

(Supplementary Ln 49-51) **“Impact of measured $ClNO_2$ on O_3 production (Figure 10) was explored by constraining the box model with diurnal variation of observations throughout each step. Constraining the model with the diurnal variation of measured $ClNO_2$, allowed the box model to capture its trend throughout the course of the day”**

Conclusions:

The authors mention that stagnation events were associated with low ClNO2 production. There was no discussion in the text, however, about the meteorology associated with these low ClNO2 events. Moreover, past studies have shown that certain types of stagnation events can actually enhance N2O5 chemistry (e.g. Baasandorj et al., 2017).

Description on the meteorology during the campaign have been included in the Methods section for clarity:

(Ln 110-115) **“Meteorology during the observation period can be classified into dynamic (May 4th - 16th), stagnation (May 17th - 22nd), transport (May 25th - 31st), and blocking period as shown in Figure 3. During the stagnant period, high pressure system was persistent in the Korean peninsula resulting in local air masses to be more dominant within the SMA compared to the dynamic and transport (May 25nd - 31st) periods. Rex block patterns were observed during the blocking period (June 1st - 6th), which also resulted in more local influence.”**

The following have been included in the discussion for further discussion on stagnation events and N_2O_5:

(Ln 236 -241) **“Stagnation events can be characterized by low wind speeds and increased atmospheric stability, possibly leading to enhanced levels of pollutants like NO_x. Previous studies have shown that these stagnant conditions can result in enhanced levels of N_2O_5 (Baasandorj et al., 2017) driven by high ozone and NO_2.**

> **However, $ClNO_2$ production was limited during stagnation events in this study. This is likely due to limited availability of chloride as shown in submicron particle measurements of aerosol mass spectrometer (AMS) at the ground site for OP and airborne over TRF (Figure 3).”**

Minor Comments:

Throughout text – change ‘ppb’ to ‘ppbv’ and ‘ppt’ to ‘pptv’

Changes have been made throughout the text.

line (ln) 16 – The authors reference the laboratory work of Roberts et al. (2008) showing Cl2 production from N2O5 uptake on acidic, chloride-containing aerosol (R5). There is no known field evidence of this reaction occurring on ambient aerosol, even at low pH. For example, the recent study by McDuffie, E. E. et al. (2018) found a negative correlation between particle acidity and Cl2(g) during the WINTER aircraft campaign, which is the opposite expected trend from this reaction. While this one study cannot confirm or deny the presence of the net N2O5 → Cl2 reaction, the authors should note that there are uncertainties regarding the occurrence of this particular reaction on ambient aerosol.

The following has been added:

> (Ln 21) **“However, this reaction has yet to be proven in ambient conditions.”**

Ln 18 – In addition to the decreased lifetime of NO3 during the day, the short lifetime of N2O5 is largely due to its thermal instability during the day.

The sentence has been revised as below:

> (Ln22-23) **“During the day, N_2O_5 exists at low levels due to its thermal instability (Malko and Troe, 1982) and the short lifetime of NO_3 (NO_3 < 5 s) from photolysis and reaction with NO (Wayne et al., 1991).”**

Ln 19 – define ‘reactive chlorine’

The sentence has been revised as below:

> (Ln 23-24) **“Particulate Cl^- and chlorine containing gas species can come from both natural sources such as sea salt…”**

Ln 35 – since there is discussion of the efficiency of ClNO2 production, it might be helpful to change R4 to the following reaction to show the dependence of ClNO2 on the ClNO2 yield and N2O5 uptake coefficient.

R4 has been replaced as below:

$$N_2O_{5(g)} \xrightarrow{\gamma(N_2O_5),Cl^-(aq)} (2-\phi)*HNO_{3(g)} + \phi*ClNO_{2(g)} \qquad \text{(R4)}$$

Paragraph starting on 37: There are many more observations than the few U.S. studies that are referenced. It is not necessary to cite all of these past studies, however, there are a growing number of observations in Asia, which should be referenced/discussed here as this is the focus area of this manuscript. Including, but not limited to: (Liu et al., 2017; Tham et al., 2018; Tham et al., 2016; Tham et al., 2014; Wang, X. et al., 2017; Wang et al., 2016; Wang, Z. et al., 2017; Wang et al., 2018; Wang et al., 2014; Yun, Hui et al., 2018). Many of these studies are discussed in the paragraph starting on line 66, but should still be referenced here when first discussing the history of ClNO2 measurements.

The sentences have been rearranged and further revised to include the studies in Asia.

> (Ln 55-64) **"More recently (in the past 5 years), increasing number of $ClNO_2$ observations have been conducted in Asia consistently showing significant levels of $ClNO_2$ present in the boundary layer (e.g., Tham et al. 2018, 2016; Wang et al. 2016, 2017c, 2014; Yun et al. 2018; Liu et al. 2017). $ClNO_2$ observations at semi-rural (Wangdu of Hebei province) and urban (Hong Kong, Jinan) regions in China have measured up to 2 ppbv and 776 pptv respectively. At the mountain top (957 m above sea level) in Hong Kong, up to 4.7 ppbv of $ClNO_2$ was reported. The high levels of $ClNO_2$ in these studies were mostly correlated with continental pollution in vicinity (e.g., power plant plumes, biomass burning). A recent study by Yun et al. (2018) reported the highest $ClNO_2$ (8.3 ppb), during a severe have event in a semi-rural site downwind of the Pearl River Delta in the winter. Overall, observations have shown that $ClNO_2$ is ubiquitous in the tropospheric boundary layer."**

Ln 57 – Add a reference for the model tendency to underestimate ClNO2. For instance, if the simulated ClNO2 yield is too large, an underestimation in emissions would not necessarily lead to an underestimation of ClNO2. The authors should additionally add a statement in this section about current uncertainties in the ClNO2 yield as this adds uncertainty to model-predicted ClNO2 as well. This topic was recently reviewed in McDuffie et al. (2018a) and references therein.

The sentences have been re-written as below :

> Ln 66-68 **"Uncertainties in model simulated $ClNO_2$ can arise from limited emission inventories, low resolution of the grid, uncertainties in γ_{N2O5} and Φ_{ClNO2} parameterization, complexity of the terrain, and meteorological conditions and these have been dealt in previous studies (e.g., Zhang et al. 2017; McDuffie et al. 2018b, a; Lowe et al. 2015; Sarwar et al. 2012, 2014; Sherwen et al. 2017)."**

Ln 66 – Add a citation to Yun, H. et al. (2018), who report the largest concentrations of ClNO2 to-date (8.3 ppbv!)

