In response to the comments posted by Dr. Kleffmann, please find below our answers.

General remarks:

1) The aim of this study is to approximate the HONO mixing ratios to investigate the global impact of HONO *photolysis during daytime* on atmospheric chemistry as stated in the title. Thus, daytime HONO mixing ratios are of high importance rather than nighttime HONO mixing ratios. This study is not aiming at simulating each peak in the HONO diurnal cycle, especially during the night, which is not feasible and of little importance for atmospheric models.

Since the derived HONO/NOx ratio of 0.02 provides good representation of daytime HONO mixing ratios (as also mentioned in the comment), the main purpose of the study is achieved. Furthermore, the nighttime average HONO/NOx ratio for all the investigated campaigns is also 0.02 (see Fig. 4) and the correlation between measured and simulated HONO mixing ratios was quite high during both daytime as well as for 24 h average conditions (see Fig. 5 and Fig. 10). Such good agreement is quite sufficient as a basis for global models.

The suggested parameterization based on the linear increase of HONO/NO2 ratio does not have any merit over the applied method for the following reasons:

1. The suggested heterogeneous formation rate constant is also an average nighttime value, therefore may not reproduce the peak in the nighttime part of the diurnal cycle. In addition, this heterogeneous rate constant was also reported to be a function of wind speed (e.g Elshorbany et al., 2009a), therefore this dependency should also be considered, not as simple as suggested.
2. Most important, the HONO/NO2 ratio (which only accounts for heterogeneous HONO formation) obviously does not account for nighttime direct emissions of HONO from combustion sources (see also e.g., Alicke et al., 2002 and Vogel et al., 2003) while HONO/NOx implicitly does.
3. As discussed in the paper (sec. 3.2.1 and Fig. 4 and Fig. 5), for measurement campaigns with nighttime HONO/NOx ratios >0.02, a slightly better representation of nighttime HONO mixing ratio can be calculated by considering the HONO/NOx dependency on NOx or wind speed (ws), while for other campaigns, nighttime HONO mixing ratios were reasonably simulated based on the HONO/NOx ratio of 0.02. However, this additional parameterization is not feasible as it is difficult to determine which location will be characterized by a different HONO/NOx ratio (> or < 0.02).

This additional parameterization will certainly require much more computational time and does not have any impact on daytime HONO chemistry.

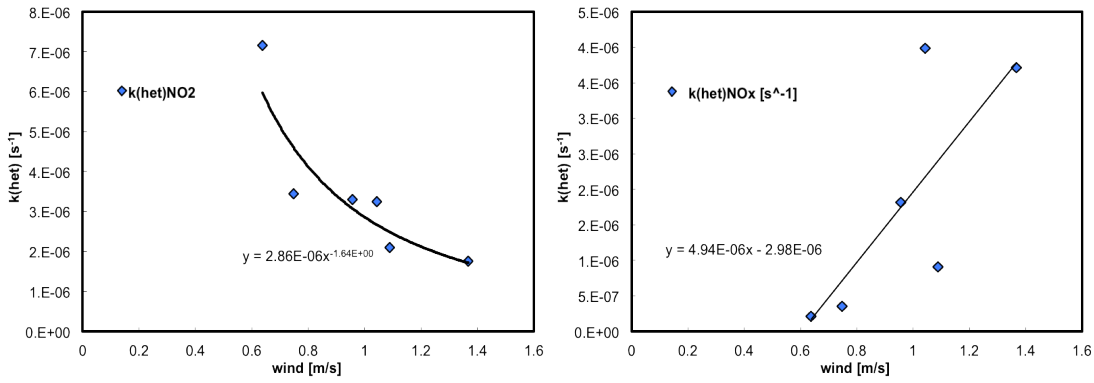
Therefore, the applied methodology in the paper provides the best approximation of HONO mixing ratios during the entire day for use in atmospheric models.

2a) This is not correct. If the HONO/NOx ratios were independent of NOx levels (as during the daytime, see Fig. 6, lower panel), then the HONO heterogeneous formation rate would have been independent of NOx, implying that NO2 does not play an important role in HONO heterogeneous formation, which is not the case.

