We would like to thank all the referees for their comments. Here are our responses:

REFEREE #1

Page 23112, lines 8-9. Isn't ozone destroyed at low NOx because the rates of HO2+O3, and O3 photolysis to O(1D) followed by reaction with water vapor become larger than the rate of HO2+NO plus RO2+NO? While radical-radical reactions become relatively more important at low NOx, they aren't the cause of net ozone loss.

The sentence has been changed to:

"At low NOx, peroxy radical self-react and react with O3; the reactions of peroxy radicals, and HO2 in particular, with O3 causes net chemical loss of ozone."

Page 23114, line 8. Suggest adding references to Andres-Hernandez and Cantrell regarding dual channel chemical amplifiers.

The references have been added.

Page 23114, line 21. Just a comment on the "absolute" nature of CRDS. The technique does determine the absolute amount of an absorber in the laser beam path, but issues such as inlet losses, artifacts (such as from NO+O3 in ambient air), and interfering ab sorbers require careful assessment. Regarding the comparison, I am surprised by the degree of scatter. You should discuss the relative uncertainties in the two measurements and assess whether the amount of scatter is what would be expected. Also, is the 9% slope within the expected uncertainties?

The uncertainties of the CRDS and the Scintrex are 5% and 20%, respectively (Osthoff et al., 2006, Fleming, Ph.D. thesis, 2005), including the uncertainty in the water correction for the Scintrex NO2 detector. Therefore, a 9% agreement between the two instruments is within the instrumental uncertainties, even though this is a relatively large discrepancy for two instruments measuring NO2. The NO2 uncertainties have been added to the text.

The comparison between the two instruments was not a formal comparison, but instead a rather limited test of the Scintrex NO2 response. Uncertainties from both the slope and scatter are larger than ideal for a direct measurement of NO2, but have been taken into account in calculation of the uncertainty associated with the RO2 measurement itself.

The phrase "absolute" has been stricken from the text since, as the reviewer correctly points out, this property of the CRDS measurement is not directly relevant to this comparison.

Page 23115. This paragraph discusses the calibration of the chemical amplifier (chain length) and issues related to the observation of the peroxy radicals. I suggest actually give the chain length or range of chain lengths used during the study. Line 20-21. The agreement between the two calibration methods does not demonstrate that the data does not require a humidity correction. The agreement could be entirely fortuitous. The argument needs more information. For example, based on previous studies, what is the expected humidity correction? Likely it is at least a factor of 2 for the humid marine boundary layer. This casts a big question over the absolute amounts of peroxy radicals from this study. In the future, I suggest using the method of standard addition of radicals to ambient air to overcome this problem. It does appear that perhaps there are discrepancies between the reported laboratory studies of the humidity effect and what is actually observed in ambient air studies.

We agree with the referee(s) that the issue of the water correction for the chemical amplification

method is important and may require more investigation in the laboratory and in the field. It is also possible than the dependence of the CL for RH>80% (the limit of most reported lab experiments) cannot be reliably extrapolated. It is quite clear from observations at higher temperature and high humidity, that this phenomenon is reproducible. We have observed this during the AMMA campaign on aircraft measurements (Andres-Hernandez et al., ACP, 2010), as well as in the tropical forests of Borneo and in the tropical Atlantic from surface measurements.

The mechanism of the humidity interference remains unclear, although it appears to be heterogeneous (i.e., wall loss) in origin. If it is so, it is possible that the walls of the inlet retain some water during the period of the calibration, when the instrument has been operated for a long time under high RH conditions; therefore the calibration with zero air is effectively carried out in humid environment (thus the data will not require a correction, which would be implicit in the calibration factor). Such a mechanism may explain the discrepancy between the laboratory studies and field observations, as the referee points out. It may also explain the similarity between the calibration factor with CH3O2 in ZA and with CH3CO3 in ambient air. The two species should have the same CL (see answer to issue raised by referee #2), if measured under similar conditions. Finally, prior studies have shown that the RO2 levels for very clean conditions can be rather accurately calculated (e.g., Fleming et al., ACP, 2006). A simple box-model with CO-CH4 chemistry gives around 40 ppt of HO2+RO2, similar to observations (without correction) of 30-35 ppt in the open ocean. Applying a humidity correction factor would lead to unrealistic concentrations of more than 100 ppt. Andres-Hernandez et al. (ACP, 2010) reported a similar issue when operating in the high RH environment of Niger, and although their instrument was aboard an aircraft (as noted by referee #2), they performed their calibration at the ground, so the two experiences are not that dissimilar. It is worth noting that the non-humidity correction does not introduce a systematic bias as would be expected against models (see Andres-Hernandez et al., ACP, 2010).