> (Ln 61-63) "**A recent study by Yun et al. (2018) reported the highest $ClNO_2$ (8.3 ppb), during a severe have event in a semi-rural site downwind of the Pearl River Delta in the winter.**"

Ln 85 – As this study also includes a box model analysis, the authors should include a reference to this part of the analysis here.

> (Ln 97) **"We present observational and box model results to evaluated the impact of ClNO2 towards regional air quality in SMA."**

Ln 100 – Were THS CIMS used at both ground sites and on the NASA DC-8? Please clarify. Much of the information in this paragraph could be moved to the SI since the iodide adduct chemistry with ClNO2 and Cl2 is not novel and has been used for many of the past measurements of these species.

We agree with the reviewer that the iodide adduct chemistry is not novel and has been used in many previous studies. However, the information included in our method section is just a very brief summary of it, which we believe will help the readers for better understanding.

The same THS quadrupole CIMS were used at both ground sites and also on the DC-8. The related sentences have been revised as below for clarity:

> (Ln 117-118) **"A THS Instruments LLC Chemical Ionization Mass Spectrometer (CIMS), using iodide (I^-) as the reagent ion was used for measuring Cl_2 and $ClNO_2$ at the two ground supersites and on the NASA DC-8."**

Ln 107 – It is unclear which CIMS instrument is being discussed here, or whether all three instruments have the same configuration. Please clarify.

We used the same CIMS system on the two ground sites and airborne. However, the inlet configuration slightly varied as described in the manuscript. For clarification, the sentence has been revised as below:

> (Ln 123-125) **"Therefore, the use of different types of inlets (e.g., the use of the donut), described above, at the two ground sites and on the DC-8 is not expected to be an issue for the quantitative comparisons in this study."**

Ln 161 – The authors use LST here and throughout the text and KST in Figures 2 and 3. Please change for consistency.

Changed

Ln 167 – Riedel et al. (2012) have also extensively discussed the correlation between Cl2 and ClNO2 in data offshore of LA, which should be cited/discussed here. It is helpful to discuss similarities and differences with additional urban areas outside of Asia.

Comparison with Riedel et al. (2012) has been included as below:

> (Ln 190-192) **"Nighttime relationship between $ClNO_2$ and Cl_2 varied day by day and did not show a clear correlation. This implies that the sources of Cl_2 and $ClNO_2$ was not consistent at night. This is similar to Riedel et al. (2012), where they reported a wide range of correlation between Cl_2 and $ClNO_2$ off the coast of LA "**

Ln 172 – 175 – Please clarify here how the presence of power plants and aerosol organics would impact the observed ClNO2/Cl2 correlation.

Ln 174-176 have been removed and the following sentence has been re-written as below:

> (Ln 203-205) **"However, in this study, the $ClNO_2$ measured at both the OP and TRF sites was weakly correlated with SO_2 ($R^2 = 0.02$), which implies that the air masses that we sampled are not fresh emissions from power plants."**

Ln 180 – As previously mentioned, it should also be noted here that there is currently no field-evidence for this reaction.

The following has been added:

> (Ln 210) "**Therefore, the efficiency of this reaction in ambient conditions requires further investigation.**"

Ln 200 – Please add details about the calculation of the nitrate radical production rate, such as the rate coefficient that was used. It is also unclear here how a 'slow nitrate radical production rate' is consistent with a rapid drop in ClNO2 at 22:00 LST.

The suggestions have been added as below:

> (Ln 251-253) **"The trend was consistent with slower nitrate radical production rate ($d[NO_3]/dt = [NO_2]*[O_3]*k$, where $k= 3.52\times10^{-17}$ at 298 K, Atkinson et al., (2004)), as O_3 was titrated to zero by NO close to midnight."**

Ln 201 – At night the boundary layer becomes vertically stratified, which results in a surface layer and residual layer.

The sentence has been removed.

Ln 211 – There have been multiple studies that have discussed the change in N2O5 chemistry with altitude. Many of these have been in the context of nitrate aerosol production. It would be good to reference some of this past work (in addition to Brown et al. (2017)) when discussing the change in ClNO2 production with altitude. For example: (Baasandorj et al., 2017; Tham et al., 2016; Young et al., 2012; Yun, Hui et al., 2018)

The references have been added and discussed as below:

> (Ln 254-259) **“However, significant levels of N_2O_5 and $ClNO_2$ could have been present in the upper part of the surface layer as shown in previous studies (Baasandorj et al., 2017; Young et al., 2012; Yun et al., 2018). According to Baasandorj et al. (2017), significant levels of N_2O_5 were observed aloft, within the boundary layer, while O_3 was completely titrated near the surface. On the other hand, airborne measurements at the LA basin (Young et al., 2012) showed a relatively uniform $ClNO_2$ profile throughout the boundary layer as O_3 did not change significantly within the measured altitude.”**

Ln 235 – Figure 9 does not show the agreement between the observed and simulated ClNO2 mixing ratios as indicated. The authors might have meant to reference Figure 7.

The reference to figure 9 has been corrected to 8 in the text.

Ln 257 – Please define net O3 production rate and explain how this was calculated from model simulations.

The following has been included in the supplementary for clarity:

> (supplementary Ln 55-57) **Net O_3 production rate was calculated in the box model as below, where f is the stoichiometric coefficient of O_3 and k is the rate constant corresponding to each reaction i. More details can be found in the supplements of Wolfe et al. (2016) :**

$$d[O_3]/dt = O_3\ \text{production rate} - O_3\ \text{loss rate} = \sum_{i=1}^{\#\ of\ reactions} fi \times (product\ of\ reactants)i \times ki$$

Table 1 – Clarify that chloride, nitrate, and sulfate are concentrations from particles < 1um in diameter.

Changed

Figure 1 – Increase the text size and resolution of the images in panel a. The insert is difficult to read.

Changed

Figure 2. The NOx observations should be averaged to the same time interval (i.e., 10 minutes) to allow for direct comparison between the sites.

Changed

Figure 3 – This timeseries makes Cl2 appear as if it has a constant background of ~2 pptv at each site. Could the authors comment on this background and discuss whether it is real or an instrument artifact?

Figure 3 has been corrected to remove data points below detection limit. For Cl_2, the detection limit was around 2.9 ppt (2 sigma, over 30 min).

Figure 6 – Expand panel a to increase the visibility of the ClNO2 data.
Changed

Figure 7 – Clarify that sunrise is at time zero. Clarify what the standard deviations are referring to. The authors should also mention why ClNO2 at t=0 is 0 ppbv when there is actually ClNO2 present at sunrise.

