

Responses to Editors and Reviewers

We appreciate the reviewers for the constructive and helpful comments, the incorporation of which has led to an improved manuscript. We have revised the manuscript appropriately and addressed the reviewer's comments in our point-by-point responses. As detailed below, the reviewer's comments are shown in black, our responses to the comments are in blue fonts, and the added/rewritten parts are presented in red fonts.

RC1: ['Comment on egusphere-2024-1638'](#), Anonymous Referee #1, 31 Jul 2024

General Comments

The work by Gaojie Chen et al. is a well written study presenting two months of ambient observations in Southeast China and has two main components. First, the work introduces interesting evidence for the formation of ClNO₂ during the daytime by a recently suggested particulate nitrate mechanism. Second, the work discusses the implications for Cl radical production from ClNO₂ photolysis.

The first component has significant implications for the understanding of ClNO₂ formation globally. However, a discussion of the traditional metrics of ClNO₂ formation, the N₂O₅ uptake rate and ClNO₂ yield, are completely absent from the paper. Without a discussion on this topic, the authors' conclusion that "NO₃⁻ photolysis contributed to daytime generation" is severely weakened. In fact, it is based only a machine learning output which gauges the "importance" of NO₃⁻ influence on ClNO₂ as well as a linear regression of ClNO₂ with NO₃⁻ × jNO₂ × aerosol S_a. In this joint correlation, insufficient evidence is provided to suggest that the photolysis component improves the correlation. As such, I request major revisions in which the authors justify their conclusion by demonstrating that the daytime observations of ClNO₂ cannot be explained by traditional N₂O₅ and ClNO₂ chemistry.

The second component is based on box modeling from the master chemical mechanism. Aside from a lack of detail on the parametrization used for N₂O₅ uptake and

CINO₂ yield, the results presented are generally sound and informative. I request that the authors include their choice of parametrization in the main text.

Response: Thanks for your valuable comments. Your review comments and suggestions are benefit to improve the quality and readability of this manuscript. We have revised the manuscript appropriately and addressed the reviewer's comments point-by-point for consideration as below.

The first component: We have added the discussions on the N₂O₅ uptake coefficient ($\gamma(\text{N}_2\text{O}_5)$) and CINO₂ yield ($\phi(\text{CINO}_2)$). Furthermore, we also provided the evidence showing that the daytime observations of CINO₂ cannot be explained by traditional N₂O₅ and CINO₂ chemistry. Please refer to our response to Specific Comment 4 for more details.

The second component: In this study, the box model is employed to evaluate the photochemical effects of CINO₂. The levels of CINO₂ in the box model were constrained by the observed levels of CINO₂ from our field measurements. This approach eliminates the necessity for parameterization of N₂O₅ uptake and CINO₂ yield to determine CINO₂ levels. Therefore, the parametrization for N₂O₅ uptake and CINO₂ yield was not utilized in the box model.

Specific Comments

1. Section 2: A description on the handling of N₂O₅ uptake and CINO₂ yield is absent from the methods. A list of previous papers is provided but it is not clear how these two parameters are handled. Both N₂O₅ uptake and CINO₂ yield will vary with the parameters investigated here (T, RH, etc.). See McDuffie et al.

McDuffie, E. E., Fibiger, D. L., Dubé, W. P., Lopez Hilfiker, F., Lee, B. H., Jaeglé, L., et al. (2018a). CINO₂ yields from aircraft measurements during the 2015 WINTER campaign and critical evaluation of the current parameterization. *Journal of Geophysical Research: Atmospheres*, 123(22), 12994–13015. <https://doi.org/10.1029/2018JD029358>

McDuffie, E. E., Fibiger, D. L., Dubé, W. P., Lopez-Hilfiker, F., Lee, B. H., Thornton, J. A., et al. (2018b). Heterogeneous N₂O₅ uptake during winter: Aircraft measurements

during the 2015 WINTER campaign and critical evaluation of current parameterizations. Journal of Geophysical Research: Atmospheres, 123(8), 4345–4372. <https://doi.org/10.1002/2018JD028336>

Response: Thanks for your comment. In this study, the box model is employed to evaluate the photochemical effects of ClNO₂. The levels of ClNO₂ in the box model were constrained by observed levels of ClNO₂ from our field measurements. This approach negates the need for parameterization of N₂O₅ uptake and ClNO₂ yield to determine ClNO₂ levels. Therefore, the parametrization for N₂O₅ uptake and ClNO₂ yield was not utilized in the box model.

