Response to interactive comments on

"High resolution vertical distribution and sources of HONO and NO$_2$ in the nocturnal boundary layer in urban Beijing, China"

by Jörg Kleffmann.

Fanhao Meng and Min Qin

<u>**General comments:**</u>

The gradient study presented in Meng et al. is a nice piece of work showing night-time gradient data on a meteorological tower up to 250 m altitude similar to our former study (Kleffmann et al., 2003). Here a measurement container is lifted on the side wiring of the tower, far away from the open steel construction of the tower, thus minimizing potential wall effects, which is a nice idea. I strongly encourage the authors to extent these measurements including other important species for the future.

Response: We would like to thank Jörg Kleffmann for his time and efforts in preparing this detailed and constructive comments. We have revised the manuscript accordingly to address his concerns. Here are our point-to-point response to Jörg Kleffmann's specific comments.

<u>**Major Comments:**</u>

1. Direct HONO emissions (section 3.3):

The authors determined the HONO/NO$_x$ emission ratio from the minimum HONO/NO$_x$ ratio, which by definition will only result in an upper limit value, since always some secondary HONO is included in ambient measurement data. If emission ratios should be derived from field data, the authors should look for sharp peaks during night-time and should evaluate only the elevations of HONO and NO$_x$ over the background levels (DHONO/DNO$_x$). In this case the risk overestimating direct emissions is minimized but still even this peak data presents an upper limit caused by potential secondary HONO formation during transport from the emission to the measurement site (will be short for sharp concentration peaks). I expect that the HONO/NO$_x$ emission ratio derived by the peak method will be considerably lower, closer to direct emission data (see test stands and tunnel data: <1%). The use of correct HONO emission data is important for the whole study, since emission corrected HONO data (considerable ca. 50% contribution, typically emissions have 10-20 % contribution) is used later for the evaluation of the other HONO sources.

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Response: Thank you very much for your valuable comments and suggestions. The emission factors have been derived from tunnel measurements in previous studies (Kirchstetter et al., 1996; Kurtenbach et al., 2001; Laing et al., 2017). However, considering the differences in vehicle types, fuel compositions, and other factors, the previously reported emission ratios might not be representative for Beijing region. In order to obtain the local emission factor, the $HONO/NO_x$ ratio was derived from field data. Following Jörg Kleffmann's suggestion, five criteria were applied to ensure as much of the freshly emitted air masses as possible: (a) only nighttime data (from 18:00 LT to next 6:00 LT) were included to avoid the fast photolysis of HONO; (b) only sharp peaks during nighttime and the elevations of HONO and NO_x over the background levels were estimated; (c) $\Delta NO/\Delta NO_x > 0.80$; (d) good correlation between HONO and NO_x; (d) short duration of the plume (< 30 min). The typical nocturnal wind speed at measurement site was 1.2 m s^{-1}, thus the duration for fresh air masses should have been less 30 min during transport from the emission to the measurement site. Criteria (b), (c), (d) and (e) were used as indicators for identifying fresh vehicular emissions.

For a month field observation, 11 fresh plumes satisfied selection criteria (Table 1). Two examples of emission plumes selected based on the preceding criteria are shown in Fig. R1.
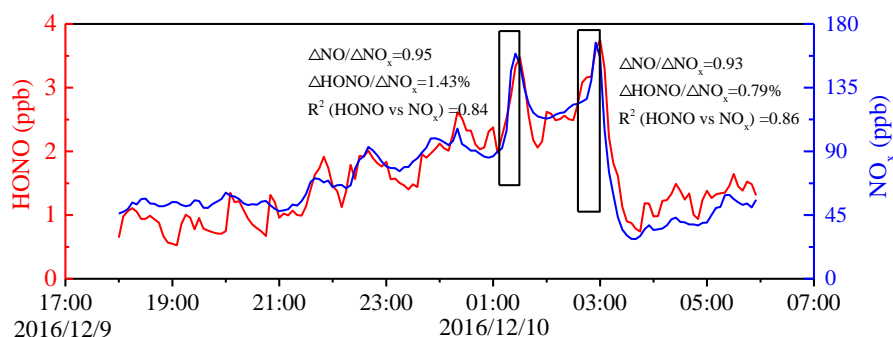

Figure R1. Temporal variation of nocturnal HONO and NO_x on December 9th to 10th, 2016. The HONO emission ratios were estimated using data collected in the black frame.