Figure 8 caption has been revised as below:

> **"Steady state $ClNO_2$, simulated from a box model constrained with airborne measurements (blue) and ground site data from TRF (red), when there was morning $ClNO_2$ peak in the morning. Averaged values of the model runs are shown here with standard deviations."**

More details of the box model simulations were added as below:

> **(Supplementary Ln 31-34) "Daytime steady state Cl_2 simulations (Figure 5) were constrained with meteorology and trace gas observations corresponding to each point of the data shown in Figure 4. The constrained parameters were kept constant throughout the 72 hours of integration time and the end points are shown in Figure 5. Steady state simulations of $ClNO_2$ production in the morning (Figure 8) were ran similarly by holding constant of all the measured parameters throughout the runs, except for $ClNO_2$."**

SUPPLEMENT
Section 1 –
As the AMS typically reports aerosol pH, the authors should compare their pH calculations to those from the AMS.
The AMS doesn't typically report aerosol pH. The AMS does provide ammonium balance but previous studies (Hennigan et al., ACP, 2015; Guo et al., JGR, 2016; etc.) have shown that this solely can't be used to determine aerosol pH. This is due to aerosol pH having a non-linear relationship with temperature and liquid water/RH (e.g., Guo et al., JGR, 2016; Guo et al., ACP, 2017; Song et al., ACP, 2018). Similarly, subtracting anions from cations to get H^+ concentration wouldn't be plausible since H^+ depends on water and temperature. Therefore, the

only way to determine pH would be through running a thermodynamic model as have been done in our study.

Figure S1 – Clarify whether this inlet configuration is for the ground-based CIMS or aircraft instrument. If for the ground-based CIMS, was it the same for both instruments?

> (Supplementary Ln 5-8) **"During the KORUS-AQ 2016 field campaign a chemical ionization mass spectrometer (CIMS) was deployed to measure Cl_2 and $ClNO_2$. These systems were deployed at the Taehwa Research Forest (TRF), Olympic Park (OP), and on-board the NASA DC-8. The configuration of the inlet at the two ground sites is shown in Figure S1. The CIMS on the DC-8 had a similar configuration but without the heating inlet."**

Figure S6 – It is unclear how the May 5th profile of ClNO2 relates to the diurnal average profile. The font size of the insert also needs to be increased.

As mentioned in the Ln 270-271, in the morning of May 5th at TRF, maximum levels of $ClNO_2$ net production was required to reconcile the measurements. This insert was included as the 'max' case along with the 'average' case. The font size of the insert has been adjusted.

Editorial Comments:

Throughout the text – check the consistency of 'aerosol' vs. 'aerosols' when using the plural form

changed

line (ln) 5 – Change "in both sites" to "at both sites"

changed

ln 6 – Either change "variation" to "variations" or "were" to "was"

changed

ln 11 – Either change to "the net ozone production rate" or to "net ozone production rates by"

changed

ln 15 – Change to ", which generated from an equilibrium reaction with…"

changed

ln 16 – Change "In acidic aerosols" to "On acidic aerosol"

changed

ln 20 – Change to "coal-fired power plants"

changed

ln 22 – Change to "N2O5 aerosol uptake coefficient (g (N2O5)), aerosol surface area, and N2O5 mean molecular speed, as well as the yield…"

changed

ln 23 – The references provided are only a subset of the relevant literature. Change to (e.g., Thornton et al. …)

changed

ln 29 – Change “level of ozone” to “ozone (O3) level”
changed
ln 37 – change to “The first ambient measurements of ClNO2 were carried out by Osthoff et al. (2008), from a ship sampling along the southeastern U.S. coast in 2006”
changed
ln 47 – change to “models”
changed
ln 53 – Change 7 ppbv and 20% so that both O3 and OH changes are either reported in percent or ppbv.
Ln 78-79 has been changed as below:
“The results showed that, compared to the simulations without ClNO$_2$ formation, monthly 8 h wintertime maximum O$_3$ and □OH increased up to 15 % and 20 %, respectively.”
ln 71 – Change “correlated to” to “correlated with”
changed
ln 78 – Change from “observation results during” to “observations from”
changed
ln 82 – Change “were conducted” to “were collected”
changed
ln 83 – Change to “and included airborne observations from the NASA DC-8…”
changed
ln 84 – Change “ground” to “ground-based”
changed
ln 95 – Change to “research flights when ClNO2 was measured’.
changed

Ln 105 – Change to “in various field conditions”
changed
Ln 106 – Change to “at the two ground sites”
changed
Ln 110 – Define slpm
revised
Ln 111 – PTFE is used on line 103, prior to the definition here.
Revised to define earlier
Ln 157 – Change to “During most nights…”
changed
Ln 161 – Change to “At both sites,…”
changed
Ln 164 – Change to “positive correlation with Cl2…”
changed
Ln 173 – Change to “the ClNO2 measured at both the OP and TRF sites was weakly correlated…”
changed

Ln 175 – Add “e.g.,” before the McDuffie et al. (2018b) and Thornton et al. (2003) references as there have been many more studies that have looked at the organic influence on N2O5 uptake.
changed

Ln 184 – Change to “at the ground sites was highly correlated with the origin of the air mass…”
changed

Ln 185 – Change to “During the nights shaded in red in Figure 3 …”
changed

Ln 186 – Change to “there was limited production of ClNO2 at the surface.” Also, change to “These periods corresponded to low contributions from air masses originating over the ocean and with limited particle chloride concentrations measured by the…”
changed

Ln 190 – Change to “when nitrate production was limited at the surface due to O3 reaction with NO, ClNO2…”
changed

Ln 198 – Change to “significant levels of ClNO2 were sustained throughout the night during most of the…”
changed

Ln 204 – Change to “near the surface”
changed

Ln 207 – Change to “a Cavity Ringdown Spectrometer (CRDS) was installed on top of the Seoul tower in May -June that measured N2O5, NOx, and O3”
changed

Ln 209 – Change to “the average nighttime O3 mixing ratio was around 50 ppbv and N2O5 was observed most nights, with mixing ratios reaching up to 5 ppbv.”
changed

Ln 226 – Change to “At night…”
changed

Ln 233 – Change to “However, the remaining flights observed an average of only 17 ± 56 pptv of ClNO2 (black circles).”
changed

Ln 255 – Change to “methods section.”
changed

Ln 270 – Place the reference to Tham and Wang in parentheses.
changed

Figure 3 - Remove the extra ‘)’ after m-3. Change to “ClNO2 and Cl2 observations and results…”
changed

Figure 5 – Change to “Diurnal variation of ClNO2 and other parameters and trace gases calculated and measured during the campaign…”
changed

Figure 9 – provide a label for the x-axis.

