Review of Jeong et al.: 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

Jeong and co-authors present an analysis of $ClNO_2$ observations from the 2016 NASA KORUS-AQ campaign. $ClNO_2$, 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 $ClNO_2$ measurements, allowing for direct comparisons between sites. Ground-based observations revealed that $ClNO_2$ was elevated at night at both sites, with higher concentrations at the TRF forested site, which was physically removed from NO emissions, which reduce O_3 concentrations and calculated nitrate radical production rates. At both sites, there were periods where $ClNO_2$ persisted at high levels throughout the morning after sunrise. Using additional box-model simulations, the authors found that morning entrainment of $ClNO_2$ from the residual layer, nor morning $ClNO_2$ 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 $ClNO_2$ concentrations. A final set of 24-hour simulations with and without $ClNO_2$ production, constrained to observations, revealed that chlorine radical initiated chemistry can increase net O_3 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 $ClNO_2$, 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 O_3 and NO_3 radical production rates and aerosol surface area, and other minor and editorial comments.

Major Comments:

Instrument Section:

Ln 130 – The authors note that $ClNO_2$ is thermally converted to NO at 325 C in a CL instrument. The thermal conversion efficiency of $ClNO_2$, 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 $ClNO_2$ measurement, the authors should discuss how they accounted for (or quantified) the thermal conversion efficiency of $ClNO_2$.

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?

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 N_2O_5 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 $ClNO_2$ production, the authors should also state how the $ClNO_2$ 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).

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

Results and Discussion Section:

Ln 161 – The statement that $ClNO_2$ rapidly photolyzes near sunrise contradicts the authors later statements on line 213 that $ClNO_2$ persists after 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 $ClNO_2$?

For example, were NO_x and O_3 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 $ClNO_2$.

Section 3.3.

It is unclear how $ClNO_2$ was used to constrain the model simulations. For example, was the model only initialized with observed $ClNO_2$ 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 $ClNO_2$, it seems important that model simulations emulate that observed behavior.

Conclusions:

The authors mention that stagnation events were associated with low $ClNO_2$ production. There was no discussion in the text, however, about the meteorology associated with these low $ClNO_2$ events. Moreover, past studies have shown that certain types of stagnation events can actually enhance N_2O_5 chemistry (e.g. Baasandorj et al., 2017).

Minor Comments:

Throughout text – change 'ppb' to 'ppbv' and 'ppt' to 'pptv'

line (ln) 16 – The authors reference the laboratory work of Roberts et al. (2008) showing Cl_2 production from N_2O_5 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 Cl_2(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 $N_2O_5 \rightarrow Cl_2$ reaction, the authors should note that there are uncertainties regarding the occurrence of this particular reaction on ambient aerosol.

Ln 18 – In addition to the decreased lifetime of NO_3 during the day, the short lifetime of N_2O_5 is largely due to its thermal instability during the day.

Ln 19 – define 'reactive chlorine'

Ln 35 – since there is discussion of the efficiency of $ClNO_2$ production, it might be helpful to change R4 to the following reaction to show the dependence of $ClNO_2$ on the $ClNO_2$ yield and N_2O_5 uptake coefficient.

$$\mathrm{N_2O_5(g)} \xrightarrow{\gamma(N_2O_5),\ Cl^-(p)} (2-\varphi) * \mathrm{HNO_3} + (\varphi) * \mathrm{ClNO_2} \qquad \text{(R4)}$$

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 $ClNO_2$ measurements.

Ln 57 – Add a reference for the model tendency to underestimate $ClNO_2$. For instance, if the simulated $ClNO_2$ yield is too large, an under-estimation in emissions would not necessarily lead to an underestimation of $ClNO_2$. The authors should additionally add a statement in this section about current uncertainties in the $ClNO_2$ yield as this adds uncertainty to model-predicted $ClNO_2$ as well. This topic was recently reviewed in McDuffie et al. (2018a) and references therein.

Ln 66 – Add a citation to Yun, H. et al. (2018), who report the largest concentrations of $ClNO_2$ to-date (8.3 ppbv!).

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 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 $ClNO_2$ and Cl_2 is not novel and has been used for many of the past measurements of these species.