Considering all these issues, we feel justified in not applying the humidity correction to the data for this study. Experience from this study is not necessarily general to all past experiments with such instruments, although, as mentioned, this has been observed in other occasions.

Thank you for pointing out the impact of humidity on the NO2 measurement itself. What does this imply for the comparison with the CRDS discussed earlier?

Nothing, the correction has been applied to the NO2 measurements before comparing to the CRDS measurements.

Lines 27-29. In discussing the total uncertainty, do the values refer to 2-sigma uncertainties (or something else). The uncertainty interval should be stated. The integration time affects these values, so it would be good to state it here as well (even if it is stated elsewhere in the paper).

This information has been added to the text.

Page 23116. Other measurements aboard the ship are discussed. Again, integration times and uncertainty intervals should be included in the discussion. The words "accuracy" and "uncertainty" appear to be used interchangeably. Just make sure all is clear to the reader.

Complete details of all the instruments on the RHB are given in Parrish et al., 2009. Some more information about the relevant instruments has been added and the use of "accuracy" and "uncertainty" corrected.

Page 23119, lines 4-6. This statement implies something about the dependence of the total peroxy

radical amount on the amount of VOC and NOx. It would be helpful to the reader if this discussion was more specific as to what is meant. The argument that values are higher in the most polluted regions does not appear to be supported by the average values presented in Figure 6. Does this statement refer to the mean or median values, or to the outliers? Line 9. The range of times included in "daytime" data should be spelled out. Lines 15-17. It would be helpful if NOx values were given for these conditions in support of this statement. Lines 18-26. This nighttime observations are interesting and this reviewer would like to see discussed more. On the night of 7 September, what were the NOx and O3 levels. NO3 reacts with aldehydes and alkenes, while O3 only reacts with alkenes. Is there enough information, perhaps including some modeling to say what reactions lead to the high peroxy radical levels? Were there NO3 measurements anywhere in the region that can be used to give an indication of its importance?

Figure 5 clearly shows that the highest values are from the Industrial Areas. The statement the referee is pointing out refers to the average values, and Figure 6 show the medians, hence the confusion. The text has been amended to clarify.

The range of times and the NOx values have been added. Regarding the night-time event please see reply to referee #2.

Summary statistics of the observations appear to be given for some of the regions, but not all. I suggest a table summarizing (mean and/ or median, standard deviation, 95% range, numbers) the daytime and nighttime values observed for each of the 5 regions (filtering by some criteria as appropriate), and also including statistics for NOx and some measure of the VOC impact (e.g. total OH reactivity or some other measure) to go along with Figure 5. This would help the reader understand the changing environments and the resulting change in peroxy radical amounts.

This table has been added.

Page 2312, line 12. Suggest changing "in the Ocean" to "observations in the Ocean category" or something similar. This paragraph discusses the sources of peroxy radicals from reactions of OH. What is the relative importance of these reactions compared to direct formation, say from oxygenated VOC photolysis? In other words, a budget of HOx+RO2 would be instructive for the reader.

The category "Ocean" has been renamed to Open Ocean. The relative production rates of peroxy radicals from the oxygenates (photolysis versus reactivity with OH) has been added to the text.

Page 23121, line 24. This statement appears to contradict the one on page 23119, line 4-6. Suggest clarifying and/or making the discussion consistent.

The statement refers to a particular period of the day (sunrise) not to the whole day (as the previous statement), therefore they are not inconsistent. The text has been amended to clarify.

Page 23122, lines 2-14. This discussion of the total peroxy radical amount versus NOx is interesting. In lines 7-9, I think you mean that the "average" levels were more influenced by outliers. Since you have calculated the expected peroxy radical levels, a plot of the observed to calculated ratios versus NOx would also be interesting. This would tell us whether we understand peroxy radical chemistry at high NOx, something that has been questioned in the literature.

Yes, we meant to say "average levels" and this has been corrected in the paper. A proper comparison with modelled HO2+RO2 and discussion of the modelled-measured agreement is however beyond the scope of this paper – which is focused on the measurements and ozone production – and will be the subject of another upcoming publication.

