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Atmospheric Measurement Techniques Discussions

Interactive comment on "Inter-comparison of MAX-DOAS measurements of tropospheric HONO slant column densities and vertical profiles during the CINDI-2 Campaign" *by* Yang Wang et al.

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

General comments This manuscript evaluates results from intercomparison exercise of HONO observations during CINDI-2 campaign performed at Cabauw, the Netherlands. Previously HONO intercomparison was conducted during MAD-CAT campaign but was limited to dSCD comparisons. Here the exercise, with more participants, was extended to vertical profiling. Tasks were carefully designed, where first the retrievals for HONO and aerosols were conducted with individual groups' own observations/processing for

which protocols were provided, and where the HONO dSCDs, and then the aerosol quantities additionally, were constrained to common values in order to compare the performance of profiling algorithms, separately from the influence from diversity in HONO dSCDs. Basically high degree of agreement was found particularly for selected instruments/groups. The random and systematic discrepancies were evaluated, mainly around the median quantities. With the state-of-the-art instruments, the relative random and systematic discrepancies for dSCDs were about 15 and 30% for low elevation angles, and were about 20% for both for VCDs and near-surface VMRs. The evaluation is important not only for providing adequate a priori errors for the MAX-DOAS retrievals themselves (as the authors mentioned) but also for adequate validation of satellite observations of HONO using MAX-DOAS. Additional findings that atmospheric variability is important for the random term and that basic agreement is reached with active DOAS measurements are also important and interesting. Basically the analytical methods and logics are sound. However, clarification is needed for some important points. Author reply: Many thanks for the positive assessment! We modified the paper based on the comments from you and the other reviewer. Please see the replies and modifications regarding your specific comments below. We give the answers to your individual questions below:

Question 1: "First, I am afraid that the true VCD values (including discrepancies) mentioned in text are 10 times lower (10ËĘ15 must be 10ËĘ14.). The authors need to check all values very carefully as such mistake is fatal." Author reply: the reviewer is right. We are very sorry for the mistakes and thank the reviewer very much for pointing them out. We also checked the values throughout the manuscript and corrected them.

Question 2: "Secondly, the authors need to state why systematic errors quantified to be 30% for dSCDs could diminish to a systematic error of 20% in VCDs." Author reply: Thanks for asking this point. However, the main text in line 474 on page 12 (of the modified manuscript) demonstrate that the systematic error of the dSCDs is typically about 15%, while the random error is typically about 30%. If the reviewer means the

lower random discrepancies of the VCDs rather than the dSCDs, our answer is that the constraint of the profile retrievals (integrated to derive VCDs from the profiles) by the a-priori profile and a-priori uncertainty co-variances (Sa) play the role. In addition, since the dSCDs and VCDs vary in large ranges, and the participating datasets to the comparisons of dSCDs and VCDs are not exactly the same and the data samplings are limited, therefore the relative uncertainties can only be very roughly estimated. In order to clarify this point, we add the following sentence in line 733 to 735 on page 18 (of the modified manuscript): It needs to be clarified that the data samplings are limited in the statistic study, thus the uncertainties of the HONO delta SCDs (and other HONO results including VCDs, near-surface VMRs, and profiles) can only be roughly estimated for typical cases.

Question 3: “Thirdly, the standard deviation is calculated over both days and instruments/retrievals (e.g., Fig. 2 and 6). While the authors conclude the variability derived from instruments/retrievals, day-to-day variability may severely affect.” Author reply: Thanks for mentioning this point. The question is probably due to the unclear description of the calculation procedures of the standard deviations. The calculation procedure is the following. The median values between the instruments/retrievals are calculated for individual time steps. The median values of the SCDs and profiles are shown in Fig. 2a and in the top of Fig. 5. Then the standard deviations are calculated against the median values. Therefore the effect of temporal variations is excluded from the standard deviations. In order to clarify this point, we modified the sentences in line 338 to 341 on page 9 (of the modified manuscript) as follows: “In order to evaluate the agreement of the HONO delta SCDs between the different participants, for the same data sets, the diurnal variation of the standard deviation of all HONO delta SCDs compared to the median values as shown in Fig. 2a is calculated and shown in Fig. 2c. Note that temporal variations of HONO delta SCDs do not impact the standard deviations because the median values in the individual time steps shown in Fig. 2a served as reference in the calculations.” We also added the following clarification in the caption of Fig. 6: “Note that the median values which served as the reference in the calculation of

the boxplots are calculated in the individual time steps, namely each hour. Therefore, temporal variations of the quantities do not contribute to the boxplots.”

Specific Comments: 1. In Abstract, the authors should highlight what was newly done with CINDI-2, beyond MAD-CAT. I believe VCDs and near surface VMRs were intercompared for the first time. Author reply: Thanks for the suggestion. We added the following sentence in the abstract, “The HONO vertical profiles, VCDs, and near-surface volume mixing ratios are compared between different MAX-DOAS instruments and profile inversion algorithms for the first time.”

