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 $ClNO_2$ during the daytime by a recently suggested particulate nitrate mechanism. Second, the work discusses the implications for Cl radical production from $ClNO_2$ photolysis.

The first component has significant implications for the understanding of $ClNO_2$ formation globally. However, a discussion of the traditional metrics of $ClNO_2$ formation, the N_2O_5 uptake rate and $ClNO_2$ yield, are completely absent from the paper. Without a discussion on this topic, the authors' conclusion that "NO_3^- photolysis contributed to daytime generation" is severely weakened. In fact, it is based only a machine learning output which gauges the "importance" of NO_3^- influence on $ClNO_2$ as well as a linear regression of $ClNO_2$ with NO_3^- $\times jNO_2 \times$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 $ClNO_2$ cannot be explained by traditional N_2O_5 and $ClNO_2$ 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 N_2O_5 uptake and

$ClNO_2$ 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 N_2O_5 uptake coefficient ($\gamma(N_2O_5)$) and $ClNO_2$ yield ($\phi(ClNO_2)$). Furthermore, we also provided the evidence showing that the daytime observations of $ClNO_2$ cannot be explained by traditional N_2O_5 and $ClNO_2$ 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 $ClNO_2$. The levels of $ClNO_2$ in the box model were constrained by the observed levels of $ClNO_2$ from our field measurements. This approach eliminates the necessity for parameterization of N_2O_5 uptake and $ClNO_2$ yield to determine $ClNO_2$ levels. Therefore, the parametrization for N_2O_5 uptake and $ClNO_2$ yield was not utilized in the box model.

Specific Comments

1. Section 2: A description on the handling of N_2O_5 uptake and $ClNO_2$ 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 N_2O_5 uptake and $ClNO_2$ 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). $ClNO_2$ 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 N_2O_5 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 $ClNO_2$. The levels of $ClNO_2$ in the box model were constrained by observed levels of $ClNO_2$ from our field measurements. This approach negates the need for parameterization of N_2O_5 uptake and $ClNO_2$ yield to determine $ClNO_2$ levels. Therefore, the parametrization for N_2O_5 uptake and $ClNO_2$ yield was not utilized in the box model.

Added/rewritten: "Due to the levels of $ClNO_2$ in the box model determined by observed levels of $ClNO_2$, the parametrization for N_2O_5 uptake and $ClNO_2$ 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 $ClNO_2$ and N_2O_5 measurements were estimated to be ~20 %."

3. Figure 5: What is the interpretation of negative "importance factors"? During the daytime, N_2O_5 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 N_2O_5 during the daytime indicate that the contribution of N_2O_5 chemistry to daytime $ClNO_2$ 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_3xjNO_2xSa is driven by pNO_3xSa 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 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).

$$\gamma(N_2O_5) = \frac{2 \times \left(P(ClNO_2) + P(NO_3^-)\right)}{cN_2O_5S_a[N_2O_5]} \quad (1)$$

$$\phi(\mathrm{ClNO_2}) = 2 \times \left(1 + \frac{P(NO_3^-)}{P(\mathrm{ClNO_2})}\right)^{-1} \quad (2)$$

Here, $P(\mathrm{ClNO_2})$ and $P(NO_3^-)$ represent the production rates of $\mathrm{ClNO_2}$ and NO_3^- induced by N_2O_5 uptake, respectively. S_a denotes the aerosol surface area, and $c(N_2O_5)$ is the mean molecular speed of N_2O_5. This method assumes that air masses remain relatively stable, and $\mathrm{ClNO_2}$ 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 $\gamma(N_2O_5)$ and $\phi(\mathrm{ClNO_2})$ during the whole measurement period.

