

General comments

Wong et al. present vertical profiles of HONO, NO₂ and O₃ obtained from long-path DOAS measurements in Houston, TX during three different nights in September 2006. The observed HONO profiles are compared with vertical HONO profiles derived from a 1-D chemistry and transport model after adjusting NO_x emissions and vertical turbulent transport to make the modeled temporal and vertical profiles of NO₂ and O₃ fit the measurement. Finally, the model is used to quantify the contributions of different HONO formation and loss processes to the net HONO production at different heights.

The separation and quantification of different HONO formation and loss processes is an important and interesting topic. Therefore, this combined presentation of observational and modeling data merits publication. However, I do have a few major questions about the comparison of the observational and model results. Also, the manuscript needs some technical revision before publication.

Specific Comments

1. Taking into account that the upper height interval was not reproduced well by the model, and that the night period on 11/12 September was not reproduced well by the model, I would ask the authors to remove their statement that "the observed HONO profiles were reproduced well by the model" from the abstract (p.30130, 1.15-16).
2. While the introduction is nicely written and just about the right level of detail, I don't think that Figure 1 fits with the text. The authors may want to expand on their explanation of Fig. 1, e.g. explain the different arrow colors. It could also be helpful to indicate reactions R1-R3 in Fig. 1, and put labels on the different ground and aerosol surfaces.
3. In Fig. 2, I do not understand how the authors arrive at the averaging dimensions associated with the light path between 70 m and 300 m. How exactly is the upper height interval between 130 m and 300 m derived from the light path arrangements between Moody Tower (70 m) and the three retroreflectors at 20/130/300 m? What is the exact horizontal averaging of the upper interval as indicated by the light red box? It seems to be different from the horizontal averaging between Moody Tower and downtown Houston as noted on p.30134, 1.17.
4. On p.30135, 1.6, the authors mention that vertical gradients disappear in the morning when the boundary layer became well-mixed. It would be very helpful to add a complementary measurement to Fig. 3 that indicates the observed mixing state of the boundary layer. On that same note, do you have complementary measurements of atmospheric stability?
5. P.30136, 1.25: Please indicate sunset and sunrise in Figs. 4-6.
6. P.30137, 1.7 and 1.20: Can you quantify the positive correlation between HONO and NO₂?
7. The used model is subdivided into 27 boxes, 9 of which are below the lowest observational height of the LP-DOAS of 20 m. I was left wondering how you initialized these nine lowest model boxes. Was there any additional observational data available closer to the ground? While you find the largest production and loss processes close to the ground (e.g. p. 30148, 1.13-14), this is also where the model is not constrained by LP-DOAS observations. This raises the question how confident you are in the model results close to the surface, e.g. as presented on p.30147, 1.12-20. A direct comparison with in-situ HONO measurements at the ground would be a very valuable addition to the manuscript.

8. P.30139, 1.18-21: How is the aerosol surface area density initialized and parameterized in the model? Did you assume a uniform aerosol profile over the NBL (p.30148, 1.18)? If so, all conclusions about HONO formation and uptake on aerosol surfaces and comparisons with ground surfaces should be made with great care. Furthermore, it should be clearly stated that the description of aerosol surface profiles is oversimplified.

With regard to the same topic: Is the gas phase transport of HONO to aerosol surfaces and to ground surfaces treated differently in the model? In fact, the transport to the surfaces may be the limiting factor in the heterogeneous conversion of NO_2 to HONO.

9. P.30140, 1.12: Please give a reference for the typical VOC concentrations that were used in the model runs!

10. On p. 30141, the authors compare the vertical mixing and the horizontal transport timescales. For the reader it would be useful to find typical values of these timescales in seconds or minutes presented in this section.

11. The model calculations showed an increase of the HONO/ NO_2 ratio with height which was not reflected in the observations. The authors discuss their findings and conclude from sensitivity studies that the increase was due to excess HONO rather than the lack of NO_2 , most likely due to an underestimation of HONO loss on aerosol surfaces (p.30144). However, there is no discussion of vertical mixing as a possible explanation. Could a different parameterization of vertical mixing also explain the model results?

12. On p.30145, 1.23-25, a deviation of the observed HONO/ NO_2 ratio from the refined model results is explained by HNO_3 conversion on fresh soot aerosol during rush hour. At what time did you observe the largest deviation?