changed

SUPPLEMENT

Ln 5 – Change to “model was run in the reverse mode”

changed

Ln 16 – Change to “model was run similarly here, and it took…”

changed

Ln 21 – Change to “pH and liquid water concentrations for sub-micron aerosol.”

changed

Figure S5. Change to “production rate of the nitrate radical.”

changed

Figure S6 – Change to “The insert in (b) is the…”

changed

Figure S7 – Change the Figure 5 reference to Figure 6.

changed

Reviewer #2

Jeong et al. reported ground-based and airborne observations of ClNO2 in Korea during the Korean- United States-Air Quality (KORUS-AQ) 2016 field campaign. They analyzed the general characteristics, the sources and the effects of ClNO2 on ozone during their measurement campaign. The study contributes to the growing body of ClNO2 measurements around the world, and the content fits the scope of *Atmospheric chemistry and Physics.* The manuscript can be improved by adding more detailed description of measurements and more in-depth analysis/discussion on the elevated levels of ClNO2 in the morning. In addition, there are a number of places which need to be modified for clarity.

Specific comments

(1) Introduction: the manuscript gave a detailed review of measurements of ClNO2 conducted in North America and Europe, but didn’t include some recent work in Asia (mostly in China). As the latter is more relevant to the present study due to proximity of the study regions, these studies should be reviewed (following the review of North America/Europe results). These studies are listed below:

· Wang, H., Lu, K., Guo, S., Wu, Z., Shang, D., Tan, Z., Wang, Y., Breton, M. L., Lou, S., and Tang, M.: Efficient N2O5 uptake and NO3 oxidation in the outflow of urban Beijing, Atmospheric Chemistry and Physics, 18, 9705-9721, 2018.

· Zhou, W., Zhao, J., Ouyang, B., Mehra, A., Xu, W., Wang, Y., Bannan, T. J., Worrall, S. D., Priestley, M., and Bacak, A.: Production of N2O5 and ClNO2 in summer in urban Beijing, China, Atmospheric Chemistry and Physics, 18, 11581-11597, 2018.

· Yun, H., Wang, W., Wang, T., Xia, M., Yu, C., Wang, Z., Poon, S. C. N., Yue, D., and Zhou, Y.: Nitrate formation from heterogeneous uptake of dinitrogen pentoxide during a severe winter haze in southern China, Atmospheric Chemistry and Physics, 2018, 23, 10.5194/acp-2018-698, 2018.

· Wang, Z., Wang, W., Tham, Y. J., Li, Q., Wang, H., Wen, L., Wang, X., and Wang, T.: Fast heterogeneous N2O5 uptake and ClNO2 production in power plant and industrial plumes observed in the nocturnal residual layer over the North China Plain, Atmospheric Chemistry and Physics, 17, 12361-12378, 2017.

· X. Wang, H. Wang, L. Xue, T. Wang, L. Wang, R. Gu, W. Wang, Y. J. Tham, Z. Wang, L. Yang, J. Chen and W. Wang, Observations of N2O5 and ClNO2 at a polluted urban surface site in North China: High N2O5 uptake coefficients and low ClNO2 product yields, Atmospheric Environment, 156. 125-134. 2017.

More studies in Asia have been added as suggested:

> (Ln 55-64) **"More recently (in the past 5 years), increasing number of ClNO$_2$ observations have been conducted in Asia consistently showing significant levels of ClNO$_2$ present in the boundary layer (e.g., Tham et al. 2018, 2016; Wang et al. 2016, 2017c, 2014; Yun et al. 2018; Liu et al. 2017). ClNO$_2$ observations at semi-rural (Wangdu of Hebei province) and urban (Hong Kong, Jinan) regions in China have measured up to 2 ppbv and 776 pptv respectively. At the mountain top (957 m above sea level) in Hong Kong, up to 4.7 ppbv of ClNO$_2$ was reported. The high levels of ClNO$_2$ in these studies were mostly correlated with continental pollution in vicinity (e.g., power plant plumes, biomass burning). A recent study by Yun et al. (2018) reported the highest ClNO$_2$ (8.3 ppb), during a severe have event in a semi-rural site downwind of the Pearl River Delta in the winter. Overall, observations have shown that ClNO$_2$ is ubiquitous in the tropospheric boundary layer."**

(2) Line 14: The lifetime of ClNO2 depends on the photolysis rate which varies among regions, seasons, weather conditions, etc. It's better to specify the condition in which the lifetime of ClNO2 is ~ 30 min and provide a reference.

The suggested information has been added as below:

> (Ln 15-17) **"Nitryl chloride (ClNO$_2$) is a night time radical reservoir that generates chlorine radicals (Cl□) upon sunrise (R1), with a lifetime (τ_{ClNO2}) of ~ 30 minutes at midday in the northern hemisphere mid-latitude summer, under clear sky conditions ($J_{ClNO2}=5.47\times10^{-04}$ s^{-1}, (Madronich and Flocke, 1998))."**

(3) Line 17: "resulting from an enhanced ClNO2 uptake coefficient of up to 3 orders of magnitude" is not clear. Is the uptake in the condition of pH<2 "3 orders of magnitude" higher than that in the condition of pH=7? Please elaborate.

> (Ln 19-21) **“In acidic aerosols (~pH 1.8), uptake of N_2O_5(g) can also produce gas-phase chlorine (Cl_2, R5), resulting from enhanced $ClNO_2$ uptake coefficient of up to 3 - 4 orders of magnitude higher than neutral pH (Roberts et al., 2008).”**

(4) Line 21: on source of chloride, Fu et al. (2018） presents a fine chloride emission inventory for China. This can be a good reference here.
· Fu, X., Wang, T., Wang, S., Zhang, L., Cai, S., Xing, J., and Hao, J.: Anthropogenic Emissions of Hydrogen Chloride and Fine Particulate Chloride in China, Environmental Science & Technology, 52, 1644-1654, 2018.

The suggested reference has been added in Ln 26.

(5) Line 25: Is the heterogeneous uptake of ClNO2 on acidic particle (Roberts et al., 2008) taken into account in the calculation of the lifetime (30h) during the nighttime?

No, it was not taken into account since this specific reaction on enhanced uptake coefficient of $ClNO_2$ on acidic particles has been reported in a laboratory study by Roberts et al. (2008) and uncertainties do remain in terms of its efficiency in ambient aerosols.

(6) Line 45: These previous studies showed that ClNO2 is ubiquitous at surface, within the boundary layer or in the lower troposphere around the world. No evidence has shown that the ClNO2 is ubiquitous in the troposphere which could reach >10 Km above the sea level in mid-latitude region.