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

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

Ln 167 – Riedel et al. (2012) have also extensively discussed the correlation between Cl_2 and $ClNO_2$ 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.

Ln 172 – 175 – Please clarify here how the presence of power plants and aerosol organics would impact the observed $ClNO_2/Cl_2$ correlation.

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

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 $ClNO_2$ at 22:00 LST.

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

Ln 211 – There have been multiple studies that have discussed the change in N_2O_5 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 $ClNO_2$ production with altitude. For example: (Baasandorj et al., 2017; Tham et al., 2016; Young et al., 2012; Yun, Hui et al., 2018)

Ln 235 – Figure 9 does not show the agreement between the observed and simulated $ClNO_2$ mixing ratios as indicated. The authors might have meant to reference Figure 7.

Ln 257 – Please define net O_3 production rate and explain how this was calculated from model simulations.

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

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

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

Figure 3 – This timeseries makes Cl_2 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 6 – Expand panel a to increase the visibility of the $ClNO_2$ data.

Figure 7 – Clarify that sunrise is at time zero. Clarify what the standard deviations are referring to. The authors should also mention why $ClNO_2$ at t=0 is 0 ppbv when there is actually $ClNO_2$ present at sunrise.

SUPPLEMENT

Section 1 –

As the AMS typically reports aerosol pH, the authors should compare their pH calculations to those from the AMS.

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?

Figure S6 – It is unclear how the May 5^{th} profile of $ClNO_2$ relates to the diurnal average profile. The font size of the insert also needs to be increased.

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

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

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

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

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

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

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

ln 22 – Change to "N_2O_5 aerosol uptake coefficient ($\gamma(N_2O_5)$), aerosol surface area, and N_2O_5 mean molecular speed, as well as the yield…"

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

ln 29 – Change "level of ozone" to "ozone (O_3) level"

ln 37 – change to "The first ambient measurements of $ClNO_2$ were carried out by Osthoff et al. (2008), from a ship sampling along the southeastern U.S. coast in 2006"

ln 47 – change to "models"

ln 53 – Change 7 ppbv and 20% so that both O_3 and OH changes are either reported in percent or ppbv.

ln 71 – Change "correlated to" to "correlated with"

ln 78 – Change from "observation results during" to "observations from"

ln 82 – Change "were conducted" to "were collected"

ln 83 – Change to "and included airborne observations from the NASA DC-8…"

ln 84 – Change "ground" to "ground-based"

ln 95 – Change to "research flights when $ClNO_2$ was measured'.

Ln 105 – Change to “in various field conditions”

Ln 106 – Change to “at the two ground sites”

Ln 110 – Define slpm

Ln 111 – PTFE is used on line 103, prior to the definition here.

Ln 157 – Change to “During most nights…”

Ln 161 – Change to “At both sites,…”

Ln 164 – Change to “positive correlation with Cl_2…”

Ln 173 – Change to “the $ClNO_2$ measured at both the OP and TRF sites was weakly correlated…”

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 N_2O_5 uptake.

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

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

Ln 186 – Change to “there was limited production of $ClNO_2$ 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…”

Ln 190 – Change to “when nitrate production was limited at the surface due to O_3 reaction with NO, $ClNO_2$…”

Ln 198 – Change to “significant levels of $ClNO_2$ were sustained throughout the night during most of the…”

Ln 204 – Change to “near the surface”

Ln 207 – Change to “a Cavity Ringdown Spectrometer (CRDS) was installed on top of the Seoul tower in May -June that measured N_2O_5, NO_x, and O_3”

Ln 209 – Change to “the average nighttime O_3 mixing ratio was around 50 ppbv and N_2O_5 was observed most nights, with mixing ratios reaching up to 5 ppbv.”

Ln 226 – Change to “At night…”

Ln 233 – Change to “However, the remaining flights observed an average of only 17 ± 56 pptv of $ClNO_2$ (black circles).”

Ln 255 – Change to “methods section.”

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

Figure 3 - Remove the extra ‘)’ after m^{-3}. Change to “$ClNO_2$ and Cl_2 observations and results…”

Figure 5 – Change to “Diurnal variation of $ClNO_2$ and other parameters and trace gases calculated and measured during the campaign…”

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

SUPPLEMENT

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

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

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

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

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

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

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