We thank Anonymous Referee #1 for his/her comments and interest and for recommending our paper for publication. Below are our replies, given the same numbers as the comments.

General comment:

The current study employs a HONO/NO_x parameterization based on data from 15 field measurement campaigns around the globe (see Table 1 and Fig. 3) performed during the last decade under very different seasonal and atmospheric conditions ranging from VOC-sensitive conditions, mainly experienced in urban polluted areas (e.g., Santiago, New York, Milan) to NO_x-limited conditions, mainly experienced in rural (e.g., HOxComp, PRIDE-PRD2004, PRIDE-PRD2006) and remote (e.g., DOMINO) areas as also mentioned in sec. 2.2, pages 12891-12892. Thus, this parameterization is not limited only to urban high NO_x conditions but represents an overview of HONO measurements over a wide range of atmospheric conditions. Even though HONO chemistry is found to be most important under high NO_x polluted conditions; we aim at a global perspective considering the impact of HONO under different atmospheric conditions.

1. As shown in Fig. 11, HONO mixing ratios were increased by about one order of magnitude all over the globe. The impact of this increase on global atmospheric chemistry depends on the photochemical regime in each region and can be positive, neutral or negative. The impact of HONO enhancement is most apparent in high NO_x polluted regions, conditions under which NO levels are sufficient to recycle RO_2/HO_2 back to OH, thus enhancing OH, HO₂, secondary oxidation products including PAN, H_2O_2 , HNO₃ and O₃ photochemical formation, while the impact was neutral or negative under low NOx conditions in rural and remote areas (see Figs 12 – 17).

The impact of HONO enhancement on global budgets of O_3 and OH is also discussed (see page 12905-12906) in terms of the relative global enhancement of O_3 and OH as a function of changes in the seasonal (summer to winter) and photochemical conditions (high and low NO_x conditions). These impacts are investigated both globally (see Figures 14 and 15) and with a focus on polluted regions (see figs. 17 and 18). Further detailed analysis of the ozone photochemical budgets is not within the scope of this study. Furthermore, we do not think that the number of the figures is important; there are already 18 global maps in 7 figures (see Figs 11-17). Thus, the impact of HONO on global atmospheric chemistry is comprehensively addressed considering different seasonal and photochemical conditions (high and low NOx). Therefore, we prefer to keep the title as it is, as it reflects the subject and content of the paper.

2. HONO measurements have been significantly improved over the last decade. For the investigated measurement campaigns in Table 1, HONO was measured by DOAS, LOPAP and aqueous-phase scrubbing and HPLC/IC analysis. The DOAS method is absolute, which detects HONO by its specific UV absorption, thus free of sampling artefacts and chemical interferences (Platt and Perner, 1980). HONO measurements by the LOPAP instrument were intercompared to that of DOAS and were in excellent agreement (Kleffmann et al., 2006), which is due to the active correction for interferences via the two-channel system and absence of sampling lines. Aqueousphase scrubbing and HPLC/IC analysis have been deployed (also in a modified form) in several field measurement campaigns (e.g., Zhou et al., 2002; Ren et al., 2006; Su et al., 2008; Ren et al., 2010; Zhang et al., 2012). Under very clean conditions (i.e., low HONO levels), measured HONO levels using wet-chemical techniques (other than LOPAP) have been reported to be biased by uncorrected interferences leading to artificially very high HONO/NOx ratios (Kleffmann et al., 2006; Kleffmann and Wiesen, 2008; Su et al., 2008; Lu et al., 2010; Zhang et al., 2012). Recent studies acknowledged this problem and corrected for these interferences (Su et al., 2008) or modified the technique to minimize them (Ren et al., 2010; Zhang et al., 2012). Under high NO_x polluted conditions this problem is less pronounced (Kleffmann et al., 2006; Kleffmann and Wiesen, 2008). In the current study, only HONO measurements by LOPAP were considered for low NO_x conditions while for high NO_x conditions, LOPAP, DOAS and aqueous phase scrubbing and HLPC/IC analyses techniques are used. It should be also mentioned that the measurement accuracy depends also on the measurement protocols (blank (zero air) and calibration frequency and which material to be used, ...etc) individually applied by each measurement group. From the good correlations between HONO, NO_x and other parameters for the investigated measurements campaign, we believe that measurement accuracy was adequate. In addition, owing to the large number of measurement campaigns investigated and the good correlations between HONO/NO_x

for all of these campaigns, the obtained correlation should be representative and is suitable to parameterize the model and to validate its results.

