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Atmospheric Chemistry and Physics Discussions

Interactive comment on "MAX-DOAS detection of glyoxal during ICARTT 2004" *by* R. Sinreich et al.

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Interactive comment on "MAX-DOAS detection of glyoxal during ICARTT 2004" by R. Sinreich et al. F. Wittrock (Referee) mail@folkard.de Received and published: 17 November 2006 The paper by Sinreich et al. reports on observations of glyoxal (CHO-CHO) using ground- and shipbased MAX-DOAS measurements during the ICARTT campaign in 2004. The measurements from the ground were carried out at MIT, Cambridge, USA, those from the ship on board the research vessel Ron Brown in the Gulf of Maine. The study focuses on a limited data set: One day in July 2004 for each instrumental platform. However, since only a very few measurements of glyoxal in the atmosphere have been published so far it contains significant and new information on this trace gas. The paper is in general clearly written and I recommend it for publication in ACP. But the authors should address for some revisions/corrections as detailed below, most of them are technical/editorial.

Author reply: We want to thank Folkard Wittrock for the positive assessment and the

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helpful comments. We mostly followed them (and explained the reasons in cases we did not follow them), as outlined in detail below.

General comments:

- There is only one major point the authors should address in their revised version. Since this paper is on the theoretical aspects of the MAX-DOAS detection of glyoxal more information and in particular more figures are necessary to illustrate the profile inversion in section 4. Similar information is given in Sinreich et al., 2005. However, the reader will have a more thorough overview on this important topic, which makes the section 5 (Results) more useful. I propose to add following: One example showing modeled and measured O4-DSCDs for one day., e.g. for the hourly averages shown in figure 3. Do the authors have taken into account all elevation angles of the MAX-DOAS to analyse the aerosol? Please add a statement on the possible influence of the aerosol type on the retrieval. This might be also one reasonable explanation for problems in aerosol modeling for measurements on board RV Ron Brown on July 17th 2004. The explanation given in the manuscript " . . . due to the relatively high AOD" is not very conclusive since an AOD of 0.2 is quite common above the oceans.

Author reply: We included an example of the O4 retrieval in section 5. Also, we modified the text in section 4 to clarify that all elevation angles were used (“ĚThey were compared to the measured O4 AMFs of the different elevation angles, which were obtained by dividing the O4 SCDs by a typical O4 VCD. Ě”). We added the following discussion of the aerosol type: “This method does not primarily focus on the inversion of the specific properties of the aerosol which due to the small wavelength range is not impossible but at least difficult to retrieve. Our approach aims on the modeling of the optical properties of the aerosol which fit to the O4 measurements and apply for the inversion of the trace gases. Thereby, ambiguities of the specific aerosol properties can occur.” Concerning the AOD, of course, in reality it could be much higher. What we wanted to express was that our method has not yielded an unequivocal result due to the high AOD. However, we added that the aerosol profile might be very complex as

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well.

- The authors have used the ratio of 3° and 18° DSCDs to estimate the mixing layer height for CHOCHO. This is of course a suggestive approach but the authors should point out more clearly that this is based on the assumption that all of the CHOCHO is well-mixed within one single layer which is probably not the case in reality (see e.g. Heckel et al., 2005). In fact, the same argument is true for the aerosol profile (see comment on figure caption 2 below). The authors might add a statement that a future retrieval algorithm will profit from the measurements at all different elevation angles and therefore provide more profile information.

Author reply: We agree with the referee and added the following paragraph: "Also, the assumption of well-mixed aerosol and trace gas layers is a rough, but not necessarily a totally unrealistic estimate. In future work, more detailed trace gas profiles could be retrieved, if the information of all elevation angles is used in the trace gas profile retrieval, which was already done by Heckel et al. (2005)."

- One obvious question by reading this paper is: What is the reason that no measurements of HCHO are presented? The retrieval of O4 in the UV already includes formaldehyde and this trace gas would provide valuable information on the interpretation of the glyoxal observations.

Author reply: In this paper we focus on the methodological aspects of the CHOCHO retrieval. The interpretation is supposed to be mainly subject of a companion paper. Additional data of HCHO would inevitably lead to a further discussion with a more meteorological and chemical focus, which is beyond this paper.

- It would be worthwhile to add information on the vertical columns of CHOCHO during the ICARTT campaign. If possible for more than one day since this pulls the manuscript together with the satellite work.

Author reply: In Fig. 4, we added a plot of the VCD of CHOCHO. More data would be

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beyond this methodological paper. (see comment above)

Minor corrections/comments: - Abstract, line 6: The authors should take into account, that in Wittrock et al. not only satellite measurements are shown but also MAX-DOAS measurements have been used for validation. Therefore the statement “we report the first detection of CHOCHO by . . . ” should be rephrased slightly. The same is valid for page 9461, line 15 (Introduction).

Author reply: We substituted “comprehensive analysis” for “detection”. Additionally, in the introduction the text now reads “In this study we present the first comprehensive analysis of CHOCHO using passive ground-based DOAS instrumentation based on initial studies by Sinreich et al. (2004). CHOCHO observations from ground-based instruments have also been reported by Wittrock et al. (2006).”.