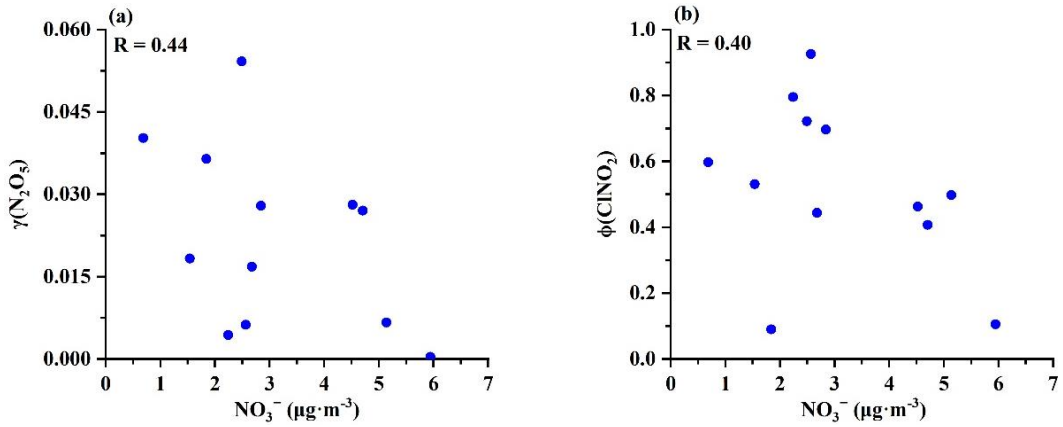

Figure 1. The relationship between field-derived $\gamma(N_2O_5)$ (a), $\phi(\mathrm{ClNO_2})$ (b) and NO_3^- concentrations.

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

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).

$$\frac{d[ClNO_2]}{dt} = k(N_2O_5)[N_2O_5]\phi(\mathrm{ClNO_2}) - JClNO_2[\mathrm{ClNO_2}] \quad (3)$$

$$k(N_2O_5) = \frac{1}{4}cN_2O_5S_a\gamma(N_2O_5) \quad (4)$$

We used a $\gamma(N_2O_5)$ value of 0.06 and a $\phi(ClNO_2)$ 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 $\phi(ClNO_2)$ of 1.0 with a $\gamma(N_2O_5)$ of 0.06 ($\phi\gamma$ = 0.06) fails to reproduce the observed levels of daytime $ClNO_2$. A larger $\gamma(N_2O_5)$ 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 $NO_3^-\times JNO_2\times S_a$, 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 $ClNO_2$ levels by decreasing $ClNO_2$ loss through photolysis. Additionally, in real atmospheric conditions, several factors beyond photolysis influence NO_3^- photolysis, including NO_3^- concentrations and particulate chloride levels.

