A point-by-point response to the reviews

We appreciate the editor and reviewers very much for your positive and constructive comments and suggestions on our manuscript. The followings are our responses to your comments. The comments of the reviewers are shown in black, our responses to the comments are presented in blue, and the new or modified texts are provided in *italics*.

Response to Reviewer #2

Comment 1: The revised manuscript has been greatly improved. There is still one question that I want to ask. After revising, the correlation between $[NO_2]^2 \times [O_3]$ and NOR was plotted in Fig. 3b. However, the correlation of them under RH < 60% condition is very weak. Hence, I think maybe it is not necessary to address the significance of the heterogeneous hydrolysis of N_2O_5 under RH < 60%. Discussion on the results at RH > 60% in detail is enough.

Answer: Thank you for your positive evaluation of our work and your valuable suggestion. These relevant sentences have been modified and deleted in the revised manuscript as following:

"As shown in Figure 3b, although the variations of $[NO_2]^2 \times [O_3]$ at the nighttime (18:00-7:00) were all positively correlated with NOR under the three different RH conditions, ~~and~~ their correlation under the RH ≥ 60% condition (R^2 = 0.552) was significantly stronger than those under the RH < 60% condition (R^2 ≤ 0.181). It has been acknowledged that the correlation between two species means the impact of changes in one species on another. The stronger the correlation is, the greater the impact is. Therefore, ~~the positive correlations between NOR and $[NO_2]^2 \times [O_3]$ indicated that the heterogeneous hydrolysis of N_2O_5 could contribute to the formation of atmospheric nitrate at the nighttime under different RH conditions. The~~ the significantly stronger correlations between NOR and $[NO_2]^2 \times [O_3]$ under the RH ≥ 60% condition than under the RH < 60% condition revealed that the heterogeneous hydrolysis of N_2O_5 made a remarkable contribution to atmospheric nitrate at the nighttime under high RH condition."

Comment 2: In addition, line 599-601 "Considering that the formation of atmospheric NO_3 radical is mainly via the oxidation of NO_2 by O_3, the heterogeneous hydrolysis of N_2O_5 occurs only at high O_3 and NO_2 levels during the nighttime (He et al., 2018; Wang et al., 2018b)." This sentence is a little incompatible with the result in Fig 3b. Because in Fig. 3b, the highest concentration of NO_2 and O_3 was shown at RH < 30% and the lowest concentration of them was shown at RH > 60%. However, the contribution of heterogeneous hydrolysis of N_2O_5 to nitrate was not consistent with that result. Hence, it is better to rethink the way of expression.

Answer: Special thanks to you for your good comment. This sentence has been changed in the revised manuscript as following:

"Considering that ~~the formation of~~ atmospheric NO_3 radical is mainly generated via the oxidation of NO_2 by O_3, the relatively high O_3 and NO_2 levels could be in favor of the ~~heterogeneous hydrolysis~~formation of N_2O_5 ~~occurs only at high O_3 and NO_2 levels~~ during the nighttime (He et al.,

2018;Wang et al., 2018b)~~. Therefore~~, and hence the correlation between $[NO_2]^2 \times [O_3]$ and NOR can represent roughly the contribution of the heterogeneous hydrolysis of N_2O_5 to atmospheric nitrate at night."