2. Page 1, line 53, 10ËĘ14 instead of 10ËĘ15? Please check also for Lines 518, 563, 571, 572, 573, 574, 715, 716, 742. Author reply: we are sorry for the mistakes. And many thanks for pointing this out! We modified the numbers and also checked the full manuscript to avoid the mistakes.

3. In Abstract, better to mention that systematic and random discrepancies were determined against median observations basically. Author reply: We added the following sentence to the abstract: “Systematic and random discrepancies of the HONO results are derived from the comparisons of all datasets against their median values.”

4. Line 135. CINDI-2Pr Author reply: The mistake is corrected.

5. Lines 254. Better to tabulate differences among tasks T1a, T1b, T2a and T2b. Aerosols in T2b are same as those in T1a and T2b? Are "aerosol retrievals at 340 nm given in Tirpitz et al. 2020" same as "aerosols retrieved from the O4 delta SCDs" mentioned in line 263? Table 3 mentioned 360 nm. Author reply: The aerosols in T2b are the same as those in T1a and T1b. The input aerosol profiles are the same as those retrieved at 360 nm given in Tirpitz et al. 2020. The “340 nm” is a mistake. We corrected it. The corresponding sentence in line 267 to 269 on page 7 (of the modified manuscript) is modified as follows: “It should be noted that, the input profiles of aerosol extinctions in the tasks T1a, T1b, and T2b are the same and are derived from the aerosol profile retrievals at 360 nm, as given in Tirpitz et al., 2020, using the common

settings by the individual participants."

6. Lines 346-349. Is this statement valid when variability is studied including days with different concentrations? Author reply: Following our reply to the question 3 in the main comment, the effect of the temporal variations of the HONO concentrations is excluded from the calculations of the standard deviations. Therefore the statement is valid.

7. Line 369. How less were the photons could be quantitatively discussed. Author reply: We quantified the reduction of the photons by citing Wagner et al (2014) in the modified manuscript. The following sentence is added in line 374 to 375 on page 9 (of the modified manuscript): "Wagner et al. 2014 demonstrated that the number of photons (proportional to the measured radiance) is typically reduced by more than 10% under optically thick clouds compared to those under clear sky conditions."

8. Line 407. Not only "codes" but instrumental characteristics, such as how well the slitfunction is represented during DOAS fit might affect? Author reply: Yes, we agree. Therefore we added the possible reason in line 409 to 411 on page 10 (of the modified manuscript): "This finding implies that random discrepancies between the data sets can be considerably attributed to the specific implementation of the DOAS fits and the characteristics of the instrumental slit functions by the individual participants."

9. Line 413. Define what is "mini". This statement should only be valid for Cabauw or cleaner sites. Author reply: We modified the sentences to clearly define the "mini" MAX-DOAS, and specify the conditions of HONO concentrations which are lower than the typical signal to noise ratios of the "mini" MAX-DOAS. The modified sentence on line 418 to 418 on page 10 (of the modified manuscript) is as follows: The "CMA" RMS values derived for a Hoffmann Mini-DOAS instruments are the largest ($\sim$1 to 1.7$\times$1015 molecules cm-2, corresponding to a "typical percentage" of 30% to 85%). The large RMS of "CMA" is consistent with its large fit error of $\sim$1$\times$1015 molecules cm-2. Therefore, we conclude that the Hoffmann Mini-DOAS instruments can hardly reach the signal to noise requirements for HONO measurements in cases of HONO

dSCDs lower than $\sim$2$\times$1015 molecules cm-2.

10. Line 416. Comma should be period Author reply: We added a comma before the "corresponding to".

11. Line 448. Did the difference occur on selected days? Or for most of the days of observation? Author reply: Since the phenomenon is found from the statistic comparisons of the measurements during the whole campaign, therefore the finding should be a general feature. The feature is clarified by adding "usually" in the sentence.

12. Line 467. I believe the quantities are not "in general" but for selected high performance instruments. Author reply: The quantities are for most of the instruments during this campaign with moderate performance. Therefore we clarified the point in line 472 on page 12 (of the modified manuscript) as follows: In general, for most of the instruments with moderate performance during this campaign,...

13. Line 471. Why DOAS fit error is discussed within systematic term here, while it was discussed under random term before? Author reply: We checked the manuscript, but "DOAS fit error" is not discussed in the line.

14. Section 3.3.3. Influence from different FOV angles for individual instruments can be ignored? Author reply: Since the typical effect of the FOV on the HONO dSCD is not considerably larger than the noise level of the HONO dSCDs, the effect can not be seen during this campaign. It could become important when the signal to noise ratios of MAX-DOAS instruments are significantly improved.