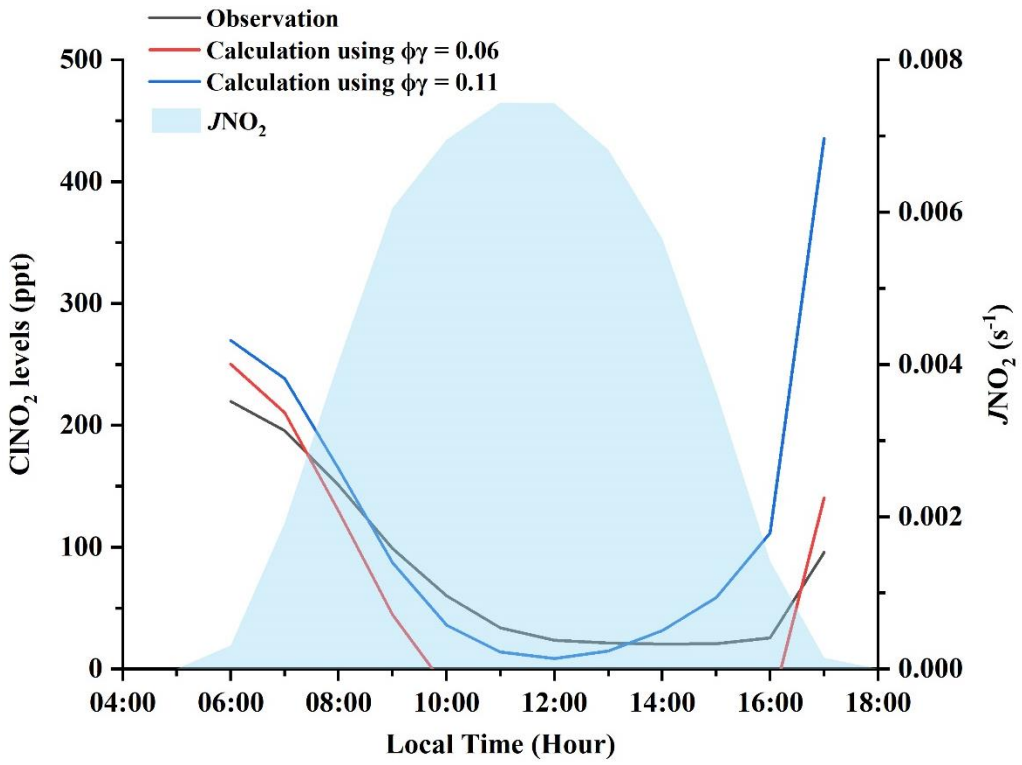

Figure 2. Comparisons of daytime $ClNO_2$ levels between observation, and calculation using Eq. (4) with a $\phi(ClNO_2)$ of 1.0 and a $\gamma(N_2O_5)$ of 0.06 ($\phi\gamma = 0.06$), or a $\phi(ClNO_2)$ of 1.0 and a $\gamma(N_2O_5)$ of 0.11 ($\phi\gamma = 0.11$).

Added/rewritten: "N_2O_5 concentrations only presented a small peak after sunset, and declined to near the detection limit in the daytime. Previous studies indicated that abundant $ClNO_2$ could be transport from upper atmosphere or air mass, which contributed to $ClNO_2$ concentrations in the early morning (Tham et al., 2016; Xia et al., 2021; Jeong et al., 2019). However, the explanations for the concentrations of $ClNO_2$ around noon remained elusive. 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).

We used a $\gamma(N_2O_5)$ value of 0.06 and a $\phi(ClNO_2)$ 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 $\phi(ClNO_2)$ of 1.0 with a $\gamma(N_2O_5)$ of 0.06 ($\phi\gamma$ = 0.06) fails to reproduce the observed levels of daytime $ClNO_2$. A larger $\gamma(N_2O_5)$ 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 $\mu g \cdot m^{-3}$) are accompanied by the elevation of $ClNO_2$, especially its concentrations reaching 6.2 $\mu g \cdot m^{-3}$. Previous studies declared that increased concentrations of NO_3^- decreased $\gamma(N_2O_5)$, 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 $\gamma(N_2O_5)$ 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 $ClNO_2$ in the urban outflows of America, significant production of $ClNO_2$ 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 N_2O_5, NO_3^-, T, RH, and UV being the most important features of affecting $ClNO_2$ concentrations, we only compared their relative importance. Therefore, Figure 5 only presents the relative importance of N_2O_5, NO_3^-, T, RH, and UV.

Added/rewritten:

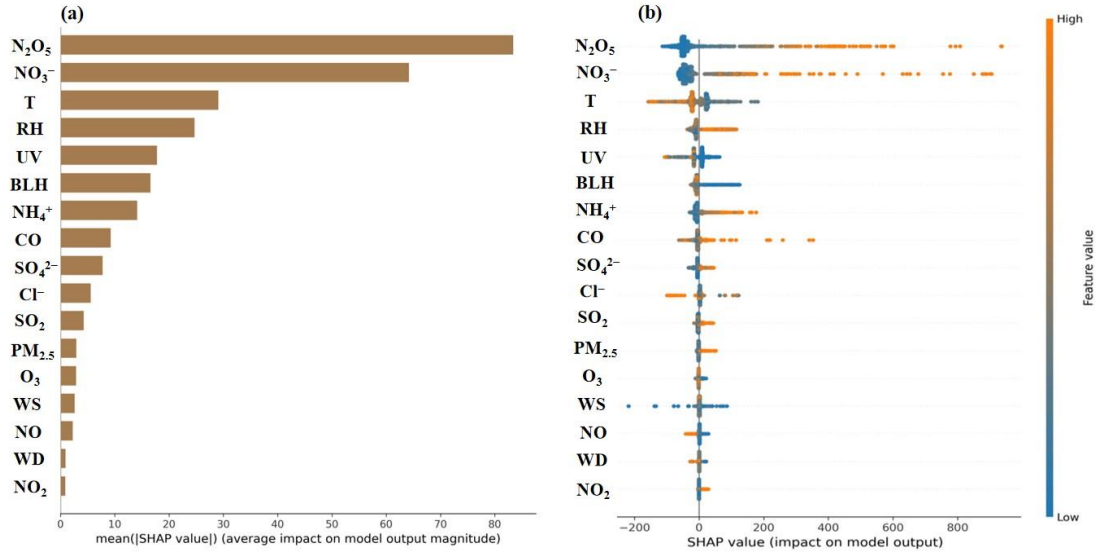

Figure 3. Relative importance of each feature to $ClNO_2$ using XGBoost-SHAP during the autumn observation period. The mean absolute SHAP value (a), summary plot of SHAP values of each feature (b).

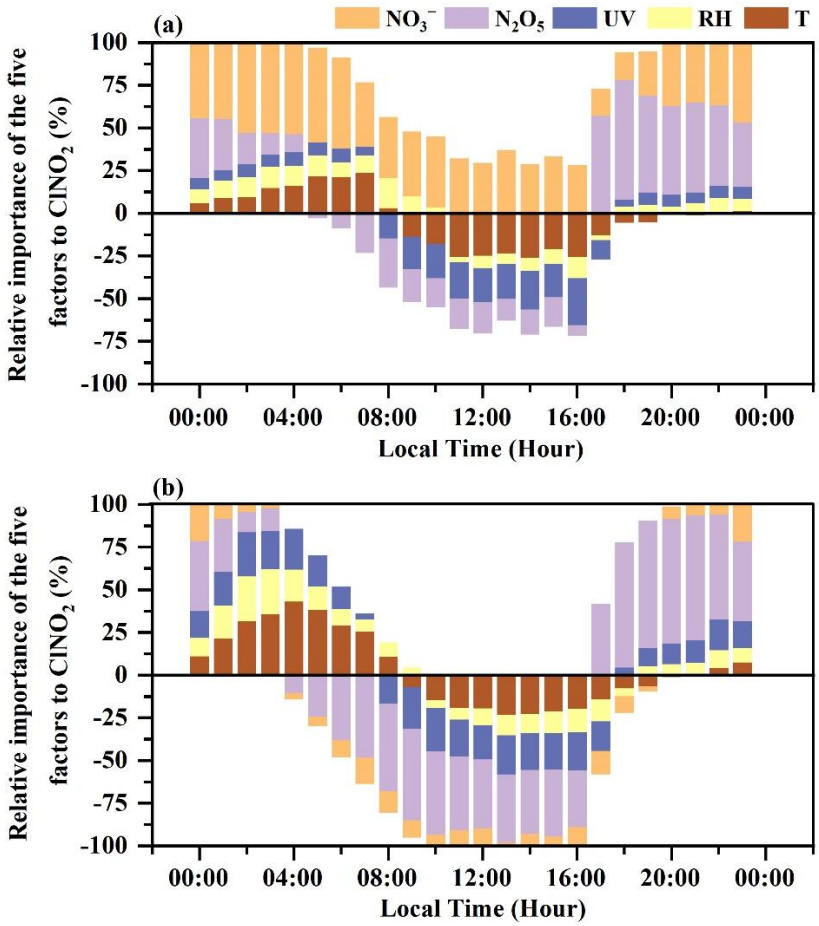

Figure 5. The diurnal variations of the relative importance of the major five factors (including N_2O_5, NO_3^-, T, RH, and UV) to $ClNO_2$ based on the SHAP values under the high (> 3.7 μg·m^{-3}) (a) and low (< 3.7 μg·m^{-3}) (b) $ClNO_2$ concentrations.