Added/rewritten: “Due to the levels of ClNO₂ in the box model determined by observed levels of ClNO₂, the parametrization for N₂O₅ uptake and ClNO₂ yield was not utilized in the box model.”

2. Section 3.1: There is no uncertainty presented with the observations in the main text. Please include the uncertainties as the uncertainties in the SI are non-negligible (~20 %).

Response: Thanks for your comment. We have included the uncertainties in the main text.

Added/rewritten: “The uncertainties of the ClNO₂ and N₂O₅ measurements were estimated to be ~20 %.”

3. Figure 5: What is the interpretation of negative “importance factors”? During the daytime, N₂O₅ is a negative importance factor. Please discuss this in the main text.

Response: Thanks for your comment. In the XGBoost-SHAP model, SHAP values are used to quantify the contribution of each feature to the prediction values, with a negative SHAP value indicating a negative contribution. Generally, negative “importance factors” suggest that the presence of these factors contributes minimally or decreases the predicted values of the dependent variable. Therefore, in our study, negative SHAP values for N₂O₅ during the daytime indicate that the contribution of N₂O₅ chemistry to daytime ClNO₂ levels was limited. We have added these discussions.

Added/rewritten: “Generally, negative “importance factors” suggest that the

presence of these factors contributes minimally or decreases the predicted values of the dependent variable. Therefore, in our study, negative SHAP values for N_2O_5 during the daytime indicate that the contribution of N_2O_5 chemistry to daytime $ClNO_2$ levels was limited.”

4. Section 3.2: A discussion on the changes in aerosol content (particulate nitrate) and the effect on N_2O_5 uptake and $ClNO_2$ yield is absent. Such a discussion is critical here. Traditionally, one expects nitrate to reduce N_2O_5 uptake (the nitrate effect) which would limit the production of $ClNO_2$. Even so, $ClNO_2$ could be enhanced in a high nitrate case if the N_2O_5 uptake and $ClNO_2$ yield are substantially greater than low nitrate air masses. According to Figure 1, there are concurrent enhancements of pCl and pNO_3 during some time periods. As pCl increases the $ClNO_2$ yield will also increase which would then be (coincidentally?) concurrent with high pNO_3 . Even more, these periods of concurrent pCl and pNO_3 appear to correlate with enhanced $PM_{2.5}$ and thus, I assume, aerosol surface area. Increases in surface area would then increase N_2O_5 uptake further promoting $ClNO_2$ and pNO_3 production. Lastly, Figure 6 suggests that the correlation between $ClNO_2$ mixing ratio and $pNO_3 \times jNO_2 \times Sa$ is driven by $pNO_3 \times Sa$ while jNO_2 has a limited or no correlation (panel d). In other words, photolysis appears to have a limited role in the production of $ClNO_2$.

While the above may be speculative, it is an example of why a lack of discussion on the $ClNO_2$ yield and N_2O_5 uptake significantly weakens the arguments made by the authors. As written, I believe there is insufficient evidence to conclude that “ NO_3^- photolysis contributed to daytime $[ClNO_2]$ generation”.