Page 23122, lines 19-20. Add the word "rate" or "rates" several times, for example after the words "photochemical ozone formation" and "photochemical production and loss", but also in several other places throughout the paper. Line 23. The loss rate of ozone "includes" the rate of the reaction of O(1D) with water vapor, but is also destroyed by HO2+O3. In very polluted regions (such as studied near Houston), the loss of NO2 (reaction with OH or other processes) results in loss of Ox which is effectively loss of O3.

The words "rate" and "rates" have been added at the appropriate places. The referee is correct in pointing out that removal of NO2 also leads to loss of ozone. The reviewer is correct that NO2 loss is an indirect O3 loss via Ox. We have added a sentence to the text to state that loss of O3 as calculated here includes only the direct terms involving O3 itself and does not include such indirect losses of ozone as Ox.

Page 23123, equation 5. While the quenching of O(1D) is due to all molecules in the air (primarily N2 and O2), there is no rate coefficient for O(1D)+M! You, of course, mean the weighted average of the quenching rate coefficients for the two processes. It should be stated as such.

The reviewer is correct in that it means the combination of the two processes: O(1D)+M is commonly used notation in the literature; the sentence "the average of O1D + N2 O2 weighted by their abundance" has been added to the text before Eq. 5.

Several places (including lines 13-14) calculations from a box model based upon the MCM are mentioned. I suggest describing the calculations earlier in the paper, including the actual mechanism used, how the model was initialized, and what times calculated values were extracted to compare with the observations and used in calculations such as discussed here (essentially moving the discussion at the end of the paragraph and expanding upon it).

The model setup and mechanism are very similar to those used in previous modelling studies (see refs on page 23123, line 19). The model is mentioned in Section 4.1 only to provide the reader with information on the origin of OH and HO2/RO2 ratio used for the calculation of ozone formation rates. The MCM model and the discussion of its results are the subject of another paper which is currently in preparation. See also reply to comment by referee #2.

Page 23124, line 4-13. In these calculations, surely the mean values (about which the changes are made) affect the sensitivity. For example, the sensitivity to NO might be very different at 1 pptv compared to 100 ppbv. Line 12-13. The statements appears to say that the sensitivity of the Net O3 production rate to the total peroxy radical level is 1.0, but Table 1 gives a value of 0.9. This should be made consistent.

The reviewer is correct. We chose to report only the campaign averages of the SI to allow an easy comparison of the relative importance of all the parameters. The difference between the text and the table is not due to inconsistency. In the table the SI is calculated by varying the concentration of HO2+RO2 by 1%. In the text we also report the effect of a variation of HO2+RO2 by 40%. This sentence has changed to clarify its meaning. Also the variation in NetO3 when changing k(NO) has been added to the text.

Lines 13-22. The discussion makes it sound like no rate coefficients for organic peroxy radical reactions with NO have been measured. There are recommendations at least for CH3O2, C2H5O2, CH3C(O)O2, and CH3C(O)CH2O2, and there are data for many more. Generic values are only used for very large or multiply substituted R-groups. It would still be useful to estimate and present weighted values (likely different for the different VOC regimes encountered) for the RO2+NO rate

coefficient since they are so important.

The text says "experimental kinetic data are not available for every individual species in the MCM", which is correct. We do however agree with the referee that changes in that generic rate coefficient have substantial impact on the results (as indicated by the sensitivity index): an estimate of the variation of Net(O3) when k(RO2+NO) is changed by +/- 30% as an indication of the overall impact of this parameter has been added to this Section.

Page 23125, line 1. I'm not sure "diurnal" is the correct word here (applying to a 24 hour period), since the values are only shown for daytime hours. Perhaps just say that the figure shows the variation of the values versus time of day. Lines 11-12. This statement does not appear to agree with the values shown in Fig.5, where peroxy radicals change greatly over the course of the day, and NOx appears relatively constant.

The referee is correct. The reason Prod(O3) does not appear to have a diurnal variation is that the data is shown in log scale. Figure 7 has been changed and the text corrected accordingly.

