Multi-axis DOAS measurements during FORMAT campaign - First results in retrieving Formaldehyde

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A. Heckel, T. Tarsu, A. Richter, F. Wittrock, J. P. Burrows, W. Junkermann[1], C. Hak[2]

Institute of Environmental Physics, University of Bremen, P. O. Box 330440, D-28334 Bremen, Germany
akheckel@iup.physik.uni-bremen.de

[1] IMK-IFU, Kreuzeckbahn 19, D-82467 Garmisch-Partenkirchen, Germany
[2] Institute of Environmental Physics, University of Heidelberg, Im Neuenheimer Feld 229, D-69120 Heidelberg, Germany

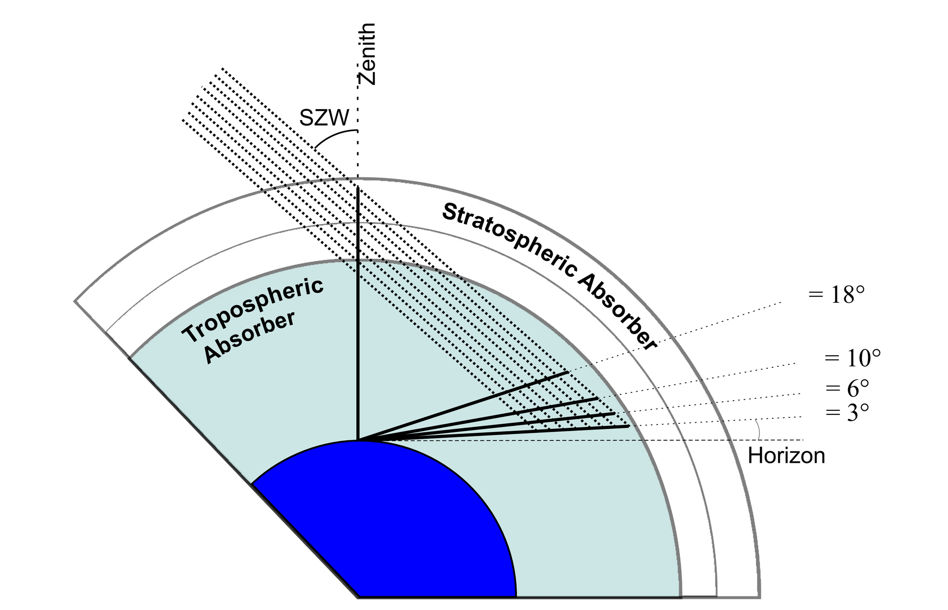
Introduction

Measuring scattered sunlight not only from zenith, but also from lines of sight with low elevation angles provides the possibility to derive vertical columns and profile information, especially for low altitudes, such as boundary layer and lower troposphere. With our MAXDOAS instrument developed by S. Fietkau, T. Medeke and F. Wittrock, it is possible to take those measurements automatically for 5 different viewing directions (zenith + 4 x off-axis). Such an instrument was used during the Format campaign in summer 2002. The DOAS analysis gives the opportunity to detect serveral atmospheric trace species, such as O_3, NO_2, HCHO and BrO, which play an important role in the atmospheric chemistry. The knowledge of the concentrations of these trace gases is necessary for the attempts to understand effects like photosmog, development of tropospheric ozon or global warming.

FORMAT

The acronym FORMAT stands for "Formaldehyde as a tracer of photooxidation in the troposphere". The project, funded by the EU, is an attempt to get a better understanding of the chemical processes in the lower troposphere, especially in the boundary layer, during photosmog episodes. Formaldehyde is therefore a good indicator for photochemical smog.
The most important aim of the project is to introduce new and to improve existing measurement techniques, in order to get accurate measurment results. Enlarging the data base it will be possible to improve the knowledge about the development of those smog events yielding better predictions to warn authorities and the public.

