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Discussions

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Interactive Comment

Interactive comment on "Impact of updated traffic emissions on HONO mixing ratios simulated for urban site in Houston, Texas" *by* B. H. Czader et al.

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— We would like to thank the Referee for time and effort put into reviewing this manuscript. Please see below our responses to your comments.

One of the major issues here is the lack of continuous or sufficient HONO measurements. I can hardly count 6 diurnal cycles of HONO measurements in Fig. 6, which is properly not adequate to discuss model-measurements comparison. From Fig. 6, HONO simulations seem to be improved only during the early morning and most apparently during the Sep. 12th. Figs 4 and 5 are shown only for Sep 12th, what about the other days?

— HONO is usually not measured routinely, which is a shortcoming of this as well as other studies (e.g., Wang et al. (2011)). Limited availability of measured data is already mentioned in the manuscript (see page 21324 lines 16-19). We hope that the new emission ratio would be tested in other areas along with different HONO measurements that would provide additional validation of HONO emissions. For our study we will add more detailed analysis of the dataset that is available to us as well as more analysis of the modeling results itself, especially on the potential impact of these higher emissions on modeled mixing ratios (see .

— Since only HONO emissions from mobile sources were increased we expect to see the largest differences in mixing ratios during early morning times when the traffic emissions are the highest, the mixing layer height low allowing for accumulation of HONO, and photochemistry not very active. We will incorporate this statement into the manuscript.

Also from Fig. 6 (since it is the only figure that show several days of HONO diurnal cycles), it seems that HONO was much better simulated with the (N) scenario on Sept. 11th, 19th and 26th, which are significantly overestimated by the new (NH) scenarios. HONO simulations seem to be improved only on the Sep. 12th, 18th.

— Variations of simulated HONO mixing ratios from day to day are influenced not only by emissions but also by other parameters, for example, the model capability to predict grow of the mixing layer as well as clouds that influence photolysis rates. To more clearly present differences between the simulations cased we prepared the average diurnal profile of measured HONO and compared it with simulated N and NH cases (see Fig. 1). It can be seen that NH case improves HONO morning peaks. As mentioned above, the increase in the morning can be explained by high traffic emission during morning times, low mixing layer height and accumulation of HONO since photochemistry is not very active.

— HONO simulations with the new ratio are improved on Sep. 12, 18 as well as on Sep. 23, 25, and 30.

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The authors should also plot the Measured vs Simulated HONO for both scenarios (N and NH) and for each complete diurnal cycle and for the mean simulated period (thought statistical mean is shown in table 3), so that we can get a clearer picture if the new ER (NH scenario) would consistently improve HONO or only under certain conditions. Why it is only improved on the 12th?

— Diurnal profiles of the measured and simulated HONO from both scenarios (N and NH) are already presented in Figure 6. As mentioned above, based on the comparison of measured NOx from very representative dataset at many stations around Houston taken during the whole month of September, we believe that N case better reflects observed NOx and since HONO is derived directly from NOx, reducing emissions of NOx resulted in HONO reduction.

— The additional figure with an average HONO profiles (see Fig. 1) will be added to the manuscript along with the following description:

"Figure 1 shows the average diurnal profiles of measured and simulated HONO mixing ratios. Since only HONO emissions from mobile sources were increased the largest differences in mixing ratios occur during early morning times when the traffic emissions are the highest, the mixing layer height low allowing for accumulation of HONO, and photochemistry not very active. The model underprediction during daytime can be explained by the fact the default model version that we used in this study does not account for the photochemical HONO sources. Also, too low modeled average profile during daytime is caused by underpredictions of HONO on Sep. 23-25 which can be attributed to stronger modeled winds in comparison to weak observed winds causing HONO to be removed from the observational site. It is worth to note that all available measured data for HONO for the September 2013 are from weekdays and the higher HONO/NOx ratio measured in Houston was calculated based on measurements taken during weekdays."

The statistical mean in table 3 is misleading because the overestimated and underesti-

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mated HONO cancel each other resulting in slightly improved mean simulated HONO. So here also, the authors should show the results for each diurnal cycle (not the mean).

— We agree that the mean values might be misleading but in addition to the mean value we also calculated and presented in table 3 the absolute mean error. The underestimated and overestimated values do not cancel each other in calculation of the absolute mean error because the absolute values are taken for the calculation (please refer to equation (1)).