Response: Thanks for your valuable comments. The N_2O_5 uptake coefficient ($\gamma(N_2O_5)$) and $ClNO_2$ yield ($\phi(ClNO_2)$) were estimated using the observational data and parameterization. We derived the values of $\gamma(N_2O_5)$ and $\phi(ClNO_2)$ based on increased rates of $ClNO_2$ and particle nitrate (NO_3^-) in the field observation (Phillips et al., 2016). Specially, $\gamma(N_2O_5)$ and $\phi(ClNO_2)$ were calculated by Eq. (1) and (2).

$$\gamma(N_2O_5) = \frac{2 \times (P(ClNO_2) + P(NO_3^-))}{cN_2O_5 S_a [N_2O_5]} \quad (1)$$

$$\phi(\text{ClNO}_2) = 2 \times \left(1 + \frac{P(\text{NO}_3^-)}{P(\text{ClNO}_2)} \right)^{-1} \quad (2)$$

Here, $P(\text{ClNO}_2)$ and $P(\text{NO}_3^-)$ represent the production rates of ClNO_2 and NO_3^- induced by N_2O_5 uptake, respectively. S_a denotes the aerosol surface area, and $c(\text{N}_2\text{O}_5)$ is the mean molecular speed of N_2O_5 . This method assumes that air masses remain relatively stable, and ClNO_2 and NO_3^- were produced through nighttime N_2O_5 heterogeneous uptake. More details on the method are provided elsewhere (Tham et al., 2018; Niu et al., 2022; Phillips et al., 2016). Using the method and selection criteria, we derived $\gamma(\text{N}_2\text{O}_5)$ and $\phi(\text{ClNO}_2)$ during the whole measurement period.

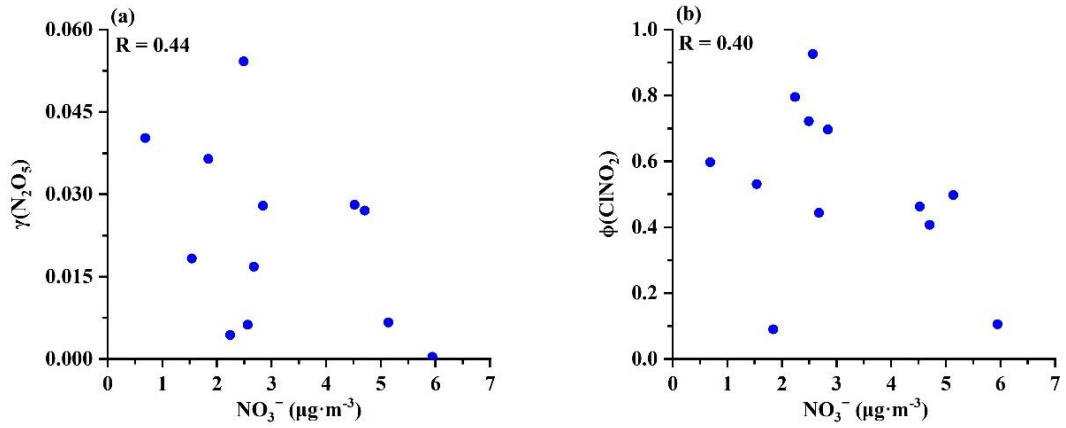


Figure 1. The relationship between field-derived $\gamma(\text{N}_2\text{O}_5)$ (a), $\phi(\text{ClNO}_2)$ (b) and NO_3^- concentrations.

The relative importance of NO_3^- derived from the XGBoost-SHAP result indicated that NO_3^- could play a vital role in affecting the concentrations of ClNO_2 besides N_2O_5 . The high NO_3^- concentrations ($> 3.7 \mu\text{g}\cdot\text{m}^{-3}$) are accompanied by the elevation of ClNO_2 , especially its concentrations reaching $6.2 \mu\text{g}\cdot\text{m}^{-3}$. Previous studies declared that the increased concentrations of NO_3^- decreased $\gamma(\text{N}_2\text{O}_5)$, which would limit the production of ClNO_2 (Wahner et al., 1998; Mentel et al., 1999; Bertram and Thornton, 2009). As depicted in Figure 1, the dependence of $\gamma(\text{N}_2\text{O}_5)$ on NO_3^- concentrations follows the nitrate suppression effect, which subsequently hindered further ClNO_2 formation. Therefore, the importance of nighttime NO_3^- for ClNO_2 levels is that they are co-products from the processes of N_2O_5 heterogeneous uptake. During our field

observation, compared to low NO_3^- conditions, ClNO_2 production was enhanced in high NO_3^- conditions. Especially in late autumn, the increased aerosol surface area and N_2O_5 levels enhanced N_2O_5 uptake, which further promoted both ClNO_2 and NO_3^- production.

