

Jeong et al. reported ground-based and airborne observations of ClNO<sub>2</sub> 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 ClNO<sub>2</sub> on ozone during their measurement campaign. The study contributes to the growing body of ClNO<sub>2</sub> 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 ClNO<sub>2</sub> 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 ClNO<sub>2</sub> 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 N<sub>2</sub>O<sub>5</sub> uptake and NO<sub>3</sub> 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 N<sub>2</sub>O<sub>5</sub> and ClNO<sub>2</sub> 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 N<sub>2</sub>O<sub>5</sub> uptake and ClNO<sub>2</sub> 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 N<sub>2</sub>O<sub>5</sub> and ClNO<sub>2</sub> at a polluted urban surface site in North China: High N<sub>2</sub>O<sub>5</sub> uptake coefficients and low ClNO<sub>2</sub> product yields, *Atmospheric Environment*, 156, 125-134, 2017.
- (2) Line 14: The lifetime of ClNO<sub>2</sub> 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 ClNO<sub>2</sub> is ~ 30 min and provide a reference.
- (3) Line 17: "resulting from an enhanced ClNO<sub>2</sub> 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.
- (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.

- (5) Line 25: Is the heterogeneous uptake of  $\text{ClNO}_2$  on acidic particle (Roberts et al., 2008) taken into account in the calculation of the lifetime (30h) during the nighttime?
- (6) Line 45: These previous studies showed that  $\text{ClNO}_2$  is ubiquitous at surface, within the boundary layer or in the lower troposphere around the world. No evidence has shown that the  $\text{ClNO}_2$  is ubiquitous in the troposphere which could reach  $>10$  Km above the sea level in mid-latitude region.
- (7) Line 55-57: another modeling study Li et al. (2017) assessed the ozone impact of  $\text{ClNO}_2$  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.
- (8) Line 58-65: the discussion on model resolution does not seem to be relevant to the present study.
- (9) Line 64-65: I assume that the authors are referring to the simulations by Sherwen et al. (2017) that underestimated the  $\text{ClNO}_2$  by 7 times. Please confirm.
- (10) Line 73-78 discusses importance of chlorine 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 Thornton et al. (2010). A recent compilation of  $\text{PM}_{2.5}$  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.
- (11) Section 2.1. It would help to include a brief description of the meteorology during the campaign.
- (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  $\text{N}_2\text{O}_5$ ? How frequent was the calibration? Was change of CIMS sensitivity to relative humidity taken into account in data reduction, and how? Could measurement of  $\text{Cl}_2$  with a Q-CIMS subject to interference? In line 115, 'the natural abundance of  $\text{Cl}_2$  and  $\text{ClNO}_2$  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.
- (13) Line 131: How was HONO measured?
- (14) Section 2.3. Add description on the calculation of the impact of  $\text{ClNO}_2$  on  $\text{O}_3$  production rate and on the running of FLEXPART model.
- (15) Line 136. Why is  $\text{NO}_2$  not constrained in the box model?

- (16) Line 138-139: elaborate how photolysis rates are determined by scaling on-board DC-8 measurements.
- (17) Line 142-145. Please include the reactions and their rates in the paper, at least in the supplement.
- (18) Line 145-148: B&T (2009) parameterization is likely to overestimate  $\text{N}_2\text{O}_5$  uptake coefficient. Also, did you assume  $\text{ClNO}_2$  yield to be unity?
- (19) Section 3.1. It would be interesting to see a comparison of the observed values in this study with those reported elsewhere.
- (20) Line 164-165 and Line 176-181: If I understand correctly, (1) when  $\text{O}_3$  is low,  $\text{Cl}_2$  level is low but  $\text{ClNO}_2$  could be high or low, so  $\text{ClNO}_2$  does not have correlation with  $\text{Cl}_2$ : (2) when  $\text{O}_3$  is elevated,  $\text{ClNO}_2$  has a good correlation with  $\text{Cl}_2$ , which could be due to  $\text{ClNO}_2$  uptake on acidic aerosol to form  $\text{Cl}_2$  (Roberts et al., 2008). Then what would be the cause of the (1) situation? Why there is no production of  $\text{Cl}_2$  from  $\text{ClNO}_2$  uptake when  $\text{O}_3$  is low, considering that the uptake of  $\text{ClNO}_2$  on aerosol does not require the presence of  $\text{O}_3$ ? Is the pH not low enough? If the  $\text{Cl}_2$  is solely produced from the  $\text{ClNO}_2$  uptake, the correlation between  $\text{ClNO}_2$  and  $\text{Cl}_2$  at night should be good as well. Is it possible that the  $\text{Cl}_2$  is mainly formed by gas phase reactions which are initiated by the photolysis of  $\text{ClNO}_2$  and the reaction of  $\text{HCl}+\text{OH}$ , both of which requires the presence of light? I would recommend the authors to apply box model sensitivity studies to understand the characteristics of  $\text{ClNO}_2$  (and  $\text{Cl}_2$ ).
- (21) Line 174-176: I don't see why the effect of organic coating on  $\text{N}_2\text{O}_5$  uptake is relevant to the correlation of  $\text{ClNO}_2$  and  $\text{Cl}_2$ .
- (22) Line 184-185: An explanation is needed for the calculation of 'source contribution of CO' using FLEXPART.
- (23) Line 184-185: Do the authors mean that the  $\text{ClNO}_2$  is highly correlated to the oceanic sources? Please elaborate.
- (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  $\text{ClNO}_2$  measurements. For example, In Line 172-174, the correlation of  $\text{ClNO}_2$  and  $\text{SO}_2$  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.  $\text{K}^+$ ) 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?
- (25) Line 189-190: What does "nitrate production was limited due to  $\text{O}_3$  titration" mean?
- (26) Figure 5(b), Wind directions should be added to see if the morning peak of  $\text{ClNO}_2$  and the peak of  $\text{SO}_2$  at 15:00 was related to wind direction change.

- (27) Line 196-197: an explanation is needed for the choice of the days.
- (28) Line 201-202: where is the information on boundary layer height coming from?
- (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 ClNO<sub>2</sub> 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 N<sub>2</sub>O<sub>5</sub> 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.
- (30) Line 234 and Figure 6(b): The maximum ClNO<sub>2</sub> 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 ClNO<sub>2</sub> concentrations were recorded at the height of ~500 m (yellowish). I suggest that the authors revisit the figures and the text.
- (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 ClNO<sub>2</sub> 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 ClNO<sub>2</sub>, that means in all these directions and distance, there is a bulk of air mass with higher ClNO<sub>2</sub> that would constantly arrive at the measurement site at 7-8 am, not before nor after. This is physically not possible.
- (32) Line 260-262: Wang et al. 2016 showed much larger contribution of ClNO<sub>2</sub> to ozone increase compared to 2% at TRF site, the latter is similar to 3% at Wangdu in Tham et al. (2016).
- (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 ClNO<sub>2</sub> plumes at a 974 m mountain-top site and suggested presence of high ClNO<sub>2</sub> 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.
- (34) Figure 7. What was this figure used for?