In the revised manuscript, statements on line 24, page 12894 has been edited to account for the intercomparisons:" During the first 6 measurement campaigns, HONO was measured by the sensitive LOPAP (Long Path Absorption Photometer) technique (Heland et al., 2001; Kleffmann et al., 2002), which showed excellent agreement with the (absolute) DOAS measurements, both in smog chamber and in urban atmospheric conditions. The good performance of the LOPAP instrument is related to the efficient correction of chemical interferences (Kleffmann et al., 2006). During MCMA-2006, HONO was measured using LP-DOAS (Dusanter et al., 2009 and references therein), which selectively detects HONO based on it specific UV absorption, thus free of sampling artefacts and chemical interferences (Platt and Perner, 1980). During the New York campaign (Ren et al., 2003) HONO was measured by aqueous-phase scrubbing and high performance liquid chromatography (HPLC) analysis (Huang et al., 2002), which has been used (also in a modified form) in several field measurement campaigns (e.g., Zhou et al., 2002; Ren et al., 2006; Ren et al., 2010; Zhang et al., 2012). It is worth mentioning that HONO measurements under very clean conditions, using wet-chemical techniques (other than LOPAP) have been reported to be biased by uncorrected interferences leading to artificially high HONO/NO_x ratios (Kleffmann et al., 2006; Kleffmann and Wiesen, 2008; Su et al., 2008, Lu et al., 2010; Zhang et al., 2012). Recent studies acknowledged this problem and corrected for these interferences (e.g., Su et al., 2008) or modified the technique to minimize them (e.g., Ren et al., 2010; Zhang et al., 2012). Under high NOx polluted conditions this problem is less pronounced (e.g., Kleffmann et al., 2006; Kleffmann and Wiesen, 2008). In the current study, only HONO measurements by LOPAP are considered for low NO_x conditions while for high NOx conditions, LOPAP, DOAS and aqueous-phase scrubbing and HPLC or ion chromatography (IC) analysis techniques are used.".

3. We respectfully disagree that the introduction is too long. We will nevertheless try to make it more to-the-point and less comprehensive in the revised version of the manuscript.

4. We did not investigate the dependency of unknown HONO sources on the aerosol surface area (as these data either were not measured or not available to us), it is not stated anywhere in the manuscript that HONO production does not depend on aerosol surface area. It is not expected that unknown HONO production should depend on aerosol surface area. Unfortunately, referee#1 did not provide any recent reference(s) for this expected strong dependency on aerosol surface area. The formation of HONO on humic acid like aerosol surfaces has been investigated in the laboratory by Stemmler et al. (2007), which was also light dependent. However, this photochemical source of HONO on aerosol surfaces was negligible compared to HONO photochemical formation on ground sources (Stemmler et al., Nature, 2006), even if all aerosol particles were consisted of only humic acid (Stemmler et al., 2007). In a recent field measurement campaign, in which nighttime gradient HONO, NO_x and aerosol particles were simultaneously measured, formation of HONO on aerosol particles was shown to be of minor importance (Kleffmann, 2007 and references therein). In his review on HONO daytime sources, Kleffmann (2007) stated that "recent kinetic studies on soot, aqueous and humid surfaces and organic particles, for which either activation or small uptake coefficient imply a minor importance of particles for the heterogeneous HONO formation in the atmosphere". Several modeling studies investigating the impact of HONO formation on aerosol surfaces and found it of negligible importance compared to the ground source, even when considering higher NO2 reaction probability on aerosol surfaces during daytime under both, high NOx (e.g., Li et al., 2010, Mexico City) and low NOx (e.g., Li et al., 2012, Pearl river Delta, China) conditions.

Furthermore, the unidentified HONO sources have been calculated for both high NOx (Santiago campaigns) and low NOx (HOxComp). The HONO/NO_x ratio used to parameterize HONO mixing ratio, is derived based on HONO/NO_x correlations of 15 filed measurement campaigns, under widely different atmospheric conditions ranging from urban high NO_x, rural and rural/remote low NO_x regions (see sec. 2.2). To the best of our knowledge, this explicit comparison study for HONO dependency under such extremely different conditions was not done before and the results presented here are novel and therefore, we think they are very useful in the understanding HONO unknown sources and for the empirical parameterization of HONO in global models.

