Atmos. Chem. Phys. Discuss., 11, C5748–C5753, 2011
www.atmos-chem-phys-discuss.net/11/C5748/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Airborne DOAS measurements in Arctic: vertical distributions of aerosol extinction coefficient and NO<sub2</sub> concentration" *by* A. Merlaud et al.

Anonymous Referee #2

Received and published: 30 June 2011

This article describes aerosol extinction coefficient and NO2 profiles measured in the Arctic troposphere using an airborne UV-VIS DOAS limb scanning instrument during the POLARCAT-France campaign (April 2008). The manuscript addresses an important topic of pollution transport to the Arctic, and its vertical distribution in the Arctic troposphere. In my opinion, it is well suited for publication in ACP.

General comments: 1. The authors tend to cite the most recent works, not giving an appropriate credit to the pioneering researchers. 2. The discussion of the profile retrieval using the optimal estimation inversion seems too general. In my opinion, it will be very helpful to present authors' implementation of the theory. For example, what

software package was used to do the non-linear least squares fitting of the measured and simulated SCD. Were all of the altitude scans analyzed simultaneously or independently? How Sa is used to optimize the dofs? How long does it take to do the inversion of the full profile? It might be useful to go into more detail about the implementation differences between the logarithmic and linear inversion. I am still not clear what spectrum was used as a reference. 3. Discussion of the results is comprehensive. Auxiliary in-situ measurements and back trajectory and emission modeling were used to support and explain the findings 4. English and style can be improved to make reading smoother.

Technical comments:

p. 13526, line 10: "aerosol extinction profiles (AEP) more consistent with the AEP calculated from the independently measured aerosol size distributions"; p. 13526, line 11: We present results from two soundings .. exchange Differential Optical Absorption Spectroscopy (DOAS) on line 18 (p. 13527) with DOAS on line 15; p. 13527, line 27: referred hereafter as O4, which has strong absorption bands; p. 13528, line 2: The vertical resolution retrieved from the ground-based measurements remains however poor; p. 13529, line 16: 100 μm wide; p. 13529, line 19: 2048 x 512 pixels2; p. 13529, line 21: Do you really mean 19'? In any case, please convert to SI units (19 ft = 5.791 m); p. 13529, line 23. Please clarify what a record is in the following sentence: "Each measurement represents a 30 s average at a certain telescope angle, the latter is changed after each completed record.";

p. 13530, line 17: compared;

p. 13530, line 18: between 0.02 and 3 μm; p. 13531, line 7: Remove "mainly"; p. 13532, line 6: Please rephrase: "DOAS analysis results are, for each considered absorber, differential slant column density (DSCD), i.e. the differences between the concentration integrated along the optical path of the measurement and the corresponding quantity in the reference spectrum";

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p. 13532, line 25:

Please replace this sentence with a list of atmospheric and observation parameters affecting atmospheric radiative transfer (with application to airborne DSCD measurements).

“The DSCDs obtained with the DOAS analysis depend on the light path, which is different for every single observation due, e.g. to the telescope scanning.”;

p. 13533, line 9: Please replace the following sentence with the one below. It is not clear so far, what reference spectrum is used in DOAS analysis.

“The quantity retrieved with DOAS being a differential SCD, the reference SCD (SCDref), i.e. the integrated concentration along the optical path in the reference spectrum, must be determined”

To calculate the true SCD from the DOAS DSCD measurements, SCD in the reference spectrum (SCDref) must be estimated (SCD = DSCD + SCDref);

p. 13533, line 14: in-situ; p. 13533, line 15: replace “like” with “such as”; p. 13533, line 17: Are you scaling the TOMSV8 climatology profile by the total O3 column over Ny-Alesund? Please clarify.

“The latter uses as input the ozone total column, estimated at 390 Dobson units in our case from the AURA AVDC values at Ny-A° lesund and Tromsø (http://avdc.gsfc.nasa.gov/).”;

p. 13533, line 27: Include reference for OPAC, and indicate that the closest wavelength to O4 360 nm absorption band in OPAC is 350 nm.