13. Regarding vertical mixing: Can you explain the steep increase of eddy diffusivity starting around 03:30 CST as shown in Fig. 7? Did you compare the adjusted eddy diffusivities used in the model runs to any in-situ turbulence measurements in order to evaluate if vertical mixing is parameterized in a reasonable fashion in the model? This is a very important issue because vertical transport was found to be the dominant source of nocturnal HONO above 20 m in all model runs (p.30149, 1.5-6).

14. I like the discussion of two regimes of net HONO formation, a "transport sensitive" and a " NO_2 sensitive" regime on p. 30150. Please also indicate both regimes in Fig. 10.

15. The authors repeatedly state that their model did not accurately reproduce the upper height interval because it was located in the residual layer. Still, they continue to evaluate and interpret HONO formation and loss rate profiles in section 6 up to 300 m, e.g. in Figs. 8 and 9, Table 3, as if the model had been shown to reproduce the profiles correctly. The authors should clearly explain how reliable they feel their conclusions are.

Technical Corrections

p.30130, 1.22: Add "an" between "showed increase".

p.30131, 1.3: Add " NO_x " between "stronger emission".

p.30131, 1.6: Change "Nitrous acid, HONO is one" to "Nitrous acid (HONO) is one".

p.30133, 1.1: Replace "occurs" with "occur".

p.30133, 1.3: "such as buildings, plants etc, or on particles." looks odd!

p.30133, 1.16: Change "gradients measurement" to "gradient measurements".

p.30133, 1.18: Change "aerosol surface" to "aerosol surfaces".

p.30135, 1.10: Should mention that Fig. 3 also shows temperature.

p.30135, 1.16: Change "condition" to "conditions".

p.30136, 1.2: Add "the" between "at lower".

p.30136, 1.3: Change "Difference" to "Differences".

p.30136, 1.10: Change "Difference" to "Differences".

p.30136, 1.18: Change "Difference" to "Differences".

p.30137, 1.12: Add comma between "September respectively".

p.30137, 1.24: Remove "and"

p.30138, 1.2: Change "6% respectively on" to "6%, respectively, on".

p.30138, 1.3-4: "HONO/NO₂ increased to 4% before midnight then was 3-4% throughout the night." looks odd.

p.30138, 1.18-19: Change "model are based on (Kurtenbach et al., 2002, 2001)." to "model are based on Kurtenbach et al. (2001, 2002)."

p.30139, 1.10: Add "an" between "with emission".

p.30139, 1.12: "currently included in the model include" should be changed.

p.30139, 1.16: Change "R3 and a NO₂ reactive" to "R3 and an NO₂ reactive".

p.30139, 1.22-23: Change "reactive uptake coefficient 10⁻⁴" to "reactive uptake coefficient of 10⁻⁴".

p.30140, 1.2 and 1.21: L should not be called "Monin-Obukhov length", but "Obukhov length".

p.30140, 1.9-12: "Because vertical gradients of other trace gases..." seems incomplete.

p.30141, 1.24-25: Change "and trace gases distribution" to "and the trace gas distribution".

p.30142, 1.6: Change "is the" to "are".

p.30143, 1.14: Change "altitudes" to "altitude".

p.30143, 1.26: Remove comma after "September".

p.30147, 1.9: Change "mixing ratios profiles" to "mixing ratio profiles".

p.30147, 1.24: Change "increases" to "increased".

p.30148, 1.1: Change section heading to "HONO formation and loss rate profiles".

p.30149, 1.2: Remove "HONO".

p.30150, 1.1: Change section heading to "Dependence of the net HONO formation at the ground on vertical mixing and NO_x emission".

p.30150, 1.7: Add comma after "Consequently".

p.30150, 1.17: Add "the" between "that impact".

p.30151, 1.1: Add "the" between "of emission".

p.30151, 1.23: Change "substantial" to "substantially".

p.30152, 1.6: Add "the" between "during morning".

p.30152, 1.11: Change "or" to "and/or".

p.30146, 1.2-3: What does "both modeling periods" refer to?

p.30159, Table 2: I assume you show mixing ratios in ppb. Please give the units of the presented values.

p.30163: Fig. 3 also shows temperature measurements. Please modify the figure caption accordingly.