The suggested explanation is also not correct. As mentioned in sec. 3.1, the increased value of the HONO/NOx ratio during nighttime is caused by the increased NO2/NOx ratio owing to the changes in the meteorological conditions discussed in sec. 3.1, leading to increased heterogeneous formation of HONO. This is confirmed by the HONO/NOx dependency on wind speed (see next).

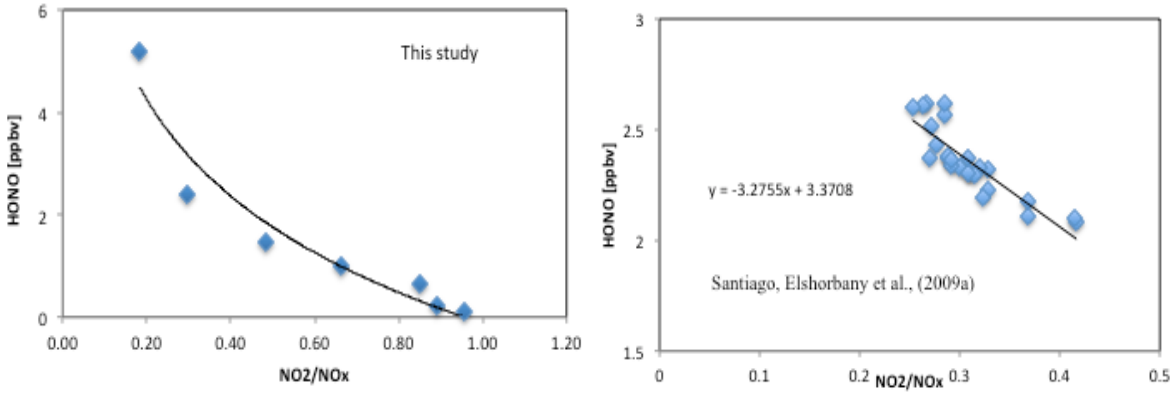
2b) Here the comparison is also not correct and confuses different aspects. In this study, HONO/**NOx** was correlated to the wind speed while in Elshorbany et al., (2009b), the heterogeneous rate constant (HONO/**NO2**/time) was correlated to the wind speed. This is a big difference.

As shown in the figure below, also for the Santiago summer campaign 2005 (Elshorbany et al., 2009a), the correlation between the heterogeneous rate constant calculated based on **NOx** (right panel) and wind speed yields also a positive correlation.



As shown in the figure below (left panel) depicted from Table 1 in the paper, the HONO mixing ratio correlates inversely with the NO2/NOx ratio for all investigated campaigns. This

is also consistent with the anti-correlation between HONO and the NO2/NOx ratio for the summer Santiago campaign (Elshorbany et al., 2009a) shown in the right panel of the figure below.



Therefore, all HONO/NOx dependencies shown in the paper are correct and consistent with all previous field measurements.

For more clarification, the following statement will be inserted on line 19 page 12895 in the revised manuscript "Similarly, Elshorbany et al. (2009a) have shown that the HONO heterogeneous rate constant in the dark (calculated based on the HONO/NO2 ratio) correlates inversely with wind speed. It is worth mentioning that positive correlation between HONO/NOx and wind speed is related to the anti-correlation typically observed between HONO and the NO2/NOx ratio for all investigated campaigns (not shown)".

3) Again, this comment is a wrong interpretation of our manuscript for the following reasons:

1. This is the first study where several HONO data sets are compared and contrasted in such a detailed manner. To the best of our knowledge, there are no previous studies that have investigated the correlation between unidentified HONO sources with meteorological parameters (actinic flux and temperature data) from different data sets under different atmospheric conditions.
2. The correlations shown in Fig. 9 are for different measurement campaigns under different atmospheric conditions (as also mentioned in the comment) and not for the same measurement campaign as reported for Santiago 2005 campaign (Elshorbany et al. 2009a); therefore, the comment misinterprets our manuscript.
3. There is no contradiction between the data reported in Elshorbany et al. (2009a, 2012) and the findings in the current study as the positive correlation reported in Elshorbany

et al. (2009a, 2012) may be only due to the similar shape of the diurnal cycles of both production rate of OH and j(NO2), which is why correlations vs j(O1D) had an intercept due to the lower frequency span (smaller diurnal cycle), and does not necessary reflect any physical dependency.