Page 23129, lines 21-25. I definitely do not like the way the data are presented in Figure 10. The size of the data points and the presentation of concentration versus NOx makes it very difficult to assess how well the observations and models agree, and how well the observations from the two time periods agree. It would be helpful, for example to bin the data based on NOx level, and to show measurement-model ratios rather than absolute amounts. I find the statement that the "agreement..was very good" completely unsatisfying and unacceptable. If a comparison is to be made, provide quantitative statistics on the agreement (e.g. mean measured and modeled ratios averages 1.xx with a standard deviation of yy). Then discuss the differences given the measurement and model uncertainties. On page 23130, line 5, the term "remarkable agreement" is used. Please be quantitative about the level of agreement. In the Summary and Conclusions section (Page 23131, lines 19-25), the terms "quantitatively compared" and "agreed reasonably well" are used. I would not say that the two datasets have been compared quantitatively, or at least that the quantitative comparisons are not given. The term reasonably well is subjective and tells the reader little about how they really agreed or disagreed.

Figure 10 has been modified and now shows the averages and the standard deviations in a range of NOx bins. The text has been changed accordingly.

REFEREE #2

Does the PERCA instrument respond to halogenated species, for example high levels of ClNO2 were observed in this region, and halogen species can oxidise NO to NO2 and generate a response.

The Scintrex NO2 detector might be sensitive to ClNO2, as it is to PAN, but these species will contribute to the background signal as well as the amplification signal so they will not affect the difference between the two channels which is used to measure RO2.

Cl itself would not directly oxidize NO to NO2, though peroxy radicals derived from VOC oxidation by Cl would. Thus is seems most likely that any effect of ClNO2 is accounted for in the measurements.

Was the chain length determined for HO2, which is expected to make up a significant amount of the total HO2+RO2? (50% is mentioned later in the paper

I didn't see any values of the chain length (CL) actually given – please provide for the species for which calibration was undertaken.

The chain length was determined for CH3O2, and in one occasion, for CH3CO3. The chain length for this species are quite close, due to the prompt conversion of these species to HO2 in the high NOx environment of the PERCA inlet, so it is unlikely to be very different. CL numbers were 100-160 in one channel and 34-45 in the other channel. Despite the difference in chain lengths, the two channels gave the same response for ambient peroxy radicals once this difference was taken into account during the data workup. These numbers have been added to the text in Section 2.1.

Perhaps surprising is that the data collected did not require a correction for relative humidity given the previous literature on this and the corrections for humidity that have been published for PERCA previously. These corrections in the past have been quite large, and this is a humid environment. The other instrument mentioned (Andres- Hernandez et al reference) was aboard an aircraft, where the calibration and relative humidity dependence might be expected to be very different (sampling mode, temperatures and pressures). I am pleased that there is a humidity correction for the Scintrex. I think more discussion is required here, as there is a great deal of confusion in the literature about the humidity correction (or not) of PERCA instruments. The main argument given here is that the zero air chain length for CH3O2 and the ambient (high humidity) chain length for CH3COO2 are the same and so no humidity dependence is necessary. I am not sure that this is a valid and logical conclusion. Could it be the case that the CLs are different for dry air for the two species, and that it happens that the CL for humid CH3OCO just matches that for dry air CH3O2? Was the instrument calibrated as a function of humidity for CH3COO2 and CH3O2? Or can laboratory (or field) calibrations for these two species from previous studies as a function of humidity be used to estimate the dependence. I am surprised that the CL for CH3CO would have no humidity dependence. Although not determined here, the HO2 CL would also be expected to have a humidity dependence (and later this is assumed to be 50% of the total species measured). It is appreciated that these are difficult things to measure, but given the humid environment of this study, it is important to be as detailed as possible.

We agree with the reviewer that the lack of a humidity dependence was a surprising result given the body of literature on this topic. However, the experimental evidence is clear – not only do the "dry" and "humid" calibrations show little difference, but corrections of the scale implied by the literature would lead to unreasonably large HO2+RO2 for most of the study. We have therefore refrained from generalizing this result, but we do feel that the corrections, as applied, are appropriate for the current set of measurements. We agree that this issue is important and may warrant more investigation in the lab. For more details, please see the response to referee #1.

Regarding CH3O2 and CH3CO3, the two species should have essentially the same chain length because CH3CO3+NO forms CH3O2 in one step with 100% yield.

The dual channel instrument helps to mitigate against varying background levels of NO2 and O3 which give a response in the PERCA instrument. In the very polluted periods, what was the relative change in NO2 signal observed in the Scintrex between radical measuring and background measuring modes (i.e. dynamic range of the modulations, which will depend on the CL, and concentrations of NOx, O3 and radicals).