To evaluate the contribution of the heterogeneous N_2O_5 uptake to daytime ClNO_2 levels, we calculated ClNO_2 production using Eq. (3), considering the loss of ClNO_2 through photolysis. This method has been employed in a previous study (Tham et al., 2016).

$$\frac{d[\text{ClNO}_2]}{dt} = k(\text{N}_2\text{O}_5)[\text{N}_2\text{O}_5]\phi(\text{ClNO}_2) - J\text{ClNO}_2[\text{ClNO}_2] \quad (3)$$

$$k(\text{N}_2\text{O}_5) = \frac{1}{4}c\text{N}_2\text{O}_5S_a\gamma(\text{N}_2\text{O}_5) \quad (4)$$

We used a $\gamma(\text{N}_2\text{O}_5)$ value of 0.06 and a $\phi(\text{ClNO}_2)$ value of 1.0 in our calculations, which represented upper-end estimates based on previous field studies (McDuffie et al., 2018a; McDuffie et al., 2018b; Tham et al., 2016). However, as shown in Figure 2, a $\phi(\text{ClNO}_2)$ of 1.0 with a $\gamma(\text{N}_2\text{O}_5)$ of 0.06 ($\phi\gamma = 0.06$) fails to reproduce the observed levels of daytime ClNO_2 . A larger $\gamma(\text{N}_2\text{O}_5)$ of 0.11 would be necessary, but such high uptake coefficients and yields are not supported by the current literature. Therefore, we believe that the observed daytime ClNO_2 levels, particularly around noon, cannot be adequately explained by heterogeneous N_2O_5 uptake alone, suggesting the presence of additional sources contributing to the formation of daytime ClNO_2 .

Notably, the laboratory research had confirmed that NO_3^- photolysis can produce ClNO_2 (Dalton et al., 2023). In our study, machine learning analysis, which gauges the “importance” of NO_3^- in affecting daytime ClNO_2 , as well as a linear regression of ClNO_2 against $\text{NO}_3^- \times J\text{NO}_2 \times S_a$, implied that NO_3^- photolysis contributed to daytime ClNO_2 concentrations at our study site. Although NO_3^- photolysis can produce ClNO_2 , this does not necessarily mean that higher photolysis intensity will result in higher ClNO_2 concentrations. It is crucial to understand the dual role of photolysis intensity in determining daytime ClNO_2 levels. Photolysis can contribute to the generation of ClNO_2 by promoting NO_3^- photolysis, while also causing the rapid decomposition of ClNO_2 . As reported in California (Mielke et al., 2013), reduced photolysis rates even

increased daytime ClNO₂ levels by decreasing ClNO₂ loss through photolysis. Additionally, in real atmospheric conditions, several factors beyond photolysis influence NO₃⁻ photolysis, including NO₃⁻ concentrations and particulate chloride levels.

