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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simultaneous HONO measurements in and above a forest canopy: influence of turbulent exchange on mixing ratio differences" *by* M. Sörgel et al.

Anonymous Referee #2

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The manuscript by Soergel and co-authors presents gradient measurements of HONO, HONO/NO_x ratios, and a number of other environmental parameters below and above a forest canopy in Germany. The manuscript provides an extensive introduction into HONO chemistry and a careful and detailed description of the experimental methods used in this study. The authors use various methods, such as the comparison of HONO gradients with micrometeorological vertical transport classifications and the temporal change of HONO and HONO/NO_x to investigate HONO chemistry. The main findings of the study are that vertical mixing through the canopy is one of the main factors controlling below canopy HONO and that relative humidity seems to have an impact on the formation of HONO at the ground.

The data presented in the manuscript is quite interesting and provides information on how heterogeneous chemistry, photolysis, and vertical mixing influence the concentrations of HONO in a forest environment. The authors provide a clear interpretation of their observations. I support publication in ACP after the authors address a number of issues that would clarify their manuscript.

Detailed comments:

- The authors use relative humidity throughout the manuscript as a measure for water in the atmosphere. This makes sense when analyzing heterogeneous chemistry as the amount of water on a surface is related to RH. On the other hand I am puzzled by the use of RH to understand the vertical structure of the atmosphere, as RH depends both on water mixing ratios and temperature. It is thus impossible to separate water vapor mixing ratios profiles from temperature profiles when showing RH alone. Vertical transport of water vapor will primarily depend on the mixing ratio gradients. It thus seems to me that, for the comparisons with HONO gradients, showing water mixing ratios or their gradients in figures 5 and 7 would make more sense.

- Section 3.2.2: It is difficult to directly compare S/V of aerosol and surface to interpret chemistry as the gas transport processes to the respective surfaces may be the limiting factor in the heterogeneous conversion of NO_2 to HONO. Gas transport to and from aerosol is much faster than that to and from the ground. Therefore, one cannot draw a clear conclusion that the low aerosol S/V necessarily means that there is no significant conversion of NO_2 to HONO on the aerosol. A more detailed analysis and discussion is needed to support the authors claim that HONO formation on the aerosol can be neglected.

- Page 21123, Line 9-11: Why would one expect a correlation of HONO concentration with NO_2? Based on HONO chemistry it should be the HONO formation rate,

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i.e. d[HONO]/dt, that correlates with NO_2 concentrations (or any other HONO precursor).

- Section 3.3: This section would benefit from a slightly more detailed explanation of the origin of the different coupling regimes. Many readers may be unfamiliar with this classification and they would have to first read other manuscripts to follow the author's arguments. Also please add a reference to the Serafimovich et al (2010) manuscript in this section, as it will otherwise get lost in the experimental part of the paper.

- The authors repeatedly state that HONO under the canopy is formed at the ground. Can it not also be formed at the bottom of the canopy?

- Page 21128, line 27-30 and Figures 5&7: Fig. 5 shows that the time around 21:00 is dominated by wave motion with a decoupling of the atmosphere above and below the canopy. Fig. 7, on the other hand, shows simultaneous increase of HONO at both altitudes at 21:00. The manuscript states that this event was due to an airmass exchange. How do the authors reconcile the conclusion that at this time mixing was not important (Fig. 5) with the fact that the HONO increase on Sept 23 occurs below and above the canopy simultaneously (Fig. 7)?

- Figures 1 and 2 would benefit from showing the actual HONO mixing ratios measured by the two instruments. In the case of Figure 1 it appears that during times of low visibility the HONO mixing ratios were also low, which would lead to large relative discrepancies between the two instruments at small absolute differences between the HONO measurements. In the case of Figure 2, showing the upper level HONO mixing ratios would allow to put the below canopy data into perspective and support the conclusions of heterogeneous formation on the ground. It would also help to show NO_2 and NO data in this plot as it is used and discussed in the manuscript, but never displayed.

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- Figure 7: While I like the aesthetics of this figure it is extremely difficult to actually see the gradient between the two HONO measurements. Please make this figure clearer.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 21109, 2010.