NO2 was highly variable throughout the campaign, but not generally variable within the 60 s interval of the modulation. So, for example, at an NO2 level of 5-15 ppbv, a background O3 level of 100-140 ppbv and HO2+RO2 level of 40-80 pptv, the additional NO2 from HO2+RO2 was 1.4-2.9 ppbv for one channel (CL = 40-45) and about 5.4 ppbv for the other channel (CL = 127-142). This signal is easily measurable.

The nighttime levels of 134 ppt are interesting, and are considered in the paper to be likely related to night-time oxidation of O3 or NO3 in the entrance to the Houston Ship canal. What was the level

of NO in the ship canal at night – one might expect this to be quite high due to local fresh sources of NOx emissions (were there any), and this may keep NO3 levels down due to the NO3+NO reaction? I think a table showing the typical levels (of e.g. NO) could be a useful addition – as well as that of supporting data (e.g. NO3 if measured). The very high HO2+RO2 levels at night are very interesting but there is insufficient data or information presented to get much insight into the chemistry responsible for this.

Night-time RO2 has not been discussed in detail in this paper, but will be analyzed in more detail in a future publication. Night-time levels of peroxy radicals of 134 ppt were not routinely observed, this was a single event, likely related to a plume emitted from one of the nearby petrochemical plants. Unfortunately NO3 measurements for that night are not available. During the event, NO was about 4 ppb and alkenes concentration was high (e.g propene = 9 ppb). This suggests ozone reactions with alkenes was responsible of the formation of such high level of peroxy radicals, because NO3 would be titrated by NO faster than it could react with any other species. This information has been added to the text.

Page 23122, line 25, rather than "atomic oxygen" it would be better to say O(3P) ground-state atomic oxygen, as O(1D) is also atomic oxygen.

This has been corrected.

Page 23123. HO2 was measured at the La Porte site, and so in line 13 something could be said about an experimental value of the HO2/(HO2+sum RO2) ratio. This could be used rather than relying solely on a model calculation for this ratio? OH was also calculated in the model, but again, this was measured at the La Porte site and so some reference to actual values rather than just model values could be discussed.

Unfortunately, HO2+RO2 was not measured at LaPorte in 2000. We acknowledge there is some confusion regarding the use and the purpose of the model(s) and the comparison with the LaPorte data and has been made clearer in the final version of the manuscript.

The only MCM model results shown in this paper are OH and HO/HO2+RO2 ratio, because they are necessary to calculate ozone formation rates. The comparison with the 2000 and 2006 observations – and related discussion of the results – are beyond the scope of this paper. The presentation of the MCM model results will be the subject of another paper.

The other model results mentioned in Section 4.3 are from a different model – not based on the MCM – tailored for the 2000 campaign. We are showing ozone production by HO2 in 2000 (modelled and measured) to have a qualitative idea of how ozone formation has changed in six years. There is no attempt to directly compare the 2000 modelled OH to the 2006 modelled OH because the focus of the paper is on peroxy radicals and ozone formation not radical chemistry in general.

A sentence clarifying the use and purpose of the two model in this paper has been added to the Introduction and to Section 4.

Page 23124. Line 4 and up to section 4.2, the MCM has a lot more detail that just a general rate constant for RO2+NO. It is true that for RO2 where k is not known, a generic value is used, but for those R where a value is known, it is present in the MCM. How different are k for RO2 where k is known? The approach here seems too simplistic given that k data do exist for a number of RO2 species. Especially given that the value of N(O3) is sensitive to the rate constant for RO2+NO.

The MCM protocol (Saunders et al., 2003) says that for "the vast majority of the ca. 900 RO2 radicals, however, kinetic data are unavailable, and the assigned rate coefficients are based on two generic expressions".

In our calculation we used a generic rate coefficient for the reaction of the sum of organic peroxy radicals with NO, which is the same used in the MCM for the majority of alkyl peroxy radicals (when kinetic data are not available) and is based on the rate coefficients of CH3O2 and C2H5O2. While we don't think this approach is simplistic, the calculation has been done for k(RO2+NO) varied by +/- 30% and the results has been added to Section 4.1 (as also requested by referee #1).