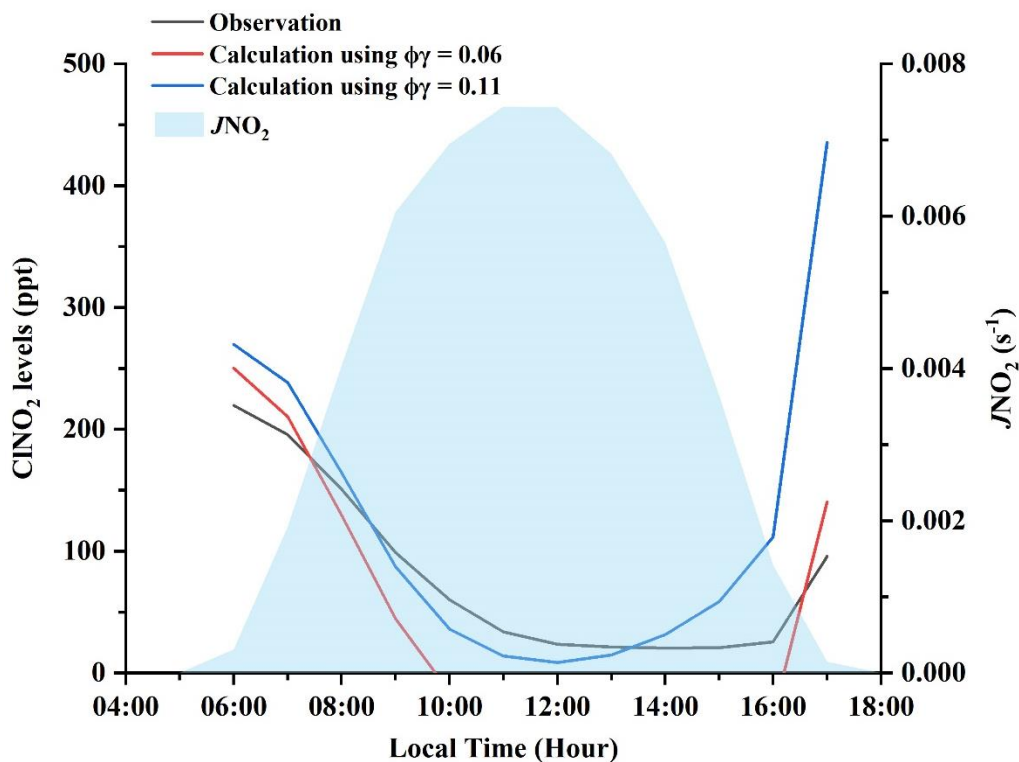


Figure 2. Comparisons of daytime ClNO₂ levels between observation, and calculation using Eq. (4) with a $\phi(\text{ClNO}_2)$ of 1.0 and a $\gamma(\text{N}_2\text{O}_5)$ of 0.06 ($\phi\gamma = 0.06$), or a $\phi(\text{ClNO}_2)$ of 1.0 and a $\gamma(\text{N}_2\text{O}_5)$ of 0.11 ($\phi\gamma = 0.11$).

Added/rewritten: “N₂O₅ concentrations only presented a small peak after sunset, and declined to near the detection limit in the daytime. Previous studies indicated that abundant ClNO₂ could be transport from upper atmosphere or air mass, which contributed to ClNO₂ concentrations in the early morning (Tham et al., 2016; Xia et al., 2021; Jeong et al., 2019). However, the explanations for the concentrations of ClNO₂ around noon remained elusive. To evaluate the contribution of the heterogeneous N₂O₅ uptake to daytime ClNO₂ levels, we calculated ClNO₂ production using Eq. (3), considering the loss of ClNO₂ through photolysis. This method has been employed in a previous study (Tham et al., 2016).

We used a $\gamma(\text{N}_2\text{O}_5)$ value of 0.06 and a $\phi(\text{ClNO}_2)$ value of 1.0 in our calculations, which represent upper-end estimates based on previous field studies (McDuffie et al., 2018a; McDuffie et al., 2018b; Tham et al., 2016). However, as shown in Figure. 2R, a $\phi(\text{ClNO}_2)$ of 1.0 with a $\gamma(\text{N}_2\text{O}_5)$ of 0.06 ($\phi\gamma = 0.06$) fails to reproduce the observed levels of daytime ClNO_2 . A larger $\gamma(\text{N}_2\text{O}_5)$ of 0.11 would be necessary, but such high uptake coefficients and yields are not supported by the current literature. Therefore, we believe that the observed daytime ClNO_2 levels, particularly around noon, cannot be adequately explained by heterogeneous N_2O_5 uptake alone, suggesting the presence of additional sources contributing to the formation of daytime ClNO_2 .”