4. Yet, there are no known sources (including those that are j(NO2) or temperature dependent) that can account for most of the observed HONO daytime concentrations (see sec. 1 in the paper). Therefore, there is no contradiction with our results, which are based on experimental results from different data sets.
5. It can be expected that the increased pollution level (of the order Santiago_W > Santiago_S > HOxCom) will be associated with stronger unidentified HONO sources, while the inverse (higher HONO unidentified sources under clean conditions) is not reasonable.
6. As mentioned in sec. 3.2.2, the correlations in Fig. 9 were limited to the measurement campaigns with a HONO/NOx ratio of >0.02 (see Fig. 8 and the corresponding text), thus not selected (we have not chosen campaigns, as indicated in the comment). The data sets that are mentioned in the comment, indicated to contradict with out results, are actually included in the linear regression analysis in Fig. 9. Furthermore, at variance with the comment, no data were presented that contradict with our correlations (e.g., values of campaign average unidentified HONO sources and j(NO2) that significantly deviated from our correlations). This linear regression analysis is based on data from different measurement campaigns, yielding high correlation coefficients and would merit further evaluation (as already mentioned in the text, page 12901, line 16).

The suggested correlation is also not helpful because dividing the unidentified HONO sources by NOx would only make sense if unidentified HONO sources directly correlate with NOx, which is not the case as shown in Fig. 6, where the daytime HONO/NOx ratio does not correlate with NOx, in agreement with previous studies (e.g., Zhang et al., 2012).

In addition, the calculated unidentified HONO sources for other campaigns using the obtained linear regression equations were found to be consistent with the reported experimental values. Nevertheless, as mentioned in line 15, page 12901, since these correlations were based on only limited number of campaigns, further measurements and evaluations are required.

These results are also in line with the results of Veitel and Febo, who showed that the highest HONO levels were measured in winter (higher pollution levels and lower actinic flux)

coinciding with high radon activity (implying a ground source). Nevertheless, for more clarification, the statement starting in line 27, page 12900 will be rephrased in the revised manuscript.

Another strong piece of evidence that supports our findings in Fig. 9 is that Villena et al., (JGR, 2011, 166, D00R07) reported HONO measurements under very clean conditions in Barrow, Alaska. They reported a maximum HONO source strength of about 90 pptv h-1 at an average maximum j(NO2) value of about 0.0078 s-1. Using the j(NO2) dependency in Fig. 9, the calculated unidentified HONO source would be about 110 pptv h-1, in excellent agreement with the measured value. This result shows that the correlations shown in Fig. 9 are robust and valid even under very different atmospheric conditions.

Minor concerns:

1) Again, this is not correct and in contradiction with all recent studies. There is no HONO source(s) that has been reported to account for most of the measured daytime HONO levels. As mentioned (sec.1, page 12887, from line 10 on), recent modeling studies (e.g., Li et al., 2011; Goncalves et al., 2012) showed that modeled HONO levels were consistently lower than observation, even when the most effective recent suggested formation mechanisms were considered. Su et al. (Nature, 2011) showed that known daytime HONO daytime sources do not account for the measured HONO levels, and suggested additional temperature-related source. In Elshorbany et al. (2010b), only an overview of the different daytime HONO sources was presented and without any comparison with models and without any quantification of their possible contribution to the measured HONO. Only some sources were suggested to be important under rural conditions and others under urban conditions. The surface based sources reported by Stemmler et al. (2006; 2007) and Zhou et al. (2003; 2011) are sources that can be important under certain conditions and may can be applied in box or regional models under certain assumptions to explain measurements but still can not be implemented in atmospheric models in absence of HONO measurements (see discussion in page 1288, from line13). Since main HONO sources are yet not known, the question of whether these sources can explain a gradient or not is till open.

2) Unknown daytime HONO sources were reported in a number of studies to correlate with NO2. Sörgel et al. (2011a) showed also that unknown HONO sources normalized with NO2 correlate with j(NO2). However, as shown in Fig. 2 and mentioned on page 12893, lines 1-10, HONO/NO2 is not a good proxy for daytime HONO mixing ratios because HONO/NO2

shows a strong opposite gradient to HONO during daytime, in contrast to HONO/NOx, of which the values are almost unchanged at different altitudes. Furthermore, in the above reference we do not see any correlation between the HONO source strength and NO2.

