Reply to Ref. #2

First of all we want to thank this reviewer for the positive assessment of our manuscript and the constructive and helpful suggestions.

General comments
The study by Y. Wang et al. reports on a detailed evaluation of state-of-science retrievals of HONO column densities from MAX-DOAS measurements of scattered UV radiation during an intensive measurement campaign in 2013 in Mainz, Germany. Following a comparison of HONO columns retrieved by 11 different groups, the authors present an in-depth analysis of the retrieval settings required for optimal fitting. This analysis also allows them to present a characterization and breakdown of the error budget of the HONO retrievals. Both aspects of the paper are scientifically important, very suitable for AMT, and in my opinion help to improve and better understand the MAX-DOAS HONO retrievals.
Strong about this manuscript is that a substantial number of dedicated and relevant sensitivity tests have been carried out to improve the fitting approach, and at the same time characterize the fitting errors. The team makes a strong case that using sequential reference spectra instead of once-per-day noontime reference spectra works best, that water vapour absorption should be accounted for in the fit, and that the 335-373 nm fitting window gives most robust retrieval results. The comparison between the sensitivity study results and the discrepancies between HONO columns observed by different groups provides excellent potential to interpret theoretical and practical uncertainties in the retrievals.
I recommend that the paper is published in AMT, but the authors should first clarify a number of issues listed below, and make the manuscript much better readable.

Author reply:
Many thanks for the positive assessment! We modified the paper based on the comments from you and reviewer 1. In order to make the main text readable without the supplement, we added some important numbers in the main part of the paper (in the parts related to the supplementary figures). We hope the revised manuscript is more smoothly readable.

Major issues:

1) The title does not cover the aspect of error analysis that is certainly an important component of this paper. I suggest modifying the title accordingly.
Author reply:
We followed the suggestion and added "error budget" in the title.

2) The paper is too long. In may places too much information is provided. There are too many references in the text to the Supplementary Material and such interruptions prevent a smooth read. The manuscript should be streamlined in many places. As an example, on page 8, L31-32 and P9, L1-15, much of the text is about supplementary figures supporting the material in Figs. 4 and 5. Isn't the material presented in Figs. 5 and 5 convincing enough to stand on its own? It would be more logical to discuss the results shown in Fig. 4 and 5 more extensively and only at the last instance mention that there is support to be found in the supplementary figures. Another option to

make the manuscript more concise is to refrain from giving all of the available information for both the FRS and noontime reference spectrum once the recommendation is given to prefer the FRS method. The same holds for the fitting windows that are ultimately not used.

Author reply:

Thanks for pointing the problem out. Based on the suggestion of the reviewer, in order to make the main text independent on the supplement, we modified the section 3.3 and section 4, in which many relevant figures are given in the supplement. In the revised version the main results are directly described in the main text. The reader doesn't need to see the figures in the supplement. These figures are only referenced in the text to allow the most interested reader to see the detailed results.

Many thanks also for the suggestion of the second option! However, we decided not to follow this suggestion. It is true that the recommended settings, especially the selection of the wavelength range and the selection of the FRS are mainly derived from the sensitivity analyses in section 4. However, it is also important to derive the same conclusion from the comparison of the results from the different instruments, because usually not all apects relevant for real measurements can be covered by the analysis of synthetic spectra. Thus we prefer to keep the retrieval results using different FRS in the main part of the paper.

3) I'm not sure if the order of the sections is optimal. If I'm correct, the 11 retrieval groups use the optimal fitting window (335-373 nm) and settings to obtain their results presented in section 3, but the motivation for this is only given in section 4. Isn't it better to present the sensitivity studies and corresponding recommendations before the actual intercomparison? This would also prevent the need to point forward to sections still to come (e.g. on P6, L14-15 "see Section 4.1")

Author reply:

The suggestion of the reviewer is also logic. However the relationships between the different parts of the paper are not only valid in one direction. For example, also from the experimental results part of the sensitivity studies performed in section 4 are motivated. Thus we prefer to keep the general structure of the paper as it is. However to better guide the reader through the manuscript, we added a clarification in the beginning of section 3:

"HONO presents prominent absorption structures in the spectral range from 335 to 390 nm. The DOAS technique (Platt and Stutz, 2008, and references therein) can be applied to spectra of scattered sunlight to retrieve SCDs of HONO. In this section we present the inter-comparison of HONO SCD results derived from real measurements and synthetic spectra between the participants. For the analyses of both sets of spectra, recommended baseline settings for the DOAS spectral analysis are provided. These baseline setting are derived from the sensitivity studies outlined in section 4 and also based on the experiences in Hendrick et al. (2014). The details of the baseline setting are given in Table 2 and described in section 3.1."

