

Interactive comment on “Daytime HONO Vertical Gradients during SHARP 2009 in Houston, TX” by K. W. Wong et al.

Anonymous Referee #1

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The role of HONO (nitrous acid) in the atmosphere continues to be of considerable interest to the atmospheric chemistry community due to the role of HONO as radical precursor. Recent evidence for significant daytime levels of HONO above the expected photostationary state have promoted HONO to the main OH sources during the day. Only few studies have addressed HONO vertical gradients during daytime. Therefore, this study is a timely and adequate investigation attempting to allocate the unknown source of daytime HONO better to its potential location, i.e., ground surface vs. airborne, gas phase or particulate. This nice and very detailed manuscript is an account of carefully analyzed DOAS measurements covering three different altitude ranges above ground. The authors use a series of lines of arguments to come to the conclusion that the unknown daytime HONO source is most likely located on the ground. While I have no reservations with these lines of arguments directly, I feel that since a main aim of the

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analysis was to find evidence for or against the HONO source being associated with aerosol surfaces, a better way of assessing the vertical gradient of the aerosol surface could have received higher priority in this campaign. However, I do not consider that a major point and I recommend publication after the relatively minor points detailed below are addressed.

1) Overall, the text is very well written and structured. I especially appreciate the long but well worked out introduction. The only general technical comment I have is that the conclusion section should be shortened a bit to avoid any reiteration of discussion there.

2) General scientific comment: The authors were obviously surprised about the significant gradient of NO₂ at times when efficient mixing should even it out. This is referred to at several places in the manuscript and would deserve some more discussion and comparison to other studies. What are the expected sinks and how do these compare to the vertical mixing speeds? Is there an unknown sink for NO₂? Since HONO to NO₂ ratios remained very similar along the vertical profile, this of course could have some ramification for the unknown HONO source.

3) page 24368, line 16: ...conversion on humid surfaces: I suggest to make this more explicit to mention both NO₂ heterogeneous hydrolysis (disproportionation) and reaction with organics. The latter reactions (in the dark) are significantly faster than the disproportionation reaction on many different substrates.

4) page 24369: discussion of role of NO₂*. For the sake of completeness, the authors should also cite the response of Li et al. to the comment by Carr et al. (2009)

5) page 24372, line 8: in the Arctic

6) page 24382, line 19: section 4: should this be 4.2; note that there is already a reference in the preceding line.

7) page 24385, top lines: in relation to the nitrophenol photolysis in the gas phase,

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nitrophenols formed in the particulate phase may also contribute to formation of HONO (Sosedova et al., 2011).

8) page 24385, 2nd paragraph: how was aerosol nitrate measured? Could photolysis of nitrate be a significant sink for aerosol nitrate?

9) page 24385 3rd paragraph: is there any impact of the complex urban ground surface structure on the difference between actinic flux and irradiance?

10) page 24388 or in the vicinity: the authors could include a comparison to the recent study by Su et al. (2011) and also discussion thereof. This should probably also added to the introduction section.

References Li, S., Matthews, J., and Sinha, A.: Response to Comment on "Atmospheric Hydroxyl Radical Production from Electronically Excited NO₂ and H₂O", *Science*, 324, 336, 10.1126/science.1166877, 2009.

Sosedova, Y., Rouviere, A., Bartels-Rausch, T., and Ammann, M.: UVA/Vis-induced nitrous acid formation on polyphenolic films exposed to gaseous NO₂, *Photochemical & Photobiological Sciences*, <http://dx.doi.org/10.1039/C1PP05113J>, 2011.

Su, H., Cheng, Y., Oswald, R., Behrendt, T., Trebs, I., Meixner, F. X., Andreae, M. O., Cheng, P., Zhang, Y., and Pöschl, U.: Soil Nitrite as a Source of Atmospheric HONO and OH Radicals, 333, 1616-1618, 10.1126/science.1207687, 2011.

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