“The relative importance of NO_3^- derived from the XGBoost-SHAP result indicated that NO_3^- could play a vital role in affecting the concentrations of ClNO_2 besides N_2O_5 . Moreover, according to Figure. 4b, the high NO_3^- concentrations ($> 3.7 \mu\text{g}\cdot\text{m}^{-3}$) are accompanied by the elevation of ClNO_2 , especially its concentrations reaching $6.2 \mu\text{g}\cdot\text{m}^{-3}$. Previous studies declared that increased concentrations of NO_3^- decreased $\gamma(\text{N}_2\text{O}_5)$, which would limit the production of ClNO_2 (Wahner et al., 1998; Mentel et al., 1999; Bertram and Thornton, 2009). As depicted in Figure 1, the dependence of $\gamma(\text{N}_2\text{O}_5)$ on NO_3^- concentrations follows the nitrate suppression effect, which subsequently hindered further ClNO_2 formation. Therefore, the importance of nighttime NO_3^- for ClNO_2 levels is that they are co-products from the processes of N_2O_5 heterogeneous uptake. During our field observation, compared to low NO_3^- conditions, ClNO_2 production was enhanced in high NO_3^- conditions. Especially in late autumn, the increased aerosol surface area and N_2O_5 levels enhanced N_2O_5 uptake, which further promoted both ClNO_2 and NO_3^- production.”

“It is crucial to understand the dual role of photolysis intensity in determining daytime ClNO_2 levels. Photolysis can contribute to the generation of ClNO_2 by promoting NO_3^- photolysis, while also causing the rapid decomposition of ClNO_2 . As reported in California (Mielke et al., 2013), reduced photolysis rates even increased daytime ClNO_2 levels by decreasing ClNO_2 loss through photolysis.”

Technical Comments

Line 76: tenths: tens

Response: Thanks for your comment. We have revised it.

Added/rewritten: “Since Osthoff et al. (2008) firstly detected over 1 ppb of ClNO₂ in the urban outflows of America, significant production of ClNO₂ was widely observed in the polluted coastal and inland areas with abundant anthropogenic emissions and chloride sources, and its concentrations were ranged from tens of ppt to several ppb.”

Figure 3, 5 and 6: Please change the color scale to a colorblind friendly version.

Response: Thanks for your comment. We have changed the color scale in Figure 3, 5 and 6 to a colorblind friendly version. Additionally, due to N₂O₅, NO₃⁻, T, RH, and UV being the most important features of affecting ClNO₂ concentrations, we only compared their relative importance. Therefore, Figure 5 only presents the relative importance of N₂O₅, NO₃⁻, T, RH, and UV.

Added/rewritten:

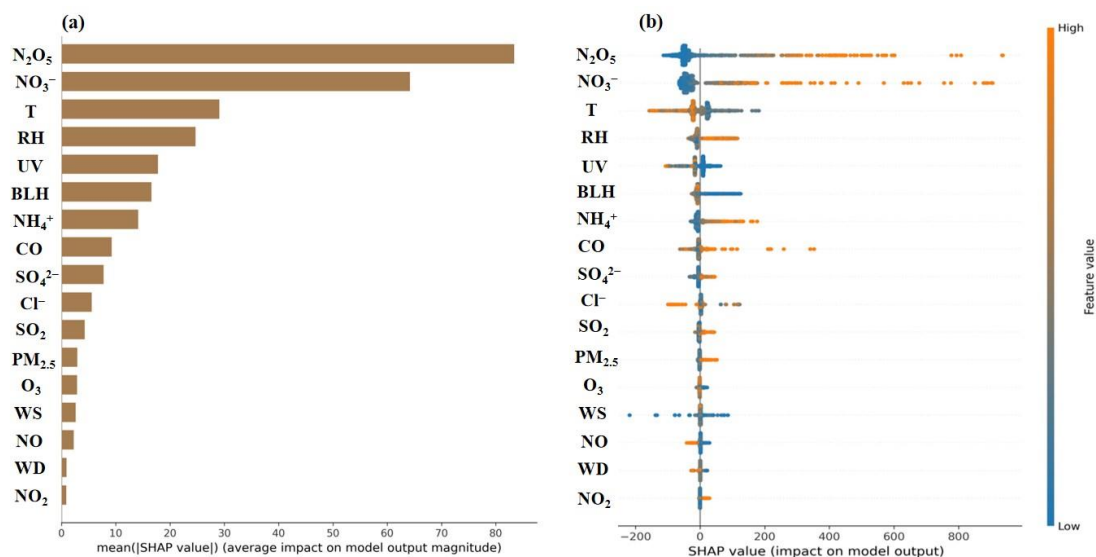


Figure 3. Relative importance of each feature to ClNO₂ using XGBoost-SHAP during the autumn observation period. The mean absolute SHAP value (a), summary plot of SHAP values of each feature (b).

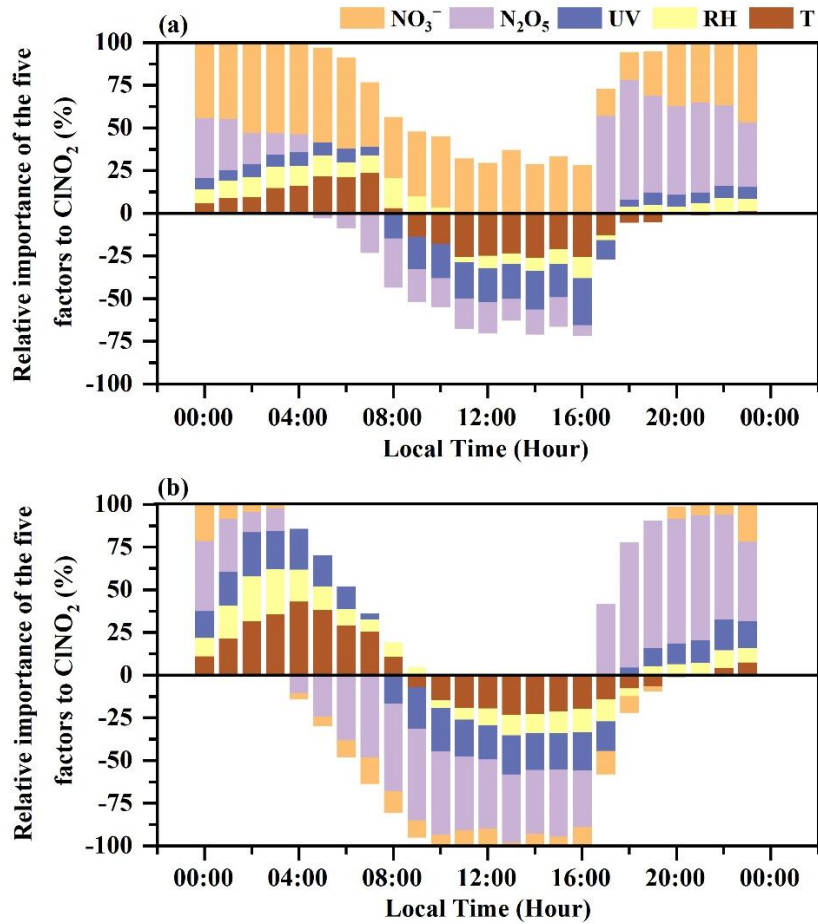


Figure 5. The diurnal variations of the relative importance of the major five factors (including N_2O_5 , NO_3^- , T, RH, and UV) to ClNO_2 based on the SHAP values under the high ($> 3.7 \mu\text{g}\cdot\text{m}^{-3}$) (a) and low ($< 3.7 \mu\text{g}\cdot\text{m}^{-3}$) (b) ClNO_2 concentrations.