The derived $HONO/NO_x$ ratios vary from 0.78% to 1.73%, with an average value of 1.28% ± 0.36%, which was larger than the 0.53%–0.8% measured in the tunnel in Wuppertal (Kurtenbach et al., 2001), but in the range of 0.19%–2.1% reported by previous studies (Kirchstetter et al., 1996; Kurtenbach et al., 2001; Su et al., 2008; Rappengluck et al., 2013; Yang et al., 2014; Xu et al., 2015; Liang et al., 2017; Zhang et al., 2018; Li et al., 2018). Comparison of derived $HONO/NO_x$ ratios with previously reported results are summarized Table S2 in the supplementary information. To minimize the risk of

overestimating the direct HONO emissions, the minimum HONO/NO$_x$ ratio of 0.78% was used as an upper limit for the emission factor (Su et al., 2008). The direct emissions contributed an average of 29.3% ± 12.4% to the ambient HONO levels at night, with an average HONO$_{emis}$/HONO value of 35.9% ± 11.8% during the clean episode and an average HONO$_{emis}$/HONO value of 26% ± 11.3% during the haze episode. The lower vehicle emissions contribution during haze episode could be caused by an odd-even car ban, which required alternate driving days for cars with even- and odd-numbered license plates.

2. HONO formation on aerosol surfaces (section 3.4.2).

The authors used heterogeneous uptake data for NO$_2$ derived in the laboratory to calculate potential HONO formation during night-time. However some studies cited/considered focus on the daytime production of HONO by photosensitized conversion (George et al., Sremmler et al.), leading to overestimation of the night-time conversion. Here references to other dark studies are recommended and it should be mentioned that the used value of 10ˆ5 represents really the upper limit night-time kinetics (typically 10ˆ-6 is used in the dark). In addition, the authors should explain that they considered a 100 % yield for the NO$_2$ conversion (see factor $\frac{1}{4}$ in eq. (1)), which is also the upper limit, e.g. when using the typically considered night-time reaction R1: 2NO$_2$+H$_2$O=>HONO+HNO$_3$ (see the first reference used in line 370) for which the maximum yield is 50 %. So please define that a redox reaction (NO$_2$+X=>HONO) is considered here. But all this will even further decrease the low contribution of particle surfaces to the night-time HONO production, which is in line with my own point of view.

Response: Thanks for the comment and suggestion. The references (George et al., 2005; Stemmler et al., 2007) have been changed to the following one:

Saastad, O. W., Ellermann, T., and Nielsen, C., J.: On the adsorption of NO and NO$_2$ on cold H2O/H2SO4 surfaces, Geophys. Res. Lett., 20, 1191-1193, https://doi.org/10.1029/93GL01621, 1993.

Following the referee #1's suggestion, the NO$_2$ uptake coefficient of 1×10^{-5} to 1×10^{-6} in the dark were used to estimate the nighttime production of HONO from heterogeneous conversion of NO$_2$ on aerosol surface. The yield for the NO$_2$ conversion and night-time reaction have been stated in the revised manuscript as suggested. A typical heterogeneous reaction (2NO$_{2(g)}$+H$_2$O$_{(ads)}$

$\xrightarrow{surface}$ HONO$_{(g)}$+HNO$_{3(ads)}$) and the maximum yield of 50% were considered in evaluating the HONO production on aerosols, the yield of hydrolysis reaction assumes that HONO and HNO$_3$ are formed by equimolar disproportionation of two NO$_2$ molecules and immediately release HONO (Finlayson-Pitts et al., 2003; Finlayson-Pitts, 2009). The reactive uptake of NO$_2$ by the aerosol was assumed to occur on all measured aerosol surface area, regardless of chemical composition. The production of HONO on aerosol surface can be estimated using the equation of Ye et al. (2018) modified to account for the disproportionation.

