In this manuscript, Wang et al. estimated particulate nitrate formation from N_2O_5 uptake at air aloft (~100 m) over urban Beijing, based on the vertical measurements of ozone, NO, and NO_2 on one episode day obtained at a tower and box model calculations. With a number of assumptions, the model suggested significant particle nitrate formation of up to 50 μg m^{-3} from the N_2O_5 heterogeneous hydrolysis in air masses aloft for this episode. The model calculations also suggested that the oxidation of NO_x to nitrate was maximized once N_2O_5 uptake coefficient was over 0.0017, and became insensitive with higher uptake coefficient.

The topic of the heterogeneous process of NO_x is of interest to the community, and the vertical measurements of chemical composition in winter haze are particularly valuable. My main concern, however, on the present work is that the model was poorly constrained by observations and had to reply on too many assumptions. The vertical measurements only included O_3, NO, and NO_2 which enabled calculation of NO_3 production, but several key parameters, such as N_2O_5, VOCs and aerosol surface area density, for loss of NO_3, N_2O_5 production and subsequent loss to nitrate were not measured, making it very difficult, if not impossible, to evaluate nighttime reactions of NO_x and for nitrate formation. In addition, the analysis was only based on one profile measured in the early evening, and too many assumptions in the model calculations were not well justified. All these make it very difficult to judge the validity of the conclusions drawn from the analysis. The authors are advised to carefully consider and reduce these uncertainties which could lead to large bias and possible errors.

Specific comments:

L16. Please elaborate what the simultaneous measurements were conducted.

L22. Please define the potential of pNO_3^-.

L57-58. It is not clear how the N_2O_5 uptake coefficient in winter will be different from summer.

L96. the abbreviation of ‘IAP’ should be spelled out upon the first mention in the text.

L110. Please specify which instruments were installed on board a movable cabin.

L111-113. Describe in more detail about the two light-weight instruments for vertical measurements of O_3, NO and NO_2. How were they calibrated, was there any intercomparison with conventional monitors?

L124. Were the daily cycles conducted in the same time periods for every day?

L129-131. The assumptions of no NO influence, no physical mixing, and no transport of the air mass may not be valid here. Without continuous or intermittent measurement as constraints, it is difficult to know the evolution of the air masses, as such one cannot testify the validity of these assumptions.

L143-147, The low theoretical equilibrium ratio of NO_3 to N_2O_5 at the low-temperature condition may not necessarily mean that the N_2O_5 formation dominates the NO_3 loss. More evidence is needed here. The k_{NO3} value of 0.02 s^{-1} assumed in the present study was much higher than other studies, for example, Brown et al., 2016, in which the NO_3 reaction with VOC contributes more than half of the total NO_3 loss.

L148-150. It is not clear how the author determined the initial concentration of NO_2 and O_3, which could affect the integrated concentrations. Were you using the iterative method suggested by Wagner et al., 2013? The retrieved results should be included in the supplementary. Any measurement constraints were used to validate this calculation?

L146. “than” should be “that”.

L162. Please clarify the exact time period for the model running from sunset to sunrise.

L164-168. There are some flaws in the Eq.1 to 5 of the box model. To simplify the differential equation, the author assumes an equilibrium between NO_3 and N_2O_5 in Eq. 3 and 4, which means the loss rate of NO_2 through R_2 and production rate from R_3 should be equal. Therefore, the NO_2 loss rate will equal to the reaction rate of R1, which contradicts Eq. 2. As suggested by Wagner et al., 2013, “the assumption of equilibrium leads to an error which accumulates as the equations are integrated”. This could affect the results when retrieving the initial NO_2 concentration and subsequent model simulation. To be more accurate, I suggest the author use the explicit equations suggested by Wagner et al., 2013.

L169-L179. The surface area was calculated based on the measurement of particle size distribution from 0.01 to 0.6 μm and could be underestimated due to the lack of information of larger particles, resulting in large uncertainty in the calculated uptake coefficients. It is necessary to provide an uncertainty estimation of how much this will affect the results.

L240-241, L245-247, L257-258. The author attributed the lower O_x level at high altitude on the night of December 19 to missing sinks of O_x with high N_2O_5 uptake, but it could also be a result of the continuous emissions of NOx near the ground leading to accumulation of NO_2 within the nocturnal boundary layer with a height around 100m. So the O_x level in the residual layer and surface doesn't have to be conserved. Here it would be good to show vertical information on meteorological parameters.

L264-265, Wrong figure number referenced here.

L265. Please explicitly define the equation used for calculating the nitrate accumulation.

L262-267. This model simulation assumes an ideal condition with no NO concentration above 150m from the sunset to 21:00, which cannot be substantiated. Thus the calculated accumulation of nitrate is questionable.

L281-282. Please elaborate how to deal with calculations of mixing.

L282-285. If the author’s hypothesis is true, it should be able to observe a sharp increase of particulate nitrate at the ground site in the morning of December 20. Any evidence on that?

L309. Eq.6 is incorrect. The particle nitrate formation is twice of N_2O_5 loss if assuming $ClNO_2$ yield is zero.

L328, “coral” should be “coal”.

L342-343. A possible reason for the small difference on k_{NO3} variation could be the NO_3 change (via VOC loss) were unaccounted for in Eq.2 that used to retrieve the initial concentration. Comparison between the full differential equations and simplified calculation is required to validate the results.

L351, delete “to be zero”.

L357, add “that” after “found”.