Experimental Setup



Scheme of the different viewing geometries used to derive column amounts and mixing ratios of tropospheric trace species

- telescope measuring five consecutive viewing directions (zenith, 3°, 6°, 10° and 18° above horizon) by use of a computer controlled turnable mirror
- a depolarizing quartz fiber bundle is connecting the telescope to the spectrometer.
- spectrometer: Acton SpectraPro-500, focal length 500 mm using a grating with 600 lines/mm
- detector: Princeton Instruments CCD with 1100x330 Pixel UV enhanced
- wavelength range: 312 - 396 nm

Analysis Method

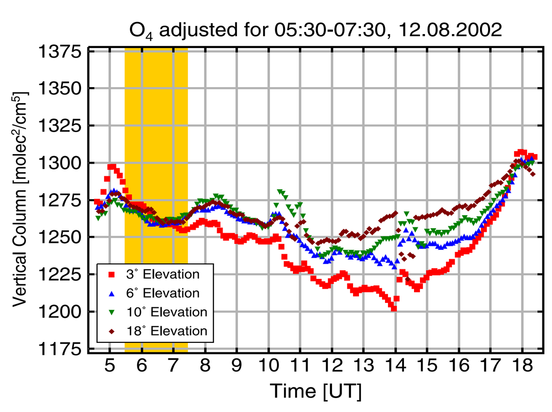
Measuring with different viewing geometries provides the possibility to derive profile information of the trace gas of interest itself as well as aerosol profile information.
Simulating the radiative transport with Sciatran [1] yields air mass factors (AMF), which describe the actual atmospheric situation. In general changes in the atmosphere lead to changes in the AMFs, resulting in serveral different sets of AMFs per day. Since the simulation of cloudy situations is much more difficult because of fast variations and a larger set of initial parameters, the current analysis is applied only for clear sky situations.
Using the equation

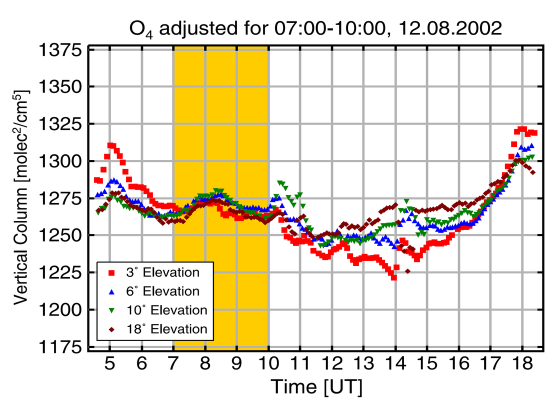
$$VC = \frac{SC}{AMF}$$

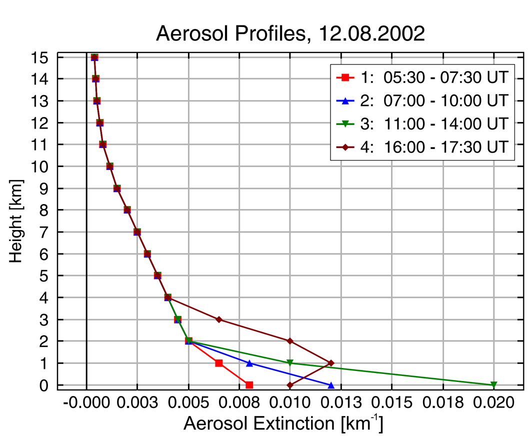
it is possible to convert measured slant columns (SC) to vertical columns (VC) by use of the calculated AMFs. This procedure leads to five independent derived vertical columns and they are equal if the AMFs describe the true atmosperic situation most realistic.
Formaldehyde is retrieved by using the fitting window 335-357nm. For the retrieval of O4 a fitting window of 350-370nm is used.
For simplification of the radiative transport modelling the days August 12th and 13th 2002 are analysed. These days had very good visibility of about 150km and only little aerosol extinction, but also little Formaldehyde compared with other days during the campaign.