On the days 23-25th, HONO measurements are still significantly underestimated, especially during the afternoon time. This underestimation should also be discussed in more details. In fact, most of the HONO unknown sources are reported during afternoon hours (e.g., Kleffmann et al., 2005; Elshorbany et al., 2012). During the early morning, the so called [HONO]pss (zero net OH source), which account for the known gas phase HONO formation from OH+NO and loss through photolysis and reaction with OH, accounts for most of the early morning peak.

— We agree that part of underprediction of HONO on those days may be due to photochemical HONO formation that is not accounted in the model. Also, on September 21-25 a cold front was passing through the Houston area, with high pressure system. On Sep. 23 – 25 the model shows stronger easterly winds than the observation that contribute to faster transport and removal of HONO from the observation site.

Why these high emission ratios, Could the authors try to shed some light on the type of fleet in Huston Metropolitan Area, compared to other cities in the US or to the fleet in Europe, does the fleet type and quality changed over time (Benzene, diesel, natural gas, hyprid cars, ::: .etc), why are ER are different that reported before?

— We will add the following discussion about that:

"The HONO/NOx ratio reported by Kurtenbach et al. (2001) is based on measurements performed between 6 am and 2 pm, for both weekdays and weekends where 22 200

± 400 vehicles were passing on weekdays and 13 300 ± 1 400 cars passing on weekends. The vehicle fleet was composed of 6.0% heavy-duty trucks, 6.0% commercial vans, 12% diesel and 75% gasoline powered passenger cars, and 1.0% motorcycles. The measurements made by Rappenglueck et al. (2013) reflect high traffic, early morning conditions (4-8 am) on weekdays. The measurements were performed at highway junction in Houston with very high traffic load (about 400 000 vehicles passing daily), which is much larger than that in the tunnel study. The vehicle fleet was represented by 93-95% of gasoline fueled vehicles and 5-7% by diesels during the morning hours. Another difference between these two studies is in vehicle speed, with a typical speed of 50-90 km/h in the tunnel studies and much lower speed during the morning peak traffic hours in Houston. "

— Also, the following will be added at the end of paragraph in line 27 on page 21320:

"Since the newly reported ratio reflects high traffic conditions during the morning rush hours on weekdays our model sensitivity study provides estimate of the upper bound of the impact of HONO emissions on pollutant levels in urban areas."

At the end, more scientific discussion of the results is still required. For example, why OH is only enhanced by ~5% though HONO is enhanced by 35% (Table 3) on Sep. 12th. What is the contribution of HONOpss to the total simulated HONO?. Here also Fig. 7 should include all other simulated cycles, i.e. not only one single event.

— We will add the following discussion based on all simulated data:

"Based on the 1 month of simulated surface concentrations the average increase in the morning OH (between 6 – 8 a.m. LT) is 14% at the location of the Moody Tower and 3% when averaged over the urban area. The ozone increase is below 1% for both the Moody Tower and the urban area. The average increase in OH during daytime (6 a.m. – 8 p.m. LT) is 7% for the Moody Tower and 1% for the urban area. The increase in ozone is again below 1%. To obtain more insights on the fate of HONO we performed additional model simulations and analysis for the Moody Tower site for Sep.

10-13, 2013. At the surface at the location of the Moody Tower the average contribution of vertical transport to the loss of HONO is 77%, horizontal transport contributes 8%, chemical removal 11 % and dry deposition 5%. At the second model layer, which corresponds to the altitude of measurements, transport (horizontal and vertical) continue to be a dominant loss process contributing on average 77% to the total HONO loss while chemical loss contributes only 23% to the total loss. The chemical loss of HONO is dominant only during couple of morning hours. Figure 2 shows hourly values of HONO mixing ratios for Sep. 10-13, 2013 along with process contributing to changes in the mixing ratios at the grid cell corresponding to measurements taken at the Moody Tower (simulated data extracted from the second model layer). This explains the fact that even though HONO mixing ratios significantly increased upon additional emissions, HONO was removed mainly by transport with only small portions taking part in chemical reactions converting it to OH and furthermore to O3. Also, the main impact of chemistry is during early morning hours following the peak in HONO."

— We believe that the above presented analysis of the 1 month dataset provide sufficient information on OH increases and do not see a need of adding more graphics in Figure 7.