- Abstract, line 16: “The paper focuses on the instrumental aspects . . . ”. If this is true, I wouldn’t recommend this study for publication. There is no special instrumental requirement necessary to observe glyoxal with the MAX-DOAS technique. Please skip this sentence or rephrase it.

Author reply: We replaced instrumental by methodological. Many thanks for this hint!

- Sometimes the authors did not introduce all acronyms before they are used the first time (e.g. MIT, MCMA).

Author reply: We added indroductions of the acronyms MIT, MCMA, FWHM, OMI, SCIAMACHY and ICARTT.

- I agree with reviewer Thomas Kurosu that a lot of references should be deleted, e.g. most PhD and Diploma theses. If no other peer-reviewed reference is available on a certain topic one might add a link with access to any time e.g. http://deposit.ddb.de/cgi-bin/dokserv?idn=962363928 (Dissertation Udo Frieß).

Author reply: We deleted all Diploma and PhD theses except for the dissertation of Udo Frieß where we included the internet link as suggested.

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- Introduction, page 9462, line 7 - 12, please add Wittrock et al., 2004 and Heckel et al., 2005 since these publications illustrate the retrieval of trace gas profiles from MAX-DOAS observations.

Author reply: We added these references.

- Introduction, page 9462, line 14 - 26: The primary quantities for ground-based DOAS measurements are differences of slant column densities (DSCD)! They have to be converted to vertical columns (VC) by the means of differences of air mass factors (DAMF). Please rephrase the paragraph making this more clear. The same is valid for the description of “The DOAS analysis” and some other text passages.

Author reply: We altered this throughout the article as suggested.

- Introduction, page 9462, line 29: Information on measurement sites not used in this study is not very meaningful.

Author reply: We deleted the information on the other measurement sites.

- DOAS analysis, page 9465, line 11: This is definitely true for NO2 but in case of CHOCHO stratospheric contributions are unlikely.

Author reply: We added “Ĕ (of course for CHOCHO no stratospheric contribution is expected)”

- Page 9466, line 16: Why O4 was not fitted including the absorption band at 380 nm? A fitting window more to the visible should give a better sensitivity to the aerosol and reduces possible interference with HCHO, BrO and ozone.

Author reply: The absorption band at 360 nm is almost two times stronger than at 380 nm, thus the signal-to-noise ratio is much better. Furthermore, a comparison of the retrievals of both absorption bands yielded the same result within the uncertainty of the measurement. Due to the different band width of O4 compared to ozone, HCHO and BrO we can neglect a possible interference (based on our experience). From all

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these points, we conclude that the O4 absorption at 360 nm is indeed a very well suited choice.

- Figure 2: Again, no SCDs but DSCDs are shown here. Please add information on the solar zenith angles during the day. The variation of the O4 on July 17 (Ron Brown) with sometimes smaller values in the 3° direction than in the 6° and 10° indicates a more complex aerosol profile than assumed in the retrieval. What is the reason for the very high NO2 for large SZA (in particular in 18° direction) on board RV Ron Brown? Stratospheric contribution since the zenith sky measurement is not close enough to the off-axis?

Author reply: We changed SCD in DSCD and added a plot which gives information on the SZA. Thereby, we recognized that, unfortunately, we had a time shift of one hour in our original manuscript (the shown DSCDs were measured one hour earlier than denoted) in the values of MIT. We apologize for this mistake. We performed the inverse modeling again, and have updated all plots accordingly. Fortunately, these new calculations did not affect the conclusions of the article, but nevertheless some small changes in the text had to be done concerning the time of the CHOCHO-to NO2 peak (changed from 1:30 PM to 12:30 PM), the mixing ratio peak (from 120 to 140 ppt) and the AOD (now shows a peak around noon). We want to thank Folkard Wittrock for this very important hint. Concerning the complexity of the aerosol profile we agree with the referee. The comparison between the modeled and measured O4 values didn't yield an unequivocal aerosol profile height (in contrast to the total vertical thickness). This indicates - as suggested by the referee - that the aerosol profile is possibly more complex than we assumed in our retrieval. We added: Ď...did not lead to an unequivocal result due to the relatively high AOD (of course, in reality even much higher AODs can occur. Nevertheless, the sensitivity of the O4 absorption on the AOD decreases with increasing AOD) and possibly the complexity of the real aerosol scenario." Thank you for the hint about the high 18° DSCDs in the morning. Above 90° SZA, our measurement routine only pointed into 18° elevation angle. Thus, these

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spectra were evaluated by the same reference which led to a stratospheric contribution. We deleted these values. In the evening, there is no indication for an artifact with the retrieval.

- Figure 3 and 4: It might be useful to combine these figures and add information on the vertical column of CHOCHO (see comment above) and the solar zenith angle.

Author reply: We added a plot of the VCDs for each site in Fig. 5 (formerly Fig. 4).

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 9459, 2006.