The beginning of section 3.1 is also modified accordingly.

4) The text in the manuscript is sometimes too vague. For instance in the abstract, the last sentence reads "However, systematic uncertainties limit the reliability of the results." Since you have a pretty decent quantitative estimate of the systematic error of the HONO columns, please indicate what you think is the detection limit, and how frequently you think this is being exceeded in practice. This gives potential users of the data a sense of the usefulness of the HONO

retrievals, for instance in the context of the diurnal cycle of HONO columns. Also, see many minor comments below, asking for clarifications.

Author reply:
We modified the sentence in the abstract as "In summary for most of the MAX-DOAS instruments for elevation angle below 5°, half daytime measurements (usually in the morning) can be over the detection limit of HONO delta SCD of 0.2×10^{15} molecules cm^{-2} with a uncertainty of ~0.9×10^{15} molecules cm^{-2}."

5) The role of clouds in the retrieval remains under-exposed. It would be interesting to distinguish the quality of the spectral fits under cloudy and clear-sky conditions.

Author reply:
Thanks for the suggestion. We checked the fit error of HONO dSCD under cloudy and clear days. We found the errors are quite similar. The reason is the most of instruments can automatically change the exposure time of spectrometer based on the sunlight intensity. Therefore the similar exposure saturation level is reached during clear and cloudy days. We clarified this point for the discussion of Fig. 4 in section 3.2 of the manuscript: "In addition fit errors of HONO dSCD under cloudy and clear days are quite similar due to the fact that the MAX-DOAS instruments automatically change the exposure time of spectrometer based on the brightness of the sky. Therefore the similar exposure saturation level is reached during clear and cloudy days.".

Minor issues:
1) P2, L14: "of the fitted from the simulated real HONO delta SCDs". Hard to follow, please rephrase.
Author reply: The sentence is deleted in the revised version.

2) P2, L21: "tropospheric atmosphere" → troposphere
Author reply: corrected.

3) P3, L26: I think it would be appropriate to introduce the 11 groups participating in the MAD-CAT campaign here.
Author reply: considering that not all the groups joined this study, we add the MAD-CAT website link in which the 11 groups are listed.

4) P4, L22: "seven of all of the eleven" → Seven of the eleven.
Author reply: corrected.

5) P4, L30: repetitive to mention the 12 June – 5 July period here since it was in 2.1
Author reply: The sentence is deleted.

6) P5, L15: it is unclear at this stage what sigma^2 NO2 represents and what it is used for. This has to do with the ordering of the section (were section 3 and 4 reversed at the last minute?)
Author reply: As the reply to the major issue 3, we prefer to keep the structure as it is. For this point, we added a note of "(the details are given in section 4.5)".

7) P6, L29-30: is there any physical or chemical reason why HONO dSCDs are high on 3 July 2013

Author reply: The high HONO in the morning is not only on 3 July 2013, but also on many other days (see Fig. 2a). Photolysis and high NO_2 concentration can cause the substantial high HONO concentration in the morning. We clarified this point in the revised manuscript: "The large HONO values in the morning could be due to a high NO_2 concentration (NO_2 dSCD of up to 1×17 molecules cm^{-2}) and a fast photolysis of HONO (e.g. Hendrick et al., 2014).". Nevertheless, the chemical sources of HONO is not the topic of this study, therefore we don't discuss this point deeply.

8) P7, L31-32: please clarify what 0.01 means here. How should the number be interpreted?

Author reply: It is clarified as "1% of the mean intensity in the fit window" in the revised version.

9) End of P9, lines 1-3 op P10: difficult to follow. I think section 3.3 is in need of a clear conclusion on what we have learned from the statistical comparison. Instead, we end with a quite detailed, unsatisfying comment on something that could be wrong with one particular instrument.

Author reply: we added the general conclusion in the end of section 3.3 in the revised manuscript as "In general the consistent temporal variation and elevation angle dependence of the HONO delta SCDs and dSCDs have been retrieved from the different instruments. The discrepancy of HONO dSCDs from the fits with a daily noon FRS between the instruments is systematically larger than that of HONO delta SCDs, which can be consistently retrieved from the fits with a daily noon and a sequential FRS."

