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Atmospheric Measurement Techniques Discussions

Interactive comment on "Eight-component retrievals from ground-based MAX-DOAS observations" *by* H. Irie et al.

H. Irie et al.

irie@jamstec.go.jp

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Reply to anonymous referee 2

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. Detailed responses to the comments are given below.

General comments:

Comment 1: HONO is a strong absorber ($\sigma \approx 5x10^{19}$ cm2) in the near uv-range. In addition, HONO can reach 10% of the NO2 at ground level. Therefore, it might influence the absorption spectra. Please comment on additional absorbers which are not considered here.

Reply: Following the published literature focusing on HCHO retrievals by DOAS (e.g., De Smedt et al., Atmos. Chem. Phys., 2008), HONO was not included in our DOAS analysis. However, we made additional DOAS analysis with HONO for both 336-359 nm (fitting window for HCHO) and 338-370 nm (for UV O4). By including HONO, its impact on O4 DSCDs was estimated to be only 0.2%, whereas HCHO DSCDs decreased by 9% on average for the entire observation period. However, when HONO was included, 43% of the HONO DSCD data showed negative values, potentially indicating that it could interfere with HCHO directly and/or indirectly through O3. Therefore, we have not included HONO in the present study. This is now stated in the revised manuscript.

Comment 2: The authors use 8 different spectral windows for their evaluation (Table 1). It is not clear to me why HCHO and AEC357 need different fitting windows. I also do not understand why NO2 was not deduced from the strongest differential absorptions around 435 nm.

Reply: Our first thought was the same as the reviewer's, but the choice of the fitting window was critical in spectral analysis, especially for HCHO. The fitting window used for HCHO is 336-359 nm, which does not fully cover the O4 absorption band around 360 nm. We realize that the best fitting window for NO2 is around 435 nm, in the case that spectral fitting targets only NO2. Our MAX-DOAS retrieval, however, targets both NO2 and O4. The AMF at 476 nm, derived from O4, was used for our NO2 profile inversion assuming that the wavelength for the NO2 AMF is 476 nm, although AMF varies with wavelength. To minimize the error due to this assumption, we have used a single fitting window, from which both NO2 and O4 DSCD values can be retrieved, where the NO2 absorption at 460-490 nm was sufficiently high to see temporal variation in NO2 at Cabauw, as shown in Fig. 5.

Comment 3: In section 3.2.1 the aerosol extinction retrieval is described in detail. The authors use the OEM to retrieve the aerosol profile in the same way as in Irie et al. (2008). However, from the present text it is not fully clear why additional Monte Carlo

calculation to create a lookup table are done. As far as I understand the LUT is used to calculate the F(xi) and the Ki as input for the OEM. Other studies (e.g Friess et al. 2006) use Optimal Estimation (with internal forward model) only, while Wagner et al. (2004, 2007) and Pikelnaya et al. (2007) and Li et al. (2010) use Monte Carlo radative transfer model to compare/fit the DSCDs. Instead of repeating part of Irie et al. (2008) it might be helpful to comment on the pro and cons of the method here compared to other approaches.

Reply: As the reviewer suggests, a lookup table (LUT) of the box-air-mass-factor (Abox) vertical profile is used to calculate O4 DSCDs in the forward model. The O4 DSCDs are then compared to those derived from the DOAS method, similarly to approaches presented in most cases in the literature. The reason why we made Monte Carlo calculations in addition to the work of Irie et al. (2008) is that for each measurement site our calculation takes into account the surface altitude at the measurement site and the altitude where the instrument was located. Accordingly, section 3.2.1 of the revised manuscript now states "A lookup table (LUT) of the box-air-mass-factor (Abox) vertical profile, which was used to calculate O4 DSCD from given aerosol profiles and observation geometries in the forward model, was created using ..." and "To simulate a realistic atmosphere, we considered the surface altitude at the measurement site and the altitude where the instrument was located."

Comment 4: In a paper by Zieger et al. (2011) on the same campaign the MAX-DOAS results do not well match the locally measured aerosol data. It might be useful here to make a not how this might affect the retrieval of the trace gases.

Reply: Following the reviewer, the revised manuscript now states that "The MAX-DOAS AEC retrieval is expected to be improved if such spatial inhomogeneity of the aerosol distributions is considered" in section 5.

Comment 5: The linear regression procedures are not explained. However, when dealing with data having statistical errors in both coordinates methods like fitexy (c.f. book

by Press et al. 1993, Numerical recipes) should be considered. They also provide meaningful measures for the goodness of the fit and the errors of the deduced parameters. This method fitexy was widely used in intercomparisons of instruments.

Reply: We appreciate this suggestion. In the revised manuscript, we now use the linear least-squares fit taking into account error ranges. This is stated in the corresponding figure captions.