The newly obtained aerosol surface areas at ground and at 260 m, which was corrected to ambient aerosol surface area (S_{aw}) for particle hygroscopicity via a growth factor (Liu et al., 2013; Wang et al., 2018), were used to estimate the nocturnal HONO production from heterogeneous reaction of NO$_2$ on aerosol surface. An estimate of HONO production on aerosols was made using an average observed S_{aw} of 2314 µm^2 cm^{-3} between 22 and 01 h (the vertical measurement periods on December 11th) and an upper limit of observed NO$_2$ of 54 ppb from the residual layer. The HONO production of 30-300 ppt in an interval of 1.5 h could account for HONO increases of 15-368 ppt in the residual layer between profile measurements during E3. Therefore, the production of HONO solely on aerosols can explain the HONO observations during the haze episode, implying that the aerosols plays an important role in nocturnal production of HONO during the haze episode. The same analysis was discussed for other vertical measurements during C2. The results indicated that the ground surface dominated HONO production by the heterogeneous uptake of NO$_2$ during the clean episode.

3. Nocturnal HONO production on the ground (section 3.4.3):

Instead of using the "two-point" equation (4), simply plot the night-time HONO/NO$_2$ ratio as a function of the time. In this case the slope determined by linear regression will statistically better describe the efficient first order NO$_2$=>HONO conversion rate constant. Since this is a rate coefficient, it should be better termed by e.g. "k(het)" and not by the term "C" (C: concentration?), which I often found in recent Chinese HONO papers? Since HONO(corr.) will be significantly higher (see point 1) also the efficient conversion rate coefficient will increase.

In this section the authors also determine a net HONO yield from the ground surface conversion of

NO₂ of the order of 10 % by using deposition velocities for HONO and NO₂. The value for HONO is to my opinion too low and will be more of the order of 2 cm/s (see cited studies and others). In addition please specify the value used for NO₂. The HONO yield determined is quite high and our recent gradient study (Laufs et al., Atmos. Chem. Phys., 17, 6907–6923, 2017) could be also cited, where we determined lower values in the range of 0.02-0.044 by a more direct approach in good agreement with the gradient study by Stutz et al., 2002 (0.03). In addition you will find there also data for the deposition velocity of HONO, confirming a higher value, at least at the low temperatures of the present study.

Response: The term "C" is first letter of the conversion, and we used the term "k_{HONO}" for conversion frequency as suggested. The conversion frequency (k_{HONO}) has been recalculated following the method introduced by Alicke et al. (2002). The local emission factor of 0.78% derived from field data was used to correct HONO concentration (i.e. $[HONO]_{corr} = [HONO] - [NO_x] \times 0.0078$). Since NO was not measured at ground level after 14:00 on December 10th, the average HONO$_{emis}$/HONO ratio of 35.9% ± 11.8% and 26% ± 11.3% were used to correct HONO observations during the clean and the haze episodes, respectively (i.e. $[HONO]_{corr} = [HONO] - [HONO]_{emis}$). The conversion frequencies, k_{HONO}, on December 9th, 10th, and 11th were 0.0082 h^{-1}, 0.0060 h^{-1}, and 0.0114 h^{-1}, respectively.