It seems that in this comment Dr. Kleffmann agrees that Fig. 9 is clear and shows that daytime HONO source depends on pollution levels.
Our observations shown in Fig. 6 and mentioned on page 12896, line 15, suggest that NOx does not directly correlate, or is not an important precursor of HONO during daytime, in agreement with previous studies. Possibly NOx can be considered as an indication of pollution levels but does not itself explain HONO unknown sources. Since HONO unknown sources and formation mechanisms are not yet known, we cannot speculate about any dependency that may or may not exist. These results are simply based on observational data and we could not find any NOx dependency.

3) First, this statement is not correct and also contradict with the statement in your review on HONO daytime sources (Kleffmann, 2007, page 1137, 3rd paragraph), stating that "The formation mechanisms of HONO in the atmosphere are not yet well understood. Direct emission can only partly explain atmospheric HONO emissions and are significantly lower than the typical values of 2-10% measured during night in the atmosphere"!!!!.
Second, as mentioned on page 12897, line 2, the HONO/NOx ratios of 0.3-0.8 estimated by Kurtenbach et al. (2001) for directly emitted HONO were estimated at NO/NOx ratio of >90% (also in Elshorbany et al., 2009a). The fraction of emitted HONO modeled by Vogel et al. (2003) in one spot location was calculated during the traffic rush hour time and measured HONO was 30-50 % underestimated by the model, which means that the calculated fraction would be much lower when compared to measured HONO. The value of 0.8% is determined in Santiago de Chile in one location is not "typical" and contradicts with the range of 0.3 – 0.8% determined by Kurtenbach et al. (2001). For example, Alicke et al. (2002) determined a direct HONO/NOx ratio of only 0.65% for the urban Milan area.
Third, as mentioned in text, owing to the variable low NO/NOx ratio (0.2 – 0.6) for the investigated measurement campaigns worldwide, under different atmospheric conditions, one single typical value for the HONO/NOx ratio cannot be derived. In addition, owing to this low NO/NOx range, direct HONO emission is not expected to contribute significantly to the unidentified HONO sources. For the same reason, Su et al. (2008) ignored direct HONO emission from their calculations.

Fourth, as mentioned on page 12897, from line 23 onward, the highest contribution of unidentified sources is during the afternoon (see also Fig. 7), when NO/NOx ratios and HONO direct emissions are low. It is also stated on page 12898, lines 23-27, that for measurement campaigns with a HONO/NOx ratio ≤0.02, HONO can be explained by HONOpss in addition to small contributions from other identified sources (could also be emission sources).
Finally and most importantly, HONO unidentified sources included in Fig. 9 were calculated only for the afternoon period and only for a HONO/NOx ratio of about 0.04 (> 0.02) (Page 12898, from line 15); therefore, even if direct emission would contribute 0.8%, this would not have any influence on our findings in Fig. 9.
Again, the average HONO/NOx ratio of 0.02 was determined from experimental data from 15 field measurement campaigns (see Fig. 3) and is an empirical ratio to calculate HONO levels. It is not important which sources are included in this ratio. It is merely an empirical formula derived from field measurements to approximate HONO mixing ratios, and it does reasonably simulate HONO for all the investigated field campaigns (see Fig. 5).

4) We also disagree with this comment for the following reasons:

1. The HONO/NOx ratios of 0.02 implemented in the model was derived from several experimental data sets (not based on the model output) and was shown to reasonably simulate HONO levels in all these data sets (see also Fig. 5).
2. In Fig. 12, S3 clearly overestimates HONO levels during the day, except for the mid-noon hours while the S2 simulation shows the best agreement with measurements throughout the day (especially during the early morning and afternoon and until midnight), not S3 !!!.
3. Compared to the reference case, S2 shows a much reasonable HONO mixing ratios relative to measurements, even during the night.
4. Nighttime HONO mixing ratios do not have any impact on atmospheric photochemistry (i.e., the aim of the study) and the additional efforts to better simulate it (if possible at all) are justified within the current objectives.
5. As mentioned on line 13, page 12903, despite the good agreement, the diurnal cycles in Fig. 12 represent larger areas than the measurements owing to the coarse resolution of the model. Therefore, the accurate comparison here is not coincidental.