

Interactive comment on “Aircraft measurements of bromine monoxide, iodine monoxide, and glyoxal profiles in the tropics: comparison with ship-based and in situ measurements” by R. Volkamer et al.

Anonymous Referee #1

Received and published: 26 February 2015

General comments

The manuscript entitled 'Aircraft measurements of bromine monoxide, iodine monoxide, and glyoxal profiles in the tropics: comparison with ship-based and in situ measurements' describes the validation of trace gas vertical profiles from MAX-DOAS with independent observations during two flights as part of the TORERO campaign. Vertical profiles of aerosols, water vapour, glyoxal, NO₂, BrO and IO are compared to ship-borne MAX-DOAS measurements, in situ water vapour and aerosol size distribu-

C199

tion observations, as well as profiles from a chemistry-transport model.

The inversion of tropospheric trace gas and aerosol profiles from ground-based, ship-borne and air-borne MAX-DOAS measurements is a strongly emerging field and a validation of this measurement technique is crucial for an assessment of their overall quality. Therefore, the topic of the manuscript fits well within the scope of AMT.

The level of agreement of the AMAX-DOAS measurements with independent observations and model results presented by Volkamer et al. is very impressive. However, I feel that the manuscript requires some modification prior to a publication in AMT. As detailed below, many open questions remain regarding the methodology for the retrieval of vertical profiles. Furthermore, a comparison of the results obtained within this study with validation results from previous airborne campaigns, which would allow for an assessment of the capabilities of MAX-DOAS compared to other measurement techniques, is missing.

I suggest to modify the title of the manuscript. Currently only measurements of BrO, IO and glyoxal profiles are mentioned in the title, but also results for NO₂, H₂O and aerosols are presented.

Specific comments

In contrast to other MAX-DOAS retrieval algorithms (e.g., Friess et al., 2006; Clemer et al., 2010), the method described here relies on absolute SCDs instead of differential dSCDs. I have to admit that I have difficulties to understand this approach and its implications. Inverse methods should be based on a forward model that accurately describes the measurement process. In case of MAX-DOAS, the primary quantity derived from a spectral analysis is the dSCD, and this is thus the quantity that should be modelled by the retrieval algorithm. An explanation why absolute instead of differential SCDs serve as input for the retrieval is missing. What is the benefit of this approach?

An important implication of the use of absolute instead of differential SCDs is the ne-

C200

cessity to accurately derive the reference amount SCD_{ref} . Little information is provided on how exactly the values for SCD_{ref} were derived. What do you mean with 'The SCD_{ref} was determined explicitly through RTM calculations' (638.11)? Why is SCD_{ref} not determined (explicitly or implicitly) by the retrieval algorithm? From a perspective of information theory, I doubt that the information content of the retrieved atmospheric state can be improved if SCD_{ref} is estimated externally from the measurements themselves.

For the same reasons, I have difficulties to understand the discussion in Section 3.4 on the impact of SCD_{ref} on the retrieved profiles. If, as the authors state, the retrieval of profiles for the SMAX measurements uses dSCDs as measurement vector, then the forward model needs to simulate $dSCD = SCD - SCD_{ref}$, i.e. the difference between the SCD simulated for the measurement geometry and the SCD simulated for the reference geometry. This is not equal to the assumption $SCD_{ref} = 0$. The retrieval algorithm should determine SCD_{ref} implicitly and there is no need to prescribe SCD_{ref} externally.

Based on radiative transfer calculations and speculations on the vertical distribution of trace gases and aerosols, Section 2.5 contains an extensive discussion on the choice of the reference spectrum. Why has this complicated approach been chosen, if there is a much simpler method to determine the spectrum with the smallest SCD, namely to analyse the dataset using an arbitrary reference spectrum, and to choose the spectrum with the smallest dSCD (plus other criteria, such as the intensity level) as reference in a final analysis run?

I suppose AMAX-DOAS measurements during ascend and/or descend were used for the retrieval, but this information is not provided explicitly. How many measurements were usually performed per altitude unit?

The agreement between the aerosol extinction profile retrieved from AMAX-DOAS O_4 and modelled based on measured aerosol size distribution and Mie calculations is very

C201

impressive. In particular, it seems to be possible to accurately determine the aerosol extinction in the upper troposphere, where Rayleigh extinction is about an order of magnitude higher. It would be important to discuss how the results of TORERO compare to closure studies from other airborne campaigns (e.g., Collins et al., In situ aerosol-size distributions and clear-column radiative closure during ACE-2, Tellus B, , 52, 498-525, 2000), and what has been done to achieve this high level of agreement between in situ and remote sensing aerosol measurements. A description of the measurement principle of the UHSAS (is it measuring dried aerosol or at ambient humidity, which particle size range is it covering), and its uncertainties is missing.

The agreement of NO_2 and H_2O profiles from AMAX-DOAS with model simulations and in situ measurements is better than anything I've seen before (see, e.g., Brunner et al., An evaluation of the performance of chemistry transport models by comparison with research aircraft observations. Part 2: Detailed comparison with two selected campaigns, ACP, 2003). In particular, most CTMs strongly overestimate NO_x in the tropical troposphere. I would therefore appreciate if more details on the chemical and dynamical scheme of the RAQMS model could be provided and if the results obtained during TORERO would be compared to previously published comparisons between airborne trace gas measurements and model simulations.

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 623, 2015.

C202