Technical corrections Page 21317, line 9: HONO photolysis during the early morning was first reported by Perner and Platt (1979) and Harris et al. (1982). Add these references before Czader et al., (2012) and write (e.g.,) at the statement's beginning.

— We will certainly add the above mentioned references and modify the manuscript according to your suggestion.

Page 21325, line 18: 12 September. Page 21325, line 22: 12 September.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 21315, 2014.

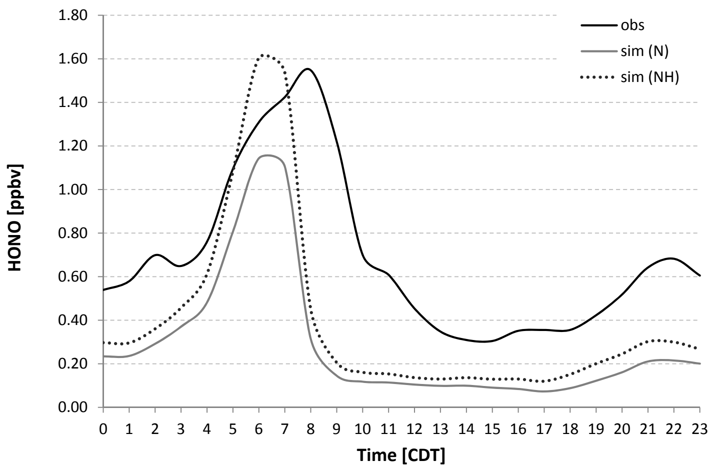

Fig. 1. Average diurnal variation of HONO at the Moody Tower measurement site.

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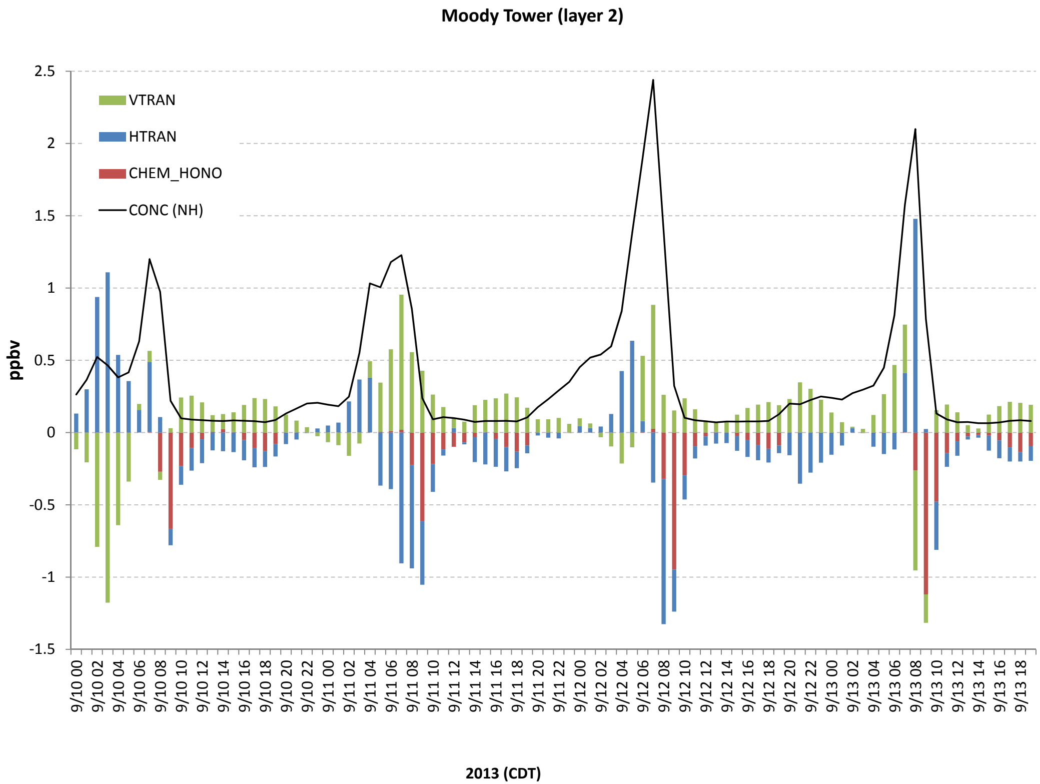

Fig. 2. HONO mixing ratios and processes contributing to changes in HONO mixing ratio at the Moody Tower site where the measurements were taken.

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