As Jörg Kleffmann suggested, the temperature-dependent deposition velocity of HONO ($V(HONO)_T = \exp(23920/T - 91.5)$) was used to estimate the dry deposition velocity of HONO (Laufs et al., 2017). The average $V_{dep,HONO}$ calculated from nocturnal observations (00:00–06:00 LT) was 1.8 cm s^{-1}, with a range of values spanning 0.9 to 3 cm s^{-1}, which was within the range of previously reported values between 0.077 and 3 cm s^{-1} (Harrison and Kitto, 1994; Harrison et al., 1996; Spindler et al., 1998; Stutz et al., 2002; Coe and Gallagher, 1992; Laufs et al., 2017). The NO₂ deposition velocity (V_{dep,NO_2}) of 0.07 cm s^{-1} (VandenBoer et al., 2013) used in calculation has been specified in the revised manuscript. Following the suggestion by the referee #1 and #3, we have significantly revised the discussion in section 3.4.3. A net HONO yield from the surface heterogeneous conversion of NO₂ has been removed in the revised manuscript. The surface production rate of HONO was calculated to compare with other HONO sources.

4. Concept used:

The authors tried to distinguish between heterogeneous night-time formation of HONO on particle surfaces and on the ground, which is an important question with respect to the decades long discussion on the sources of HONO in the atmosphere. Here first, I am missing a more focused introduction of the basic problem. There are ground level studies, which found a nice correlation of HONO/NO$_2$ with the particle surface area and which propose HONO formation on particles. On the other hand there are gradient studies of HONO (and NO$_2$) which typically determine a negative gradient, pointing to the ground as most important surface. However, none of these studies can answer the question! The correlation with particles could be also explained by the variation of the vertical mixing and the fact that both, HONO and particles are emitted/formed near the ground. Here pioneering studies by A. Febo found nice correlation of HONO with Radon during night-time and there is clearly no chemical link between both species. On the other hand the gradients could be also explained by formation on particle surfaces, if there is also a negative gradient in the particle surface area density (S/V). So what we need are gradient measurements over a few hundred meters (like the present study) but including besides HONO and its precursors (most probably NO$_2$) also the surface area density (see our former study Kleffmann et al., 2003). Only if there is no gradient in the particles, a negative gradient of the HONO/NO$_2$ ratio will show HONO formation on the ground. Thus, second, I strongly recommend that the authors use this nice tower set-up and include in future campaigns also fast particles and NO measurements!

The later is important since the titration reaction of NO+O$_3$ may mask the observation leading e.g. to artificial gradients of the HONO/NO$_2$ ratio not resulting by any HONO processes, but simply by a changing NO$_2$/NO$_x$ ratio. For example, the decreasing HONO/NO$_2$ ratio shown in Figure S4 (see first two gradients near the ground) is not plausible since the HONO/NO$_x$ ratio is expected to increase during the night. Most probably NO was converted into NO$_2$ leading to lower HONO/NO$_2$ ratio.

Response: We would like to thank Jörg Kleffmann for his introduction to the basic problem of heterogeneous nighttime formation of HONO and his constructive suggestions. The primary reaction surfaces for the nocturnal heterogeneous production of HONO is still controversial, and the role of the aerosols in the heterogeneous production of HONO remains an open question. In this study, we try to explain this problem by investigating the high-resolution vertical profiles of HONO at night. The aerosols as an important nocturnal source of HONO during haze episode was observed. However, this campaign only includes the limited vertical measurement data (only HONO and NO$_2$ vertical profiles).

As Jörg Kleffmann said, the vertical measurements including besides HONO and NO$_2$ also the aerosol surface areas and NO are needed to investigate the HONO chemistry, while the gradient measurement based on the nice tower set-up provides the basis for our future research. A more comprehensive vertical measurement campaign will perfomed in the future, including not only the vertical profiles of HONO and NO$_2$, but also the aerosol surface area and NO measurement.

Minor Comments:

1. There are numerous errors in the references, please check, examples: Line 60: Vogel et al.; missing blanks between the references (throughout the whole manuscript). Line 68: Bröske et al., etc.

Response: As suggested, we have checked and corrected the references in the revised manuscript.