15. Line 531. The better result in T2a (compared to T1a) might be partly from the fact that only high-performers took part in the T2a exercise? Author reply: The performance in T1a is mainly dependent on the different instruments. Since only one instrument is used in the T2a, the instrumental performance in T1a will not impact the discussion in T2a. Differences of the results in T2a are mainly due to the different profile inversion algorithms, which are almost same as those used in T1a. The M3 (LMU) is only used

in T1a, but not in T2a. However, the algorithm performs well in T1a. Therefore we conclude that the better results in T2a compared to T1a are mainly attributed to the fact that the same set of HONO dSCDs is used in T2a by all the participants.

16. Line 561. 0 to 0.2 km, 0.2 to 0.4 km, and 0.4 to 0.6 km (4 km in the figure; be consistent) Author reply: Thanks for pointing out this mistake. It has been corrected in the modified manuscript. Printer-friendly version Discussion paper 17. Line 566. What are the "modelled" quantities? Fitted dSCDs for some elevation angle? Readers may even wonder if they are from chemical transport models. Author reply: In order to clarify the “modelled HONO delta SCDs”, we added a sentence in line 570 to 572 on page 14 (of the modified manuscript) as follows: It needs to be noted that the modelled HONO delta SCDs represent the HONO delta SCDs which are simulated by a RTM, which is included in individual profile inversion algorithms.

18. Line 603. I did not clearly see the systematically low values at high altitudes from LMU in Fig. 8. Author reply: The HONO VMRs in the height interval of 0.4 to 4km have a slightly negative bias, which is bigger than others in Fig. 8. Although it is not a significant bias, its effect on the VCD can still be considerable since the height interval range is 3.6 km. We modified the sentence by replacing “systematically” by “slightly” to describe it more appropriately.

19. Line 617. Was any similarity found for NO2, a potential precursor of HONO? Author reply: The comparisons of NO2 near-surface concentrations between MAX-DOAS measurements and LP-DOAS given in Tirpitz, et al., 2020 do not indicate a systematic underestimation of MAX-DOAS. However we see an underestimation for HONO in our study. The reason might be attributed to the much shorter lifetime of HONO than NO2. The information is added in line 627 to 629 on page 16 (of the modified manuscript) as follows: However, the comparisons of NO2 near-surface concentrations between MAX-DOAS measurements and LP-DOAS, as given in Tirpitz, et al., 2020, do not indicate a systematic underestimation of MAX-DOAS. The different feature for NO2 and HONO might be attributed to the much shorter lifetime of HONO than NO2.

20. Line 618, 625, lifetime Author reply: We corrected it accordingly.

21. Section 5.1 Was constant Sa used for the general comparison exercise previously described? Any feedback comment from this exercise? Author reply: The constant Sa is not used in the general comparisons. The special test indicates that a Sa adjustment is also important in the inversion algorithms based on optimal estimation.

22. Section 5.1. Need to discuss negative values found with profile 1 for AUTH and INTA. Author reply: The negative values are allowed to be derived using the “BePro”, although they are unrealistic in the real atmosphere. Differently, negative values are avoided in the “PriAM” algorithm due to the logarithmic transformation. The effect is stronger only for profile 1, mainly because the true profile 1 is far away from the a-priori 2 and 3. In order to clarify the point, we added the following sentence in line 663 to 665 on page 16 (of the modified manuscript): In addition, it needs to be clarified that negative values are allowed to be derived using “BePro”, although they are unrealistic in the real atmosphere. In contrast, negative values are avoided in the “PriAM” algorithm due to the logarithmic transformation.

23. Line 721. Define "good" spectrometer. Author reply: we modified the description to: For MAX-DOAS instruments with moderate performance during this campaign.

24. Line 767. Are the uncertainties for dSCDs? Author reply: Yes, the uncertainties are for HONO dSCDs. We modified the sentence accordingly in the modified manuscript.

25. Figure 3. Scatterplots should be presented (maybe in supplementary) to show to how low dSCDs agreement is found. Author reply: Many thanks for the suggestion! However, we think the scatterplots will not help the discussions in the manuscript, because the parameters derived from the scatterplots are already quite many and well demonstrate systematic and random discrepancies.

26. Figure 4, Put labels (a), (b) and (c). What are the "subplots" mentioned in the caption? Author reply: We added the labels in the caption.

27. Figure 5. The scale only starts from zero and thus negative values are not shown. AUTH sometimes went to negative range here, as shown in Figure 9? Author reply: Yes. However, the negative values are quite small, not as large as those shown in Fig. 9 for profile 1 with a priori 2 and 3. Since the a-priori 1, which is closer to the typical HONO profile shape, is used in the common setting of the profile retrievals, the negative values at high altitudes are quite small. We added the following clarification in the caption of Fig. 5: Note that the colormap starts from zero. Negative values can appear in some datasets, but are generally insignificant since their mean values are about -0.007 ppb.

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