Comment 6: Section 3.2.2, Page 652: The ∆Abox depends not only on the AOD but also on the absorption cross section of the considered absorber (Marquard et al. 2000, Rozanov and Rozanov, 2010). So, it is hard to understand how to re-calculate the ∆Abox LUT (in which the absorber is a specific trace gas, e.g. NO2) based on an existing ∆Abox LUT (in which the aborber is O4) which was created at different wavelength. In priciple, when switching from one absorber or wavelength to another, an accurate AMF can only be achieved by a new RTM calculation. Moreover, the LUT setup for the trace gas retrieval is not so clearly described. Please make a note how the AMF LUT during the trace gases retrieval was created and about its wavelength and cross section dependence.

Reply: As pointed out by the reviewer, we realize that the ∆Abox values can differ for different species, as the ∆Abox values depend on the concentration profile and wavelength. On the other hand, Wagner et al. (2007) have argued in their RTM intercomparison paper (p. 1811) that "The great advantage of calculating box-AMFs is that they can serve as a universal data base to calculate appropriate (total) AMFs for arbitrary species with different height profiles." This argument should be valid for optically thin absorbers (optical depth « 1) at the same wavelength. Strictly speaking, ∆Abox values can differ for different species analyzed in the present study, as the reviewer suggests, but the dependence of ∆Abox values on the concentration profile should be very small, as the optical depth « 1. Therefore, we have omitted this dependence. In contrast, the wavelength dependence is much more important. To take the wavelength dependence into account, ∆Abox values were re-calculated from existing LUTs us-

ing an AOD modified considering its wavelength dependence. The revised manuscript now states that "Then, the ΔAbox profiles that correspond to the converted AOD (but at the same wavelength as the aerosol retrieval) are re-calculated from the ΔAbox LUT prepared for aerosol retrievals and used for trace gas retrievals. The dependence of ΔAbox on the concentration profile of trace gases has been omitted, since it should be very small as they are optically thin absorbers (the optical depth « 1) (Wagner et al., 2007)."

Comment 7: A weak part of the manuscript is the discussion on the HCHO and CHOCHO. In my opinion, an interpretation of the ratios of HCHO to CHOCHO requires more information (VOC levels, local sources, etc.) which is not in the focus of this manuscript. I suggest to focus on the the comparison of MAX-DOAS retrieved data with the locally measured data and, if measured data are not available, with model results. An interpretation in terms of chemistry (ratio of HCHO to CHOCHO) is outside the scope here.

Reply: We agree with the reviewer. Since no proper independent measurements were available, we needed to rely on the CHIMERE model. However, while CHIMERE CHOCHO values seemed too low, the ratio of HCHO to CHOCHO was used to argue that the concentration level of retrieved CHOCHO was not far from reality, compared to CHIMERE data. We also think that detailed discussion of the chemistry should not be made here. The revised manuscript now states that "To confirm this, the quantitative validation of MAX-DOAS retrievals using accurate independent observations of CHOCHO is desirable."

Technical corrections:

Comment 8: Page 644: Please cite Platt and Stutz (2008) instead of Platt (1994).

Reply: Done.

Comment 9: Section "Conclusion" should be renamed to "Summary and conclusion"

Reply: Done.

Comment 10: Figure 1 is a bit confusing since the different wavelength ranges are mixed around the 8 panels. I suggest to have one wavelength scale and the different spectra stacked with fitting ranges (see also note on Table 1) marked.

Reply: We tried to revise Fig. 1 following the reviewer's suggestion, but plots for some species, whose fitting window was relatively short, had been shrunk. The revised manuscript now uses the same figure as the original, but we have added the statement "Note that different wavelength ranges are used for the 8 panels" in the figure caption.

Comment 11: Figure 2: Why using K instread of AEC in the rest of the manuscript?

Reply: The AEC is now used in Fig. 2 of the revised manuscript.

Comment 12: Figure 5: Very dense figure. Please make a note in the caption why the CHIMERE results are scaled.

Reply: Although it might be hard to see some details, we believe that showing 8 different time series in one figure is convenient for readers to see the overall behavior. In the figure caption, we now mention "CHIMERE NO2, HCHO, and CHOCHO are scaled by factors of 0.6, 2.5, and 4.5, respectively, to improve agreement (see the text for more details)."

Comment 13: Figure 6: Are error bars for CIMEL available?

Reply: Error bars for CIMEL have been omitted for clarity, as the error was generally as small as $\sim$0.01.

Comment 14: Figure 9/10: Are these regression lines meaningful? (see also note on regressions above)

Reply: We believe that correlation coefficient R given in each plot indicates how meaningful the regression lines are. We have added the sentence "The correlation coefficient R is given in the plot" in the figure captions.

Interactive comment on Atmos. Meas. Tech. Discuss., 4, 639, 2011.