2. Line 76: the photocatalytic conversion of NO$_2$ on TiO$_2$ ("mineral dust") is not a redox reaction.

Response: Following the suggestion by the referee #3, the introduction section has been revised. This statement has been removed in the revised manuscript.

3. Line 77-82, R2: This reaction is not a photosensitized conversion.

Response: Thanks for the comment. This reaction (R2) is a heterogeneous reaction, and we adjusted it to a more suitable position (line 95).

4. Lines 84-88: In the cited studies neither all used a DOAS, nor was the DOAS installed on an elevator.

Response: The statements and cited references have been revised in this paragraph, as below:

Lines 122–128:

"Vertical gradient observations provide evidence regarding surfaces and in situ HONO formation, which can help to understand the nighttime HONO sources. Methods of long-path differential optical absorption spectroscopy (LP-DOAS) (Stutz et al., 2002; Wong et al., 2011; Wong et al., 2012), instruments mounted on a movable elevator of a tall tower or a fixed height on a building (Kleffmann et al., 2003; VandenBoer et al., 2013; Villena et al., 2011) and aircraft measurements (Zhang et al., 2009; Li et al., 2014; Ye et al., 2018) have been applied for HONO vertical gradient observations in Europe and the America."

5. Line 93-94: In our gradient study in Chile the measurement frequency was very high, only the

vertical resolution was low (should be "or").

Response: Revision has made as Jörg Kleffmann suggested.

6. Line 106-110: Please add our recent flux study Laufs et al., 2017 (see above).

Response: The introduction section of the manuscript has been revised accordingly to address the referee's comments. This paragraph (lines 163–170) have been removed in the revised manuscript, but the flux literature has been added to other parts of the manuscript.

7. Lines 381-384, Fig.11: I do not understand that statement. The figure shows only the column

average HONO concentration is near (81%) to the ground level. That could be also explained by a

particle source?

Response: Correlation between the column average HONO concentration and HONO measured from the ground level to 10 m in height have been reanalyzed during the clean and the haze episodes, respectively. The production of HONO on aerosols explained the HONO observations in the residual layer during E3. If the production of HONO was indeed dominated by heterogeneous reaction of NO_2 on aerosol and ground surfaces, the column average HONO concentration is expected to be irrelevant or relevant to the mixing ratio of HONO observed from the ground level to 10 m. Figure 10a showed that column average concentration of HONO is irrelevant to the amounts of HONO observed between the ground level and 10 m in height ($R^2 = 0.27$), which suggested that the aerosols dominated the production of HONO aloft by the heterogeneous reaction of NO_2 during the haze episode. However, a high correlation ($R^2 = 0.93$) between the column average HONO concentration and HONO measured from the ground level to 10 m was observed during C2 (Figure 10b), which suggested a surface HONO source at night during the clean episode. The HONO observed throughout the depth of the boundary layer was primarily derived from heterogeneous conversion of NO_2 on the ground followed by vertical transport throughout the column. The statements of Figure 10 (the original Figure 11) have been significantly revised in the manuscript to make our argument more clearly.

8. Figures 7+8: The concentrations in the residual layer typically represent daytime levels (in the

absence of a volume source of HONO). Here HONO levels of 1-2 ppb are observed, which should be

discussed. Such high HONO levels at 250 m altitude are really exceptional!

Response: We sincerely thank the Jörg Kleffmann for his constructive advice. We completely agree with Jörg Kleffmann that high HONO levels in the residual layer (typically represents HONO daytime levels) are well worth studying. However, this study primarily focused on the investigation of the vertical distribution of nocturnal HONO and relative importance of aerosol and ground surfaces in nocturnal HONO production at different pollution levels. The limited vertical measurement and ancillary data restricted the analysis on daytime HONO levels. In the future, we plan to conduct further in-depth research on the daytime HONO. Combined with more comprehensive vertical measurements, the vertical distribution and sources/sinks of daytime HONO will be studied.

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