Re-written as below:

> (Ln 63) “**Overall, observations have shown that $ClNO_2$ is ubiquitous in the tropospheric boundary layer.**”

(7) Line 55-57: another modeling study Li et al. (2017) assessed the ozone impact of ClNO2 in East Asia including Korea.
· Zhang, L., Li, Q., Wang, T., Ahmadov, R., Zhang, Q., Li, M., and Lv, M.: Combined impacts of nitrous acid and nitryl chloride on lower-tropospheric ozone: new module development in WRF-Chem and application to China, Atmospheric Chemistry and Physics, 17, 9733-9750,2017.

The reference has been added as below:

> (Ln 84-85) **“Another modeling study of WRF-CHEM embedded with an updated chlorine chemistry, simulated 3-6 % of surface O_3 increase in the North China Plain and Yangtze River Delta during the summer (Zhang et al., 2017).”**

(8) Line 58-65: the discussion on model resolution does not seem to be relevant to the present study.

The discussion on model resolution was included to present the discrepancies remaining between observation and model results. These discrepancies lead to the importance of more observations being required in different chemical regimes.

(9) Line 64-65: I assume that the authors are referring to the simulations by Sherwen et al. (2017) that underestimated the ClNO2 by 7 times. Please confirm.

The reference has been added in the text as below:

> (Ln 73-74) **“Compared to observations, the simulations underestimated the $ClNO_2$ maxima levels by ~ 7 times in inland areas (Sherwen et al., 2017).”**

(10) Line 73-78 discusses importance of chorine source in coastal cities and gives the reader an impression that it is only important near coast. But measurement data have shown it is present far inland as shown in Thronton et al. (2010). A recent compilation of PM2.5 data also shows high levels of chloride are present in inland regions of China (Yang et al., STOTEN, 2017). The relevant sentence should be modified.

· Yang, X., Wang, T., Xia, M., Gao, X., Li, Q., Zhang, N., Gao, Y., Lee, S., Wang, X., and Xue, L.: Abundance and origin of fine particulate chloride in continental China, Science of The Total Environment, 624, 1041-1051,2018.

As mentioned in Ln 52 (“Recent studies show that high levels of $ClNO_2$ are also present in mid-continental regions.”) and in the following sentences, we give examples of field observations affected by continental pollution.

(11) Section 2.1. It would help to include a brief description of the meteorology during the campaign.

The following has been added as suggested:

> (Ln 110-115) **“Meteorology during the observation period can be classified into dynamic (May 4th - 16th), stagnation (May 17th - 22nd), transport (May 25th - 31st), and blocking period as shown in Figure 3. During the stagnant period, high pressure system was persistent in the Korean peninsula resulting in local air masses to be more dominant within the SMA compared to the dynamic and transport (May 25nd - 31st) periods. Rex block patterns were observed during the blocking period (June 1st - 6th), which also resulted in more local influence.”**

(12) Section 2.2 on CIMS and calibration: more detailed information is needed. What is the length of the sample line? Was it washed or replaced regularly in order to reduce the loss of N2O5? How frequent was the calibration? Was change of CIMS sensitivity to relative humidity taken into account in data reduction, and how? Could measurement of Cl2 with a Q-CIMS subject to interference? In line 115, 'the natural abundance of Cl2 and ClNO2 isotopes are approximately 9:6:1", what do the authors mean by this statement? No figure was shown and it is not clear how the isotopic ratios behave.

The sampling lines were cleaned regularly and $ClNO_2$ calibration was carried out before and after the campaign and with additional calibrations during the campaign for Cl_2. N_2O_5 data are not presented in this study. The sensitivity of the CIMS was taken into account by normalizing the signals to the water cluster (amu 147 I□$H_2^{18}O$). In the manuscript, it says "The natural abundance of Cl_2 and $ClNO_2$ isotopes are approximately 9:6:1 and 3:1 respectively." which means natural Cl_2 has an isotopic ratio of 9:6:1 and $ClNO_2$ has 3:1. The I□$^{37}Cl^{37}Cl$ (amu 201) was subject to interference with elevated signals that didn't match the isotopic ratio, therefore wasn't considered in our Cl_2 data.

(13) Line 131: How was HONO measured?

HONO is measured as NO_y with the chemiluminescence instrument (Thermo scientific 42i). This is done by conversion of HONO to NO on the oxidized molybdenum surface. The generation of $ClNO_2$ was monitored for a couple of days to ensure minimum production of HONO (Thaler et al., 2011). The data that showed a stable signal of $ClNO_2$ after 2-3 days of the experiment were considered in the calibration.

(14) Section 2.3. Add description on the calculation of the impact of ClNO2 on O3 production rate and on the running of FLEXPART model.

The following has been added in the supplementary:

> (supplementary Ln 55-57) **Net O_3 production rate was calculated in the box model as below, where f is the stoichiometric coefficient of O_3 and k is the rate constant corresponding to each reaction i. More details can be found in the supplements of Wolfe et al. (2016) :**
>
> $$d[O_3]/dt = O_3 \text{ production rate} - O_3 \text{ loss rate} = \sum_{i=1}^{\#\ of\ reactions} fi \times (product\ of\ reactants)i \times ki$$

The following has been added:

> (Ln 173 - 180) **"The FLEXible PARTi-cle dispersion model (FLEXPART v9.1, https://www.flexpart.eu) was used for the air mass source contribution (Figure 3) and backward trajectory analysis (Figure 9). The backward trajectories reported in our**

study were initialized 9:00 LST at TRF, following it 24 hours back in time. The trajectories were driven by the National Centers for Environmental Prediction (NCEP) Global Forecast System (GFS) with a 0.25 degree resolution. Influence of air mass originating from the ocean at TRF and OP was calculated every 6 hours following an air mass 5 days back in time. Meteorology was driven by WRF with a 5 km horizontal resolution. Since emissions of CO are very low in the ocean, and assumed to be inert in the model, it was used as a tracer for contribution of air originating from the ocean within a given air mass at each ground site.”

(15) Line 136. Why is NO2 not constrained in the box model?

(Supplementary Ln 49-54) **“Impact of measured $ClNO_2$ on O_3 production (Figure 10) was explored by constraining the box model with diurnal variation of observations throughout each step. Constraining the model with the diurnal variation of measured $ClNO_2$, allowed the box model to capture its trend throughout the course of the day. Since our purpose of the simulations were to explore the possible impact of $ClNO_2$ on O_3 production, NO_2 and O_3 were only constrained initially at the first step with observations and then calculated based on the chemistry embedded in the model. More specifically, the initial concentration of each following step was taken from the value in the previous step..”**

(16) Line 138-139: elaborate how photolysis rates are determined by scaling on-board DC-8 measurements.