Page 23128, what is the source of isoprene. Biogenics are listed, but is there an anthropogenic source of isoprene, particularly given the very rich mixture of VOCs at high levels that were observed? High correlations of 1-3 butadiene and isoprene have been observed in the past.

There are indeed industrial sources of isoprene in Houston, as noted, for example, by Stutz *et al.* (Atmos. Environ, 44, 4099-4106, 2010). However, Gilman *et al.* (JGR, 114, D00F06, 2009) suggest that the isoprene is mostly biogenic. This has been made clearer in the text.

Page 23129. Line 15. Measurements at La Porte in 2000 are compared with the measurements from this paper, but there is no reference given, e.g. for the HO2 measurements. Wasn't OH measured as well? I would like to see more discussion of the assumption of HO2 being approximately 50% of the total HO2+sum(RO2). HO2 and the sum HO2+RO2 have been measured at quite a few other urban areas, and so there is further information that can be used to support this or otherwise.

The reference for the HO2 measurements has been added. OH was also measured at the site in 2000, but this species is not a subject of this paper. The estimate of HO2 being half of HO2+RO2 derives from the results of the MCM box-model. A sentence that clarifies this has been added to the text.

Also, Figure 10 displays a large range of modelled and measured values (not explicitly said which measurements agree very well with the model presumably this statement means for all four parameters in the 4 panels?), and the conclusion that there is very good agreement (line 25 on page 23129) is not obviously supported by this figure. Plots of modelled versus measured would enable this statement to be made with more confidence, and the current discussion is very inadequate. What is meant by very good agreement? Ratios need to be given, and the errors in the measurement and the model considered in order to be quantitative about what very good agreement means. This would enable the conclusions given in section 5 to be made with more confidence.

This figure and the related text has been changed to make it clearer. See reply to similar comment from referee #1.

Page 23129, line 29, a reference needs to be given for the P(O3) calculation from HO2 measured in the 2000 campaign, or is this done in this paper

The calculation is done in this paper.

REFEREE #3

1) It has been well documented that the PERCA technique generally requires a correction due to a water dependence of the chain length. As a result, it is surprising that the data presented here did not require a humidity correction. The authors appear to base this conclusion on measurements of the chain length for CH3O2 in dry air that were similar to measurements of the chain length for CH3O2 in dry air that were similar to measurements of the chain length for CH3C(O)O2 measured in humid air. Was the chain length measured as a function of relative humidity? How was the water dependence minimized? More details on the instrument calibration as a function of relative humidity are needed to give the reader more confidence in the

measurements.

Again, the reviewer raises a point raised by the previous reviewers, and one that we considered in some detail prior to submission of this paper. Please see our reply to similar comments by referees #1 and #2 above, for details.

2) The measured total peroxy radical concentrations show a large variability as shown in Figure 4, and much of this variability is due to the different NOx and VOC concentrations encountered during the measurement period. Although the diurnal profiles are shown with the NOx profiles for each location in Figure 5, it would be useful to include a time series of NOx and perhaps VOC concentrations in Figure 4 to better illustrate the influence of NOx and VOCs on the day-to-day variability, and especially the high nighttime values shown in this Figure.

We have added NOx and selected VOC (propane, ethene, formaldehyde) to Figure 4.

3) The calculation of the net production of O3 requires a knowledge of the concentration of HO2 and OH, and the authors state that they used values based on the results of a box model based on the MCM. What are the values of [OH], [HO2], and [HO2]/[HO2 + RO2] used for this calculation in each region, and how do these calculated values compare to any available measurements from TexAQS 2000 or in other similar environments?

A figure with OH and HO2/HO2+RO2 ratio has been added. See also reply to similar question by referee #2.

4) The plot of P(O3) as a function of NOx in Figure 10 does demonstrate that ozone production measured in 2006 is NOx limited. However the authors also claim that the measured and calculated values of HO2, HO2 + RO2 and P(O3) from 2000 and 2006 agree very well, but this is not clear from Figure 10. Although difficult to read, it appears that the measured HO2 + RO2 from 2006 is significantly lower than 2x HO2 measured in 2000. A plot HO2 + RO2 vs. 2x HO2 binned for different NOx concentrations would provide a clearer comparison of the agreement, which could then be quantified. Similar plots for the remaining comparisons in this Figure would also allow for a more quantitative comparison of the agreement between the measurements and model.

This figure and the related text has been changed to make it clearer. See reply to similar comment by referee #1.