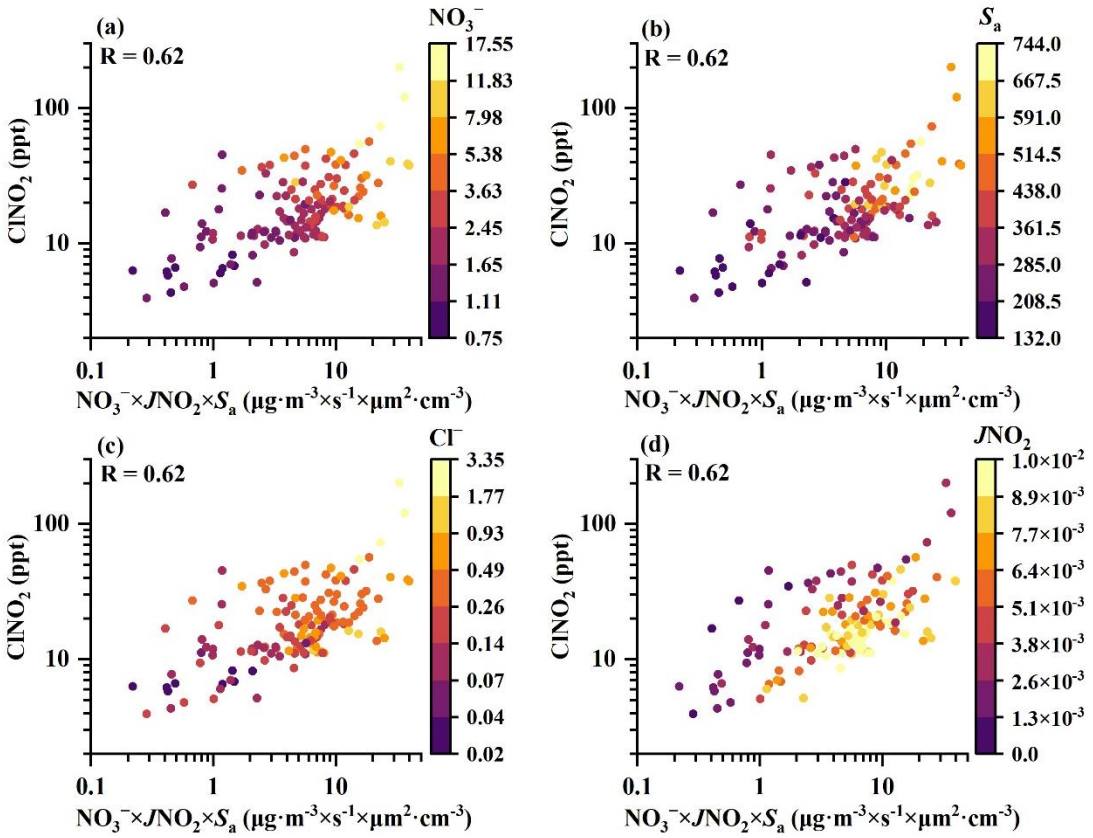

Figure 6. The relationship of daytime $ClNO_2$ concentrations (12:00-15:00 Local Time) and a proxy of nitrate (NO_3^-) photolysis ($NO_3^-\times JNO_2\times S_a$). The color of the dots denotes the NO_3^- (a), S_a (b), Cl^- (c), JNO_2 (d), respectively.

Line 215: averagely: average

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

Added/rewritten: "Therefore, the average daily concentrations of NO_3^- were classified as high (> 3.7 $\mu g\cdot m^{-3}$) and low (< 3.7 $\mu g\cdot m^{-3}$) NO_3^- cases to further elucidate the impacts of NO_3^- on the formation of $ClNO_2$."

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 $ClNO_2$ concentrations correlated well (R = 0.62) with the product of a proxy of NO_3^- photolysis ($NO_3^-\times JNO_2\times S_a$) on aerosol surfaces ($S_a$), implying that the photolysis of NO_3^- contributed to the daytime concentrations of $ClNO_2$ at our study site."

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