(Ln 158 - 166) “**Photolysis rate constants were derived through the hybrid method (Wolfe et al., 2016) in the F0AM box model. This method uses clear sky solar spectra from the tropospheric ultraviolet and visible radiation model (TUV v 5.2) and cross sections and quantum yields suggested by IUPAC. To capture the effects of pollution on photolysis rates, the ratio of the measured J_{NO2} to the F0AM modeled J_{NO2} was calculated. This ratio was then applied to other photolysis rate constants calculated in the model. Measured J_{NO2} was taken from the DC-8 actinic flux measurements (Charged-coupled device Actinic Flux Spectroradiometer; CAFS) when flying near SMA at altitudes under 1 km. A diurnal cycle was applied to the DC-8 measurement to determine j-values at other times of day. Photolysis rate constants of $ClNO_2$, Cl_2, and $ClONO_2$ were not present in the F0AM model and therefore taken directly from the DC-8 measurements.”**

(17) Line 142-145. Please include the reactions and their rates in the paper, at least in the supplement.

The following sentences have been revised to add related references and sources of the reactions embedded in the model:

> (Ln 166-173) **“The Master Chemical Mechanism v3.3.1 (MCM) was taken from http://mcm.leeds.ac.uk/MCM and embedded in the box model. MCM v3.3.1 has a detailed gas photochemistry (i.e., 5832 species and 17224 reactions), including the oxidation of CH_4 and 142 non-methane primary emitted VOCs (Jenkin et al., 2015). Since MCM v3.3.1 only includes Cl· reactions with alkane species, additional chlorine chemistry was embedded in the model, similar to what Riedel et al. (2014) reported. This was done by including multiple Cl· precursors (e.g., Cl_2, $ClNO_2$, HCl, $ClONO_2$, HOCl) and Cl· reactions with non-alkane VOCs, such as alkene, alcohol, aromatics, alkynes, ketones, organic acids and nitrates. All the reactions embedded in the model can be found in the supplementary of Riedel et al. (2014) and Wolfe et al. (2016).”**

(18) Line 145-148: B&T (2009) parameterization is likely to overestimate N2O5 uptake coefficient. Also, did you assume ClNO2 yield to be unity?

The purpose of the box model simulation of $ClNO_2$ was to explore whether the runs can reproduce the levels that we observed in the morning. Based on the simulations, the model highly underestimated the measured levels by more than 60 times. Therefore, an overestimation of the N_2O_5 uptake coefficient or the yield of $ClNO_2$ would not change our conclusion.

The following has been added in the supplementary:

> (Ln 44-45) **“The yields (*Φ*) of the three heterogeneous reactions were assumed to be 1, therefore the steady state simulations would be an upper-limit of Cl_2 or $ClNO_2$ production.”**

(19) Section 3.1. It would be interesting to see a comparison of the observed values in this study with those reported elsewhere.

$ClNO_2$ observations from previous studies have been dealt in the introduction.

(20) Line 164-165 and Line 176-181: If I understand correctly, (1) when O3 is low, Cl2 level is low but ClNO2 could be high or low, so ClNO2 does not have correlation with Cl2: (2) when O3 is elevated, ClNO2 has a good correlation with Cl2, which could be due to ClNO2 uptake on acidic aerosol to form Cl2 (Roberts et al., 2008). Then what would be the cause of the (1) situation? Why there is no production of Cl2 from ClNO2 uptake when O3 is low, considering that the uptake of ClNO2 on aerosol does not require the presence of O3? Is the pH not low enough? If the Cl2 is solely produced from the ClNO2 uptake, the correlation between ClNO2 and Cl2 at night should be good as well. Is it possible that the Cl2 is mainly formed by gas phase reactions which are initiated by the photolysis of ClNO2 and the reaction of HCl+OH, both of which requires the presence of light? I would recommend the authors to apply box model sensitivity studies to understand the characteristics of ClNO2 (and Cl2).

Box model analysis on the daytime Cl_2 production has been added in the study as below:

(Ln 210 - 230) **“Another possibility is the autocatalytic production of Cl_2 from heterogeneous reactions of gas-phase $ClONO_2$ (i.e., $ClONO_{2(g)} + Cl^{-}_{(aq)} + H^{+}_{(aq)} \rightarrow Cl_{2(g)} + HNO_3$, (Gebel and Finlayson-Pitts, 2001; Deiber et al., 2004)) and HOCl (i.e., $HOCl_{(g)} + Cl^{-}_{(aq)} + H^{+}_{(aq)} \rightarrow Cl_{2(g)} + H_2O$, (Vogt et al., 1996)) on particles. These reactions are also favored as particle acidity increases. In order to further investigate its possibility, daytime Cl_2 was simulated by constraining the box model with measurements of $ClNO_2$ and other trace gases corresponding to each data point in Figure 4. Based on the availability of parameters, we were able to simulate 1680 and 1229 runs for the OP and TRF, respectively. This corresponds to more than 96 % of the daytime data points shown in Figure 4. $ClONO_2$ and HOCl were set to 0.06 (Deiber et al., 2004; Hanson et al., 1994; Hanson and Ravishankara, 1994), which is an upper-limit of previous laboratory studies, and the yields were assumed to be unity. HCl generation from hydrogen abstraction of VOCs by Cl□ were included in the mechanisms used in the model runs. The end points of the 72 hour simulation results are presented in Figure 5. As shown in the Figure, the box model simulations were able to reproduce the positive correlation between Cl_2 and $ClNO_2$. Moreover, modeled Cl_2 was suppressed in low O_3 conditions, which corresponds to the observations. This can be explained by Cl□ reacting with O_3, producing ClO□, leading to gas-phase $ClONO_2$ and HOCl. These can react on acidic aerosols to generate Cl_2. Sources of Cl□ could be from photo-labile gas-phase chlorine compounds (e.g., Cl_2, $ClNO_2$, $ClONO_2$, HOCl) or oxidation of gas-phase HCl by OH. Although the reaction between HCl and OH is relative slow ($k = 7.86 \times 10^{-13}$ cm^3 $molecule^{-1}s^{-1}$ at 298K, (Atkinson et al., 2007)), it has been reported to be a significant source of Cl in the daytime (Riedel et al., 2012). A sensitivity test was carried out by comparing modeled Cl_2 between runs with and without HCl production from oxidation of VOCs by Cl□ (Figure S4 c,d). The results show that production of Cl_2 was suppressed by 40 - 70 % when HCl was not generated in the model. This significant contribution of gas-phase HCl as a Cl source, should be an upper-limit as the deposition of HCl was not considered in the model. Nonetheless, our analysis leads us to conclude that the mechanisms we have explored could be the main contributions of the daytime Cl_2 production during KORUS-AQ.”**

