Reply to Referee 2:

We would like to thank anonymous referee 2 for his/her detailed and helpful comments, helping us to improve the manuscript.

Page 15124 / Section 2: While I can understand not wanting to repeat the details of each instrument in this manuscript, it would very helpful for the reader to get a better idea of the possible systematic and random uncertainties of the instruments and the data that went into the interpretation in Section 3. If the authors do not want to write much text, a table with this information could be added.

> *Information about uncertainties of the instruments has been included in section 2 (see also comments to referee 1)*

Page 15125, lines 8-14: What was the residence time in the tubing? Would the O3 + NO reaction in the tube shift the NO/NO2 pseudo steady state?

> *The residence time in the tubing was about 3 seconds. The shift in NO mixing ratios is thus in the order of the given instrument error of about 5 % for NO/*O_3 *mixing ratios measured during the campaign.*

Page 15128, line 26: As NO enters the calculation of [HONO]PSS, isn't it obvious that [HONO]PSS shows a correlation with NO? I am not sure that I follow the author's argument here. I am also unclear what the significance of a correlation of HONO with NO and NO2 is. Shouldn't the HONO formation rate be correlated with NO2, rather than HONO itself? Please clarify this section.

> *Indeed, it is not surprising that [HONO]PSS correlates with NO. This sentence was misleading as the comparison with OH was missing. The dependence was meant in a way that most of the variation of all input parameters was due to NO ([NO]* r^2 *=0.78, [OH]* r^2*= 0.006 and j(HONO)* r^2 *= 0.01). The statement has been clarified accordingly.*
>
> *The focus of this study is that the HONO formation rate correlates with* NO_2 *(and j(*NO_2*)).*

Page 15129, lines 18 – 21: This statement is only correct if NOX is predominantly in the form of NO2 (as it seems to be the case in most of the nights here). In the absence of ozone, for example, addition of NOX in the form of NO at night could, according to our current understanding, decrease the HONO/NOX ratio without any actual impact on HONO chemistry. Why not use HONO/NO2 in this discussion to avoid misunderstandings. Adding HONO/NO2 in Figure 1 and 2 may also be helpful in understanding the NO2 to HONO conversion.

> *As this paper was focused on daytime chemistry we used HONO/NOx because this ratio is more appropriate during daytime. HONO/NOx is insensitive to NOx inter-conversion reactions. During daytime NO is a non-negligible source of HONO and values of direct emissions are given as HONO/NOx ratios. Additionally the ratio of HONO formation rate and* NO_2 *(which is more appropriate /see above comment) is discussed in detail in the following section. Therefore we prefer to keep HONO/NOx in Fig. 2. As the comment of the referee refers to the nighttime formation, we agree including* HONO/NO_2 *into Fig. 1 although NO values were mostly below the detection limit and therefore HONO/NOx ~*HONO/NO_2 *during nighttime. We respond to the concern of the referee about chemical alteration of the ratio due to NO emissions by expanding the excerpt of Su et al. (2008) who discussed the problems of the scaling approaches in detail.*

Page 15130, line 25 ff: A recent ACP paper by Wong et al., 2011, provides a detailed description on the impact of nocturnal vertical mixing on HONO, as well as new data on the nocturnal NO2 to HONO conversion.

> *The paper by Wong et al. 2011 has been included into the discussion.*

Page 15132: The discussion of advection and HONO from industrial sources should be expanded. The authors state that a large industrial area was located 15km upwind from the site. While the time from this source to the site of <2hours is too long to play a role during the day, it may have impacted nocturnal and morning HONO. How well is the HONO/NOX emission ratio of this source known?

> *To our knowledge, no HONO/NOx emission rates are available for this industrialized area in the literature. That is why we stick to the values reported by Kurtenbach et al. (2001). This might be justified, as traffic will still be the dominant NOx source in Huelva in spite of the industrial plants located there. Furthermore, applying a net conversion rate of* NO_2 *to HONO of about 1 %* h^{-1} *(which is within the range of reported values), heterogeneous formation (Phet) adds 1 to 2 % to the HONO/NOx ratio depending on transport time (1-2 hours). That is why we cannot quantify the influence of direct emissions from Huelva without knowing their emission ratio, the amount of emissions and the transport. We added this discussion to section 3.2.2 "Including "known" heterogeneous formation into PSS calculations ", as in accordance with the comments of referee 1 we moved all of the discussion regarding Phet to this section.*

Page 15134, lines 7-14 and Figure 4: Please provide a more detailed statistical analysis of these correlation plots. Add a linear fit line to Figures 4a,b, as it is very difficult to see if there is indeed a correlation between HONO production and J(NO2).