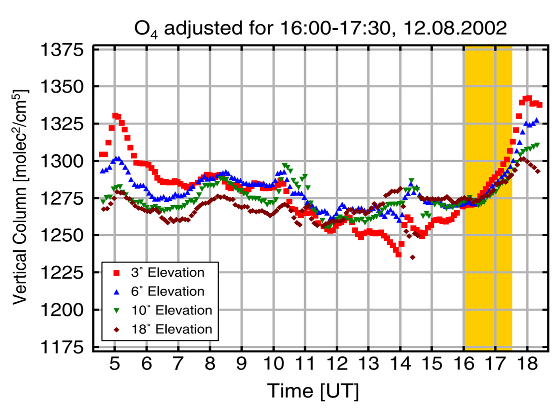
Aerosol Retrieval

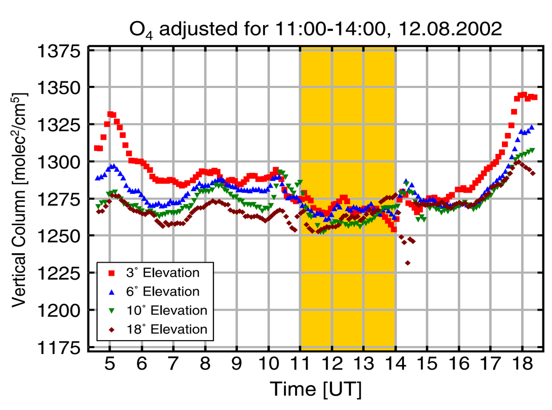
O_4 is a powerful indicator for the radiative conditions in the atmosphere. It's vertical profile is mainly determined by pressure and temperature and therefore can be computed. Hence O_4 is useful to derive parameters like the aerosol profile or albedo.
The plots show the O_4 total vertical columns for the four different off-axis directions. By adjusting the aerosol profile it is possible to match these columns for certain time periods yielding the corresponding diurnal variation of the aerosol profile. As shown the aerosol extinction is increasing during the morning and lifted in the afternoon, which is reasonable assuming a convective boundary layer.
Please note that around 14:00 - 15:00 UT there was a disturbance possibly due to little cloud cover. Therefore an interval between 14:00 - 16:00 UT is excluded from the analysis of O_4 as well as HCHO (see section on the right hand side).