M. Hess, P. Koepke, and I. Schult (1998): Optical Properties of Aerosols and clouds: The software package OPAC, Bull. Am. Met. Soc., 79, 831-844.;

p. 13533, line 29: The full limb scan from -5° to + 5° (Δ = 1°, 30 sec per position) takes about 5.5 min. For the first sounding, (as0831) this corresponds to 1.3km change in

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altitude. It is not clear if 30° azimuth change takes place within the full limb scan (5.5 min) or just 30 sec at each angle;

p 13534, line 2

Why 9 intermediary SCDs? If they represent SCD at each elevation angle in the full limb scan should it be 10 (or 11 including reference?). Or is it within a 30 sec observation at a single elevation angle? Please explain how you deal with the difference in altitude between -5° and 5° elevation angles.

Please make it clear that the assumption is that the aerosols and NO2 are homogeneously distributed within 30° (in azimuth direction);

p. 13534, line 5: This sentence seems redundant: "Once the atmospheric state and the geometry are defined, the sensitivity of the measurement to a parameter x can be expressed as the derivative δSDC/δx.";

p. 13534, line 16 In earlier sections you talk about scanning $\pm$ 5° from the horizon (elevation angle), here "close to the horizon (90°)," you change to zenith angle. Please be consistent;

p. 13534, line 28: it is not clear why you are referring to Fig. 1 here and Fig. 4 on the next line;

p. 13535, line 5: I would recommend rephrasing this sentence. The sensitivity of the airborne limb measurements close to the ground will be the same as ground-based MAX-DOAS. The ability to describe free troposphere comes from the fact that horizon scanning (with high sensitivity) is done at multiple altitudes. "But this sensitivity decreases rapidly with altitude contrary to our airborne set-up, which indicates that this approach is particularly well suited for the study of the free troposphere.";

p. 13540, line 1: Could you please clarify this sentence? Does it mean that you use 11 reference spectra one for each elevation angle ($\pm$ 5°, Δ = 1°) collected at 6km altitude? "O4 and NO2 DSCDs presented in the following are relative to their respective columns

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in the same spectrum, selected at the top of the as0831 sounding, near 6 km altitude.";

p. 13540, line 9: Strictly speaking, only binary, not absolute, O2O2 absorption cross section [Pfeilsticker et al., 2001] is used/measured since the equilibrium constant between [O2] and [O2O2] collision complex is not known. It also means that the absolute O2O2 vertical distribution is not known, but rather pseudo column density in molecules2/cm6. "There remain some uncertainties regarding the absolute value of the O4 absorption cross-section and measured DSCDs are commonly corrected with ad hoc scaling factors to retrieve extinction.";

p. 13547, line 26: Please replace "The first sounding shows an interesting mixing" with the following "HYSPLIT back trajectories calculated for the first sounding suggest a potential mixing";

p. 13548, line 4: Please replace "The second sounding:" with the following "HYSPLIT results for the second sounding";

p. 13548, line 24: add "s" to indicate p. 13550, line 2: please delete "there" p. 13550, line 6: please replace "novel" with "recently developed"

Citations: p 13555 line 17: correct citation 10.1175/1520-0477(1995)076<2403:TAHP>2.0.CO;2;

Figures: Reference to figures varies (Figure x and Fig. x) please be consistent

Fig. 3: Please indicate what the observation conditions of the spectra used in this fit are: viewing elevation angle, reference spectrum, SZA, RAA, and aircraft altitude.

Fig. 4: How where the viewing elevation (telescope line-of-sight) angles selected for this figure? Do they represent the maximum sensitivity at each aircraft altitude? It might be more informative to use [molecules*cm-2 / molecules*cm-3] on the left panel.

Fig. 5: please indicate the wavelength at which aerosol extinction was calculated (350 nm).

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Fig. 7: SCD = Slope* DSCD + SCDref. Abs. cross section correction factor = slope

Fig. 10: Comparison of the aerosol extinction profile retrieval using linear and logarithmic weighting functions for the sounding of the as0831 flight (8 April 2008).

Fig. 11. Only averaging kernels and retrieved profiles are shown (no weighting functions)

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13525, 2011.