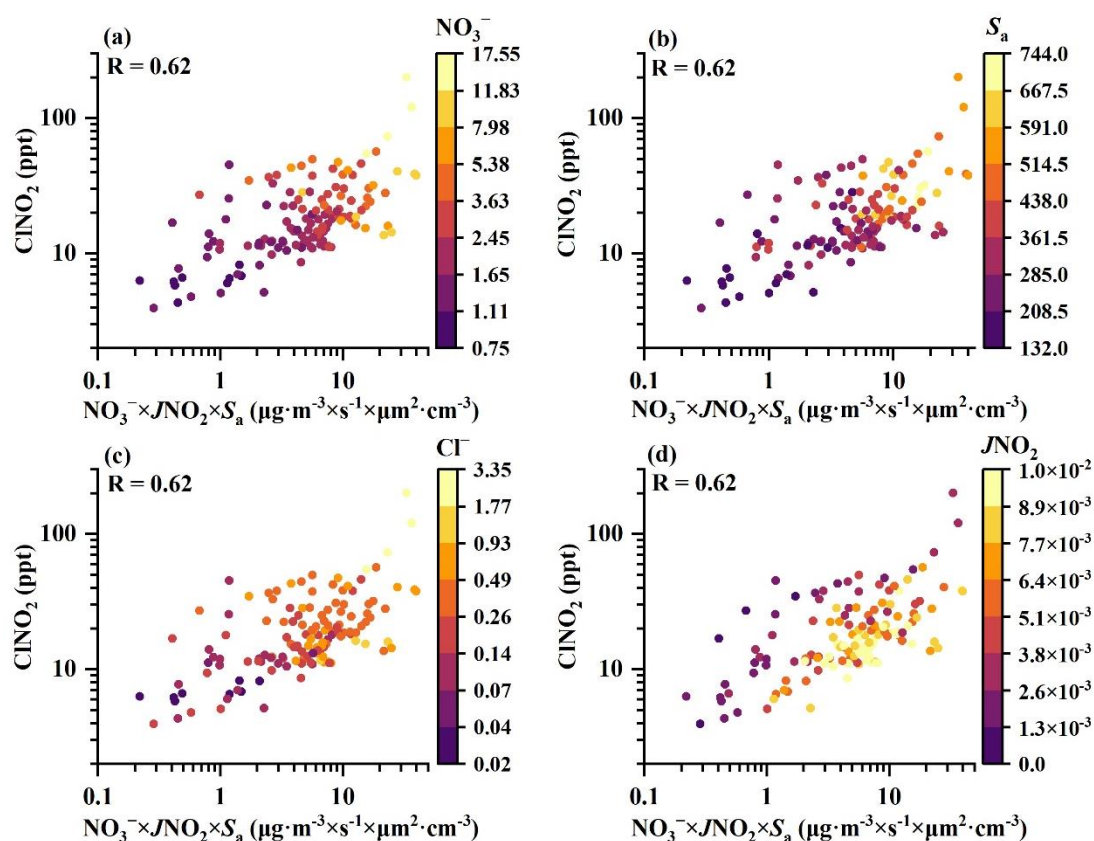


Figure 6. The relationship of daytime ClNO₂ concentrations (12:00-15:00 Local Time) and a proxy of nitrate (NO₃⁻) photolysis (NO₃⁻ × JNO₂ × S_a). The color of the dots denotes the NO₃⁻ (a), S_a (b), Cl⁻ (c), JNO₂ (d), respectively.

Line 215: averagely: average

Response: Thanks for your comment. We have corrected it.

Added/rewritten: “Therefore, the average daily concentrations of NO₃⁻ were classified as high (> 3.7 µg·m⁻³) and low (< 3.7 µg·m⁻³) NO₃⁻ cases to further elucidate the impacts of NO₃⁻ on the formation of ClNO₂.”

Line 224: corrected: correlated

Response: Thanks for your comment. We have revised it.

Added/rewritten: “As depicted in Figure 6, it is observed that daytime ClNO₂ concentrations correlated well (R = 0.62) with the product of a proxy of NO₃⁻ photolysis (NO₃⁻ × JNO₂ × S_a) on aerosol surfaces (S_a), implying that the photolysis of NO₃⁻ contributed to the daytime concentrations of ClNO₂ at our study site.”

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