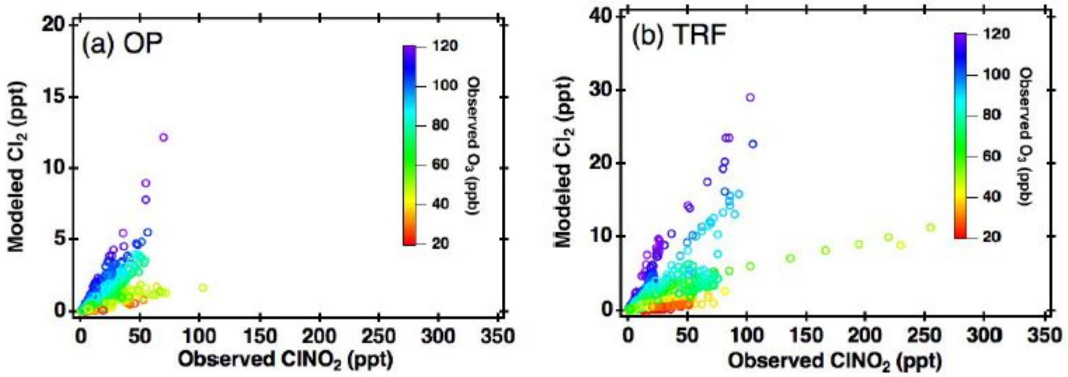

Figure 5. Correlation between box model simulated daytime (11:00 - 18:00 local time) Cl_2 and measured $ClNO_2$ at (a) OP and (b)TRF, color coded with measured O_3.

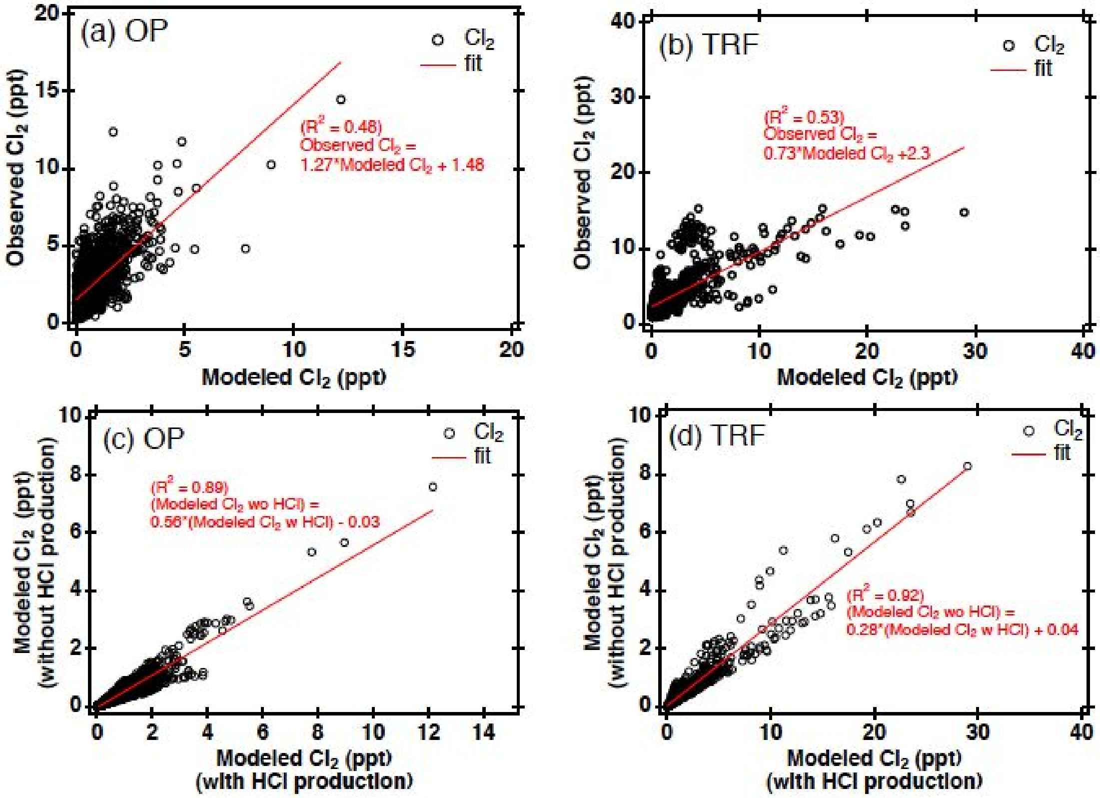

Figure S4. Correlation between measured Cl_2 and modeled Cl_2 at (a) OP and (b) TRF. Sensitivity tests of HCl were carried out (c and d) by switching off HCl production from chlorine radicals reacting with VOCs.

(21) Line 174-176: I don't see why the effect of organic coating on N2O5 uptake is relevant to the correlation of ClNO2 and Cl2.

Ln 174-176 have been removed

(22) Line 184-185: An explanation is needed for the calculation of 'source contribution of CO' using FLEXPART.

The following has been added for clarity:

> (Ln 173 - 180) **"The FLEXible PARTi-cle dispersion model (FLEXPART v9.1, https://www.flexpart.eu) was used for the air mass source contribution (Figure 3) and backward trajectory analysis (Figure 9)….. Influence of air mass originating from the ocean at TRF and OP was calculated every 6 hours following an air mass 5 days back in time. Meteorology was driven by WRF with a 5 km horizontal resolution. Since emissions of CO are very low in the ocean, and assumed to be inert in the model, it was used as a tracer for contribution of air originating from the ocean within a given air mass at each ground site."**

(23) Line 184-185: Do the authors mean that the ClNO2 is highly correlated to the oceanic sources?Please elaborate.

> (Ln 241 - 242) **"Whether the chloride is from the ocean or anthropogenic emissions is uncertain since large point sources, such as power plants or petrochemical facilities, are also present along the west coast of the SMA"**

(24) Line 188-189: Apart from the back trajectory analysis, there are other methods to determine/estimate the source of chloride. Please refer to the previous studies on ClNO2 measurements. For example, In Line 172-174, the correlation of ClNO2 and SO2 is extremely low, so the coal-burning activity is not responsible for the chloride measured during the campaign. Any evidence of biomass burning, chemical signature (e.g. K+) or the fire detected by the satellite? Any evidence of sea-salt aerosol, e.g. how is the correlation of chloride and sodium? What about waste burning?