> *We tried to add linear fit lines to Figures 4a,b, but the correlations are weak. That is why we call it "correlation scheme" instead of correlation (see comment below). We prefer adding an upper limit line to Fig. 4b, as this seems to be better constrained. The weak correlations are due to the inclusion of all available data points which carry all uncertainties discussed in the manuscript. Therefore, we prefer including a linear fit also to Fig. 5 (as insert), as these data are only from clear days and thus assumed to be less influenced by uncertainties (see original manuscript).*

Page 15134, line 24: Please clarify what a "correlation scheme" is.

> *We should rather state that after applying the normalization with* NO_2 *all data points lie on or below an upper limit which is increasing linearly with j(NO2).*

Page 15135 / Section 3.4: As already discussed above, this section should be expanded to consider other possible daytime sources. For example, a recent manuscript by Zhou et al, 2011, proposed the photolysis of HNO3 on the canopy as a source of daytime HONO. Other publications, have proposed the photo-enhanced conversion on humic acid like substances on the surface and the aerosol. Some of these mechanisms are discussed in the introduction but are not considered sufficiently in the data interpretation.

> *The current study was focused on the HONO sources which could be directly calculated from the available data. Therefore, we avoided to speculate about the magnitude of other sources for which we lack important information for their quantification. As both referees claimed that these sources should be considered, we included rough estimates of these sources based on the numbers given in the respective publications (Stemmler et al., 2006 and Zhou et al., 2011).*

Page 15135, lines 4 – 21: This text seems out of place here and should be moved to section 3.5, or removed completely if a better OH budget calculation cannot be presented.

> *The paragraph (Lines 6-21) has been removed completely.*

Page 15137/ Section 3.5: As mentioned above, this section together with Table 1 is a half-hearted attempt to provide an OH budget calculation. The authors should decide whether they want to extend this discussion to consider other OH sources, such as aldehyde photolysis, O_3 + VOC, and HO_2 +NO, or shorten this section and remove Table 1 to solely show the comparison of OH formation from ozone and HONO photolysis.

> *As we removed Table 1 and the above mentioned paragraph (i.e. the whole OH budget discussion), section 3.5 only shows the comparison of OH formation from ozone and HONO photolysis as suggested by the referee.*

Figure 1: It is basically impossible to see daytime HONO in this figure. Please plot HONO and $[HONO]_{PSS}$ on a log scale to show both nighttime and daytime periods with sufficient resolution.

> *Figure 1 has been changed. HONO and $[HONO]_{PSS}$ are now plotted on a log scale.*

Figure 2: Please use colors in this figure. The gray scales are very difficult to distinguish.

> *We believe that this comment was referred to Fig.3 as Fig.2 does not use grey scales. We changed Fig. 3 in accordance with the comments of both referees.*

Cited literature:

Kurtenbach, R., Becker, K. H., Gomes, J. A. G., Kleffmann, J., Lörzer, J. C., Spittler, M., Wiesen, P., Ackermann, R., Geyer, A., and Platt, U.: Investigations of emissions and heterogeneous formation of HONO in a road traffic tunnel, Atmos. Environ., 35, 3385-3394, 2001.

Stemmler, K., Ammann, M., Donders, C., Kleffmann, J., and George, C.: Photosensitized reduction of nitrogen dioxide on humic acid as a source of nitrous acid, Nature, 440, 195-198, 2006.

Zhou, X., Zhang, N., TerAvest, M., Tang, D., Hou, J., Bertman, S., Alaghmand, M., Shepson, P. B., Carroll, M. A., Griffith, S., Dusanter, S., and Stevens, P. S.: Nitric acid photolysis on forest canopy surfaces as a source for tropospheric nitrous acid, Nature Geoscience, 10.1038/NGEO1164, 2011.