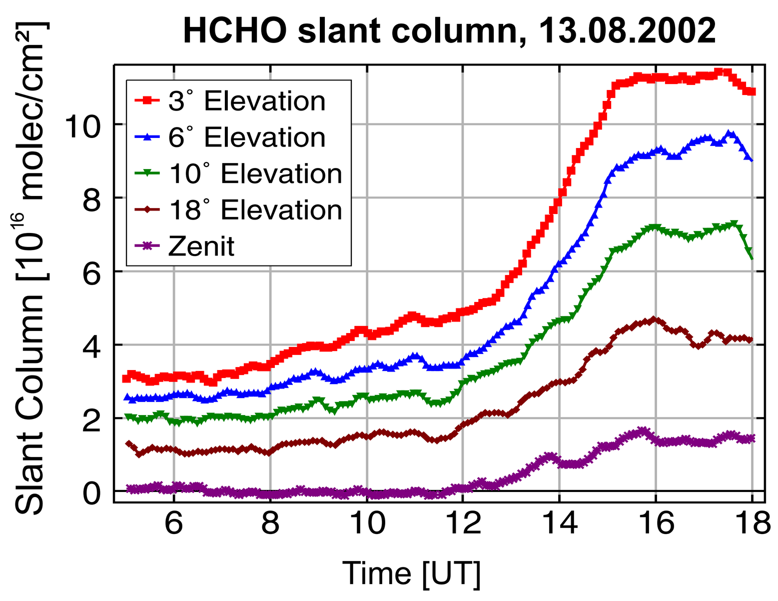


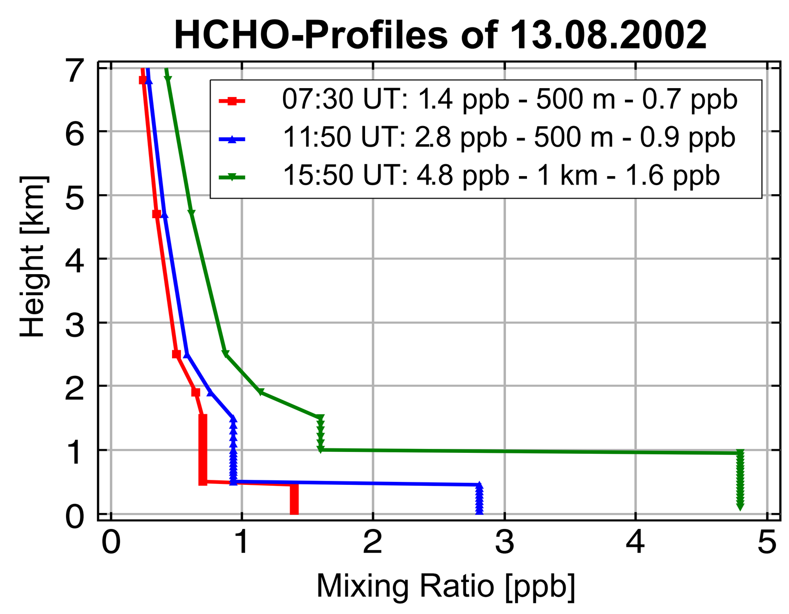


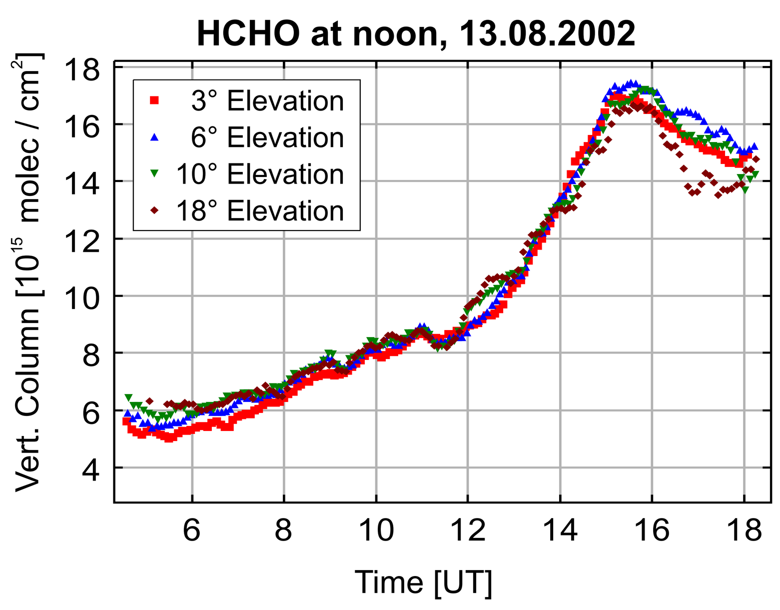


Formaldehyde Retrieval

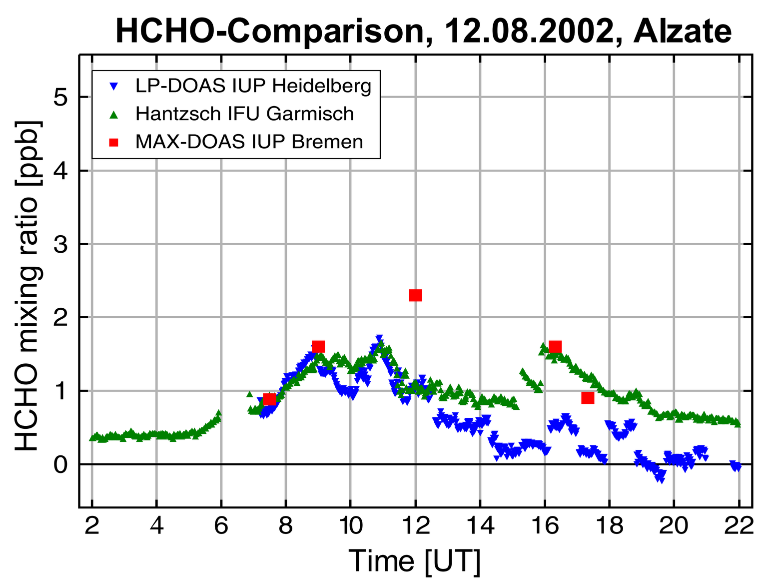
After setting parameters like albedo or aerosol by analysing O_4 it is possible to derive the formaldehyde profiles. As an example the slant columns as well as vertical columns adjusted around noon are presented for the August 13th 2002. Also shown are the profiles which were associated to certain times. By assuming that the profiles correspond to the atmospheric conditions at the specified time of measurement, mixing ratios given by a profile are the real mixing ratios.

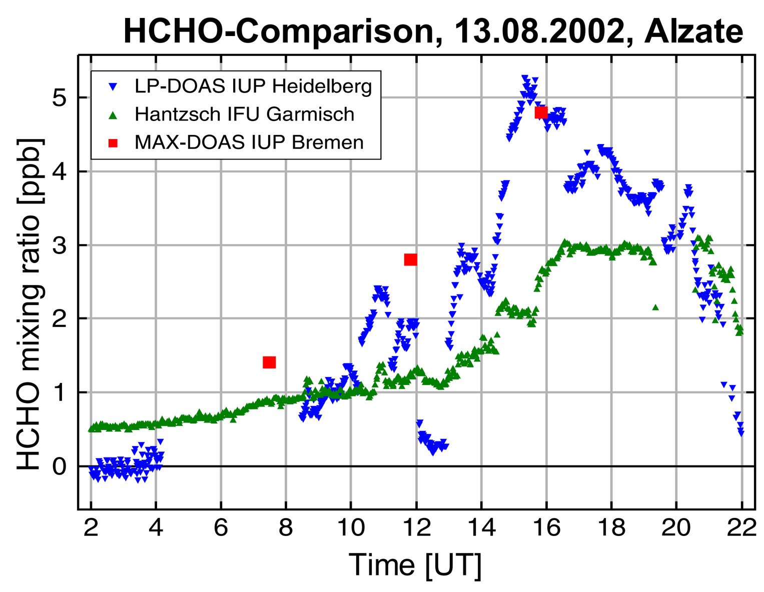






The figures below show the derived mixing ratios in comparison to other in-situ measurments placed at the same location. The Hantzsch instrument from the Institute for atmospheric research in Garmisch, operated by W. Junkermann, sampled the air at a height of approximately 2m above ground. The long path DOAS instrument, by C. Hak, Institute of Environmental Physics, Heidelberg, was measuring along a path between ground level and 50m height and a path length of about 10km. In contrast, the mixing ratios of our MAXDOAS instrument were assigned to a layer of minimal 500m height. These differences in the experimental setup are assumed to be responsible for the deviations seen in the plots.





Conclusions

- First time of analysing formaldehyde in a systematic manner using a groundbased MAXDOAS instrument.
- First time of deriving mixing ratios which have good quality and are comparable to in-situ measurments.
- Additionally, it was possible to retrieve information about the aerosol extinction

Selected References

[1] Rozanov, A., V. Rozanov and J.P. Burrows, *A numerical radiativ transfer model for a spherical planetary atmosphere: combined differential approach involving the Picard iterative approximation*, J. Quant. Spec. Rad. Trans., 69, 491-512, 2001
[2] Wittrock, F., A. Richter, A. Ladstätter-Weißenmayer and J.P. Burrows, *Global observations of Formaldehyde*, in proceedings of the ERS-ENVISAT symposium, Gothenburg October 2002, ESA publication SP-461, 2000

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