5. We respectfully disagree with referee #1 that HO_x recycling is known to be effective down to 0.1 ppb NO_x. First, there is no typical value above which HO_x recycling is effective. HO_x recycling is efficient if NO levels (not NO_x as stated in the comment) are sufficient to recycle all RO₂/HO₂ radicals that have resulted from the OH oxidation of VOC (OH+VOC \rightarrow RO₂/HO₂) back to OH (RO₂/HO₂ + NO \rightarrow OH) (e.g., Elshorbany et al., 2010a; 2010b). Therefore, the NO level at which HO_x recycling is efficient depends on the abundance of RO₂/HO₂ resulting from oxidation of VOCs, which can vary by orders of magnitude between different regions, different seasons and for different atmospheric conditions and even during the day. For example, during the HOxComp campaign (Elshorbany et al., 2012), two photochemical chemical regimes were experienced by measurement site, high NO_x in the early morning and low NO_x during afternoon. During the high NO_x period (NO>1 ppbv), HO₂/OH ratio was only about 19 indicating high recycling efficiency but increases strongly to an average value of 125 during the low NO_x period (NO< 1 ppbv) indicating low recycling efficiency. It is quite apparent from Fig. 12 that enhancements in HO_x and secondary oxidation products (in this region) appear only when NO levels are higher than 1 ppbv in excellent agreement with a detailed sensitivity analysis during HOxComp (Elshorbany et al., 2012), a measurement campaign held at this time within this region. Therefore, the results are representative and in excellent agreement with previous studies.

6. This study is not aiming at validating the model results, which has been already previously extensively evaluated (Jöckel et al., 2006; Lelieveld et al., 2007 and several others, see the text). In this study, we investigate only the impact of using realistic HONO levels on the simulated secondary oxidation products. The ozone levels in New York City indeed reached 141 ppbv in June 2005, as shown in the mentioned reference (page 12904, line 25). Also in 2006, hourly maximum ozone exceeded 168 3 ppbv times in July. (http://www.dec.ny.gov/docs/air pdf/06O3exc.pdf). Thus, these values are quite close to the measured values and are expected during this time of the year in New York City. The O₃ enhancement of 40 ppbv obtained from Fig. 13 represents especially polluted conditions (as mentioned on page 12904, lines 16) and corresponds to about 30% O₃ enhancement, which is typical and in agreement with all previous studies

applying small-scale models as mentioned on page 12905, lines26-29 and page 12906, lines 1-13. Thus, these results are meaningful.

7. The comparison in this comment is not accurate. Owing to the coarse model resolution, it is not necessary that the model results match the HONO field measurements exactly. The successful simulation of HONO in these regions (as stated in the comment), based on the HONO/NO_x ratio from 15 field measurement campaigns worldwide (not only for specific regions), indicates that this parameterization is also successful and representative. In the model we implemented the HONO/NO_x ratio to simulate HONO levels but not O₃. The agreement obtained for O₃ enhancement between our model results and previous studies employing small-scale models shows that our model results of HONO impact on global O₃ chemistry are consistent and in agreement with previous studies. In addition, referee #1 did not show any data that is in variance or significantly deviate from our results. Therefore, we do not see any conflict from the good agreement of O₃ enhancement between our model results and previous studies.

8. The "the buffering effect" of $HONO/NO_x$ is meant here to mention that the $HONO/NO_x$ ratio does not change with altitude in contrast to $HONO/NO_2$ as discussed in detail in page 12893, lines 8-18. For more clarity these words are deleted and the statement is rephrased.

9. This statement in the conclusion is discussed in details in sec. 3.2.1, page 12895. For further clarification the following statements will be added on line 19, page12895, "Similarly, Elshorbany et al., (2009a) showed that the HONO heterogeneous rate constant in the dark (calculated based on HONO/NO₂ ratio) correlates inversely with the wind speed. The positive correlation between HONO/NO_x and the wind speed in Fig. 4 is related to the anti-correlation typically observed between HONO and NO₂/NO_x ratio for all investigated campaigns (see Table 1)." in the revised manuscript.