At the Taehwa Research Forest, there were no aerosol composition measurements carried out during the campaign. FLEXPART back trajectory analysis initialized at 9:00 LST shows an overall westerlies when there was a second morning peak of $ClNO_2$. The following has been added in the discussion:

> (Ln 299-305) **"During KORUS, the DC-8 did not fly to the west of the SMA in the early morning. However, there are large point sources, such as petrochemical facilities and industries, and vehicular emissions to the west and south west of the SMA region. Sullivan et al. (2019) reported that this resulted in enhanced levels of O_3 in receptor regions (i.e., Taehwa Research Forest) downwind when westerlies were prevalent. Therefore, favorable conditions such as high chloride content in aerosols from both anthropogenic and natural sources and high levels of NO_x-O_3 could have**

> **lead to significant levels of $ClNO_2$ to build up and transported to TRF before being completely photolyzed. During the campaign, influence of large biomass burning was negligible as reported in Tang et al. (2018, 2019).”**

(25) Line 189-190: What does “nitrate production was limited due to O3 titration” mean?

The sentence has been removed.

(26) Figure 5(b), Wind directions should be added to see if the morning peak of ClNO2 and the peak of SO2 at 15:00 was related to wind direction change.

We did not observe any changes in the wind direction when the morning peak of $ClNO_2$ was happening.

(27) Line 196-197: an explanation is needed for the choice of the days.

The following sentence has been added and the explanation on the profiles are mentioned in the following sentences.

> (Ln 246) **“...The description on these profiles are further explained in the following sentences.”**

(28) Line 201-202: where is the information on boundary layer height coming from?

The “200 - 300 m” boundary layer height in Line 201-202 was referring to a typical nocturnal boundary layer height during the night, which is not specific for our study. In our study, we used vertical profiles of potential temperature to get the boundary layer heights (Figure 6). Since the DC-8 only flew during the daytime (mostly between 8 am to 4 pm local time), we do not have nighttime measurements of vertical potential temperature profiles during KORUS. Line 201-202 has been removed.

(29) Line 231-234: More detailed discussion of the vertical profiles is needed. How do you define residual layer? In line 210-212, the authors suggested that the ClNO2 in the residual layer could be higher than those at ground surface based on a previous tower measurement in the same region (5 ppb of N2O5 at 360m a.s.l.). Need to reconcile these statements. Was the wind direction different at different altitudes? I suggest the author compare individual vertical profile with ground measurements to better reveal their relationship.

In our study, the boundary layer height was determined based on the vertical profile of the airborne potential temperature measurements. This was averaged during the same timeframe and region as the $ClNO_2$ morning time data shown in Figure 6 (i.e., 8:00 - 8:30 local time). In terms of our statement in ln 210-212, we were suggesting that it is possible that enhanced $ClNO_2$ could have been present in the upper surface layer not the residual layer.

(30) Line 234 and Figure 6(b): The maximum ClNO2 on May 25 appears to be ~200 ppt, while that on May 31 is ~750 ppt and that on June 10 is ~1250 ppt. As to those on other days, dozens samples showed more than 500 ppt even close to 1500ppt in the residual layer (between the nocturnal boundary layer and the boundary layer at midday, in the present study between 200/300m and 1000/2000m). In Figure 6(a), similar results could also be found near the TRF site, over 1000 ppt ClNO2 concentrations were recorded at the height of ~500 m (yellowish). I suggest that the authors revisit the figures and the text.

As in comment #28 and #29, we estimated the nocturnal boundary layer based on the vertical profile of the potential temperature measured on-board the DC-8 in the morning. Based on this measurement, at 8:00-8:30 am local time, the boundary layer height is estimated to be 500 - 600 m. Between this height and above, we did not observe significant $ClNO_2$ levels that could reconcile the ground observations except for the days with colored green square (May 31st), purple triangle (May 25th), and green square (May 31st).

(31) Line 243-251: the evidence for contribution of horizontal transport to the morning peak is not convincing. According to the authors, in half of the days, they measured second peak of ClNO2 at 7-8 am, and in these days, the air masses came from various directions mostly from northwest to southwest and various distance (approximately 200 to 400 Km in 24h) (Fig 8). If the horizontal transport (advection) within the boundary layer is the cause of the second peak of ClNO2, that means in all these directions and distance, there is a bulk of air mass with higher ClNO2 that would constantly arrive at the measurement site at 7-8 am, not before nor after. This is physically not possible.

Following has been added for further discussion:

> (Ln 299-305) **"During KORUS, the DC-8 did not fly to the west of the SMA in the early morning. However, there are large point sources, such as petrochemical facilities and industries, and vehicular emissions to the west and south west of the SMA region. Sullivan et al. (2019) reported that this resulted in enhanced levels of O_3 in receptor regions (i.e., Taehwa Research Forest) downwind when westerlies were prevalent. Therefore, favorable conditions such as high chloride content in aerosols from both anthropogenic and natural sources and high levels of NO_x-O_3 could have lead to significant levels of $ClNO_2$ to build up and transported to TRF before being completely photolyzed. During the campaign, influence of large biomass burning was negligible as reported in Tang et al. (2018, 2019)."**

(32) Line 260-262: Wang et al. 2016 showed much larger contribution of ClNO2 to ozone increase compared to 2% at TRF site, the latter is similar to 3% at Wangdu in Tham et al. (2016).

Line 260 is referring to OH not O_3

(33) Line 270-272: the comment on the result of Tham et al. (2016) and Wang et al. (2016) is an incorrect interpretation of their findings. The three factors (downward transport, horizontal transport and local chemical production) may impact different locations differently. Tham et al. presented evidence for downwind transport in the early morning hours at a polluted rural site in the North China plain. Wang et al. (2016) measured the high ClNO2 plumes at a 974 m mountain- top site and suggested presence of high ClNO2 in the upper boundary layer in south China. They did not say that downward transport should apply to all locations. One has to analyze his/her own case.

We agree on the comment. The following has been added as suggested:

> (Ln 327-332) **"Previous studies have attributed high sustained $ClNO_2$ in the morning to transport from the residual layer (Tham et al., 2016; Wang et al., 2016). In this study, box model runs of heterogeneous and gas-phase production of $ClNO_2$ could not reconcile the observed levels. Moreover, airborne observations in the early morning showed negligible $ClNO_2$ levels in the residual layer in most of the days. Therefore, local transport of $ClNO_2$ from highly polluted airmasses from the west is the most plausible explanation. This shows that different meteorological or chemical conditions of the sites can lead to various causes of high $ClNO_2$ levels in the early morning."**

(34) Figure 7. What was this figure used for?

The reference to figure 9 has been corrected to 7 in the text.

References (*only the references in our responses have been added)

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