10) P10, L5-6: "real atmospheric values for real MAX-DOAS measurements"?

Author reply: The sentence is modified as "In general it is difficult to quantify the biases of the retrieved HONO dSCDs with respect to the reality in the atmosphere for real MAX-DOAS measurements as the true HONO column is not known."

11) P10, L30: "than the half of that" → than half of that

Author reply: corrected.

12) P11, L17-18: nonlinear fits . . . were not included

Author reply: corrected.

13) P12, L14: can you elaborate on the increase in HONO with an increase in H2O delta SCDs? Is there a good reason to expect this?

Author reply: The correlation is probably due to the interference of the absorption structures of H_2O with those of HONO in the DOAS fits. As demonstrated in the paper, if the H_2O cross section is not included in the DOAS fit, the H_2O absorption can contribute to residual structures, which can interfere with the retrieved HONO structures. The interference is stronger for larger H_2O absorptions. We modified the sentence to make the point clear in the revised manuscript: "These findings demonstrate that the H_2O absorption could mainly contribute to the residual structure around 363nm if the H_2O cross section is not included in the DOAS fit, and can considerably interfere with the HONO absorption. And the interference is stronger for larger H_2O absorptions.".

14) P12, L24: peek→peak

Author reply: corrected

15) P12, L26-27: this has been said already.
Author reply: The scaling factor of H_2O cross section is only mentioned in section 3.4 for the RTM simulation of synthetic spectra. Here the factor is mentioned again for the DOAS fit. They are for different activity. Therefore we prefer to keep it.

16) P12, L29: "bands" or are they rather lines?
Author reply: The HONO absorption structures are smooth as shown in Fig. 9. Therefore "bands" are better than "lines".

17) P14, L27: dependence of the Ring spectrum
Author reply: corrected

18) P15, L6-7: it would be helpful to quantify here what variations you think are due to different Ring settings. This helps is evaluating the overall error budget of the HONO retrievals.
Author reply: We added the numbers as "(about 0.35, 0.2, and 0.12 $\times 10^{15}$ molecules cm^{-2} in the spectral ranges of 335-361, 335-373, and 335-390 nm, respectively on averaged)" in the revised manuscript.

19) P16, L7: non-linear
Author reply: corrected

20) P16, L15-16: same as for section 4.5: please quantify the error associated with the intensity offset uncertainty, and conclude as to its relevance.
Author reply: The numbers are added as "which is 0.55, 0.35, and 0.25 $\times 10^{15}$ molecules cm^{-2} in the spectral ranges of 335-361, 335-373, and 335-390 nm, respectively on averaged" in the revised manuscript.

21) P16, L25: "instrumental function"→ instrument transfer function or slit function?
Author reply: slit function. We modified all the word of "instrumental function" as "slit function" in the revised manuscript.

22) P17, L4: "noises" → noise
Author reply: corrected

23) P18, L8: please calrify what the correlation coefficients refer to.
Author reply: we clarified it as "The correlation coefficients of HONO delta SCDs derived from the different instruments with the reference values" in the revised manuscript.

24) P18, L10: it would be useful here to explain the typical diurnal variation in HONO, and make clear that the retrievals are able to capture the temporal changes to large extent. Perhaps also indicate when (what column densities, those typically around noon?) the retrievals are running into detection limit issues.
Author reply: We added the description as "The maximum value of the HONO delta SCDs is about 6×10^{15} molecules cm^{-2} and usually in the morning. The HONO delta SCD rapidly decrease

after sunrise due to the photolysis of HONO, and below the detection limit of 0.2×10^{15} molecules cm^{-2} in the afternoon." in the revised manuscript.

25) P18, L15: before the paragraph ends, I think

Author reply: We moved the whole sentence of "In addition the deviations of the HONO dSCDs derived from the fits with daily noon FRS between the instruments are generally larger than those of the HONO delta SCDs mainly due to the different HONO absorptions in the noon FRS and the interferences by the stratospheric species, e.g. ozone." before the sentence of "Furthermore, there are no considerable systematic differences of the HONO delta SCDs from the fits with the sequential FRS and the daily noon FRS for all the instruments except the mini MAX-DOAS instrument." in the revised manuscript.