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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The promotion effect of nitrous acid on aerosol formation in wintertime Beijing: possible contribution of traffic-related emission" *by* Yongchun Liu et al.

Anonymous Referee #2

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This paper reports the possible promotion effect of nitrous acid (HONO) on the formation of secondary organic aerosol (SOA) and nitrate in winter based on a five-month comprehensive observation in urban Beijing. Evidence for the relationship between secondary aerosol formation and the consumed HONO was obtained from the observations. The detailed budget of ambient HONO was explored, and vehicle emission was proposed as a significant source of HONO in Beijing. Overall, the manuscript is logically organized and well written, and the measurement data are much valuable. I would like to recommend that it can be considered for publication after the following major and specific comments being properly addressed.

Major Comments:

Section 2.2. HONO budget calculation: the description of the budget calculation is not clear enough. Firstly, the method used in this study was the budget analysis, other than the stationary state analysis. For a thorough budget analysis, physical terms (such as vertical and horizontal transport) should be considered for equations (1) and (12). At least, the authors need to evaluate if the physical processes were negligible for the analysis presented here. Secondly, a number of parameters (e.g., Fhono, Coh, gamma, Yhono, Jnitrate, Jhono, etc.) are required for the analysis. It is not clear how these parameters were obtained or approximated in this study. Although they were described more or less in the following Section 3.3, the authors may need clearly state the data source and uncertainties of these key parameters at their first appearance. This may help the readers better understand the overall methodology of this study.

Section 3.3 and Figures 3&4: following the first comment, the description and discussion of the HONO budget are also not clear and need clarification. The contributions of heterogeneous reactions of NO2 on aerosol and ground surfaces were too low, and they were even lower than the OH+NO reactions during the nighttime. This is unusual. Is it reasonable to approximate the nighttime OH concentrations linearly with the temperature? Furthermore, the heterogeneous reactions of NO2 on aerosol and ground surfaces were highly dependent on the NO2 concentrations, the uptake coefficients, and surface density, some of which are highly uncertain. It is not clear what values were actually adopted for the uptake coefficients of NO2 on aerosol and ground surfaces, and how much were the ambient NO2 levels and surface density? More details about the calculation of HONO budget are needed.

The authors attempted to quantify the contributions of vehicle-emitted NO to ambient HONO via NO+OH reaction based on the source apportionment of NO emissions. This is not convincing because the homogeneous HONO formation is generally limited by OH other than NO. This means that the produced HONO should be not linearly dependent to the NO emissions. It can be concluded that vehicle emission should

contribute significantly to not only direct HONO emission but also HONO formation through reactions of NO and NO2. However, the current quantification analysis needs be more careful.

Specific Comments:

Line 44: fine particulate matter with diameter less than or equal to 2.5. . .

Line 78: on polluted days

Lines 84-94: to my knowledge, there have been a number of observational studies of HONO in recent years in China, and similar HONO budget analyses were performed. I suggest the authors to comprehensively review the existing results about the HONO sources in China and compare them against the source analysis results obtained in the present study.

Line 138: replace “nitrous acid” by “nitric acid”

Line 146: Equation (1) describes the budget analysis other than the stationary state analysis. Both methods are different. Transport terms need be considered here.

Lines 198-202 and Fig. 1: it would be much better if the authors could also plot the other related parameters, such as NOx and meteorological parameters, in Fig. 1. It is difficult for the readers to look at the same measurements separately from main text and supplement.

Line 228: the increase in temperature. . .

Line 262: what does “in RO2 chemistry” mean? Rephrase this sentence.

Line 266: replace “dominating” by “dominant”

Lines 270-282: it is not clear how the POH-HONO and POH-O3 were calculated from the current discussion. The calculated POH-HONO and POH-O3 levels in winter and April-June seem to be too high. Detailed calculation methods should be given here.

Usually, the OH+NO reactions should be subtracted from the photolysis of HONO to denote the real contribution of HONO to the OH source.

Line 291: photolysis of HCHO is actually the primary source of HO2.

Line 330: delete “and”.

Lines 333-336: Several recent papers about the nitrate aerosol trend and formation mechanisms in China are highly relevant to this study, and should be acknowledged. Wen et al., Summertime fine particulate nitrate pollution in the North China Plain: increasing trends, formation mechanisms, and implications for control policy, Atmospheric Chemistry and Physics, 18, 11261-11275, 2018.

Sun et al., Two years of online measurement of fine particulate nitrate in the western Yangtze River Delta: influences of thermodynamics and N2O5 hydrolysis, Atmospheric Chemistry and Physics, 18, 17177-17190, 2018.

Line 363: high or low HONO concentration?

Line 408: Tan et al., (2019)

Line 407-408: it would be better if the authors could provide the estimated OH levels here.

Lines 430-431: it is not clear how the photolysis frequency of nitrate was corrected? Details are needed here. What are the J values used finally?

Line 553-554: again, it is budget analysis other than stationary state calculation.

Line 568: indirect production

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