Interactive comment on “Airborne DOAS measurements in Arctic: vertical distributions of aerosol extinction coefficient and NO<sub>2</sub> concentration” by A. Merlaud et al.

A. Merlaud et al.
alexism@oma.be

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Referee #1 comments (comments received and published: 20 June 2011)

UV/vis limb measurements from the ground, aircrafts, balloons or satellite is not a new technique at all, and therefore the employed technique builds on past experiences. In fact, I wonder whether the heritage of your aircraft Limb measurement is actually ground-based AMAX-DOAS, rather than UV/vis measurements from space? Digging into these past studies, I feel that the UV/vis limb technique actually traces back to the SME and SBUV satellite measurements in the early 1980 rather than AMAX ‘reinvented’ much later. Appropriate references are e.g.,


Reply: We agree that a reference to satellite-borne measurements was missing in the introduction and thus added the suggested reference on SME, in the second paragraph of the introduction. We explain further that uv-vis limb geometry from space is appropriate to study the stratosphere but not the troposphere, which adds rationale to our experiment.

“On the other hand, satellite-borne instruments using UV-visible limb radience measurements date back to the Solar Mesosphere Explorer (SME, Mount et al. (1984)). This geometry provides a good vertical resolution in the stratosphere, but for tropospheric studies from satellite, nadir-looking instruments have to be used, as the Ozone Monitoring Instrument (OMI, Levelt et al. (2006))”

Considering the second suggested reference, we note that SBUV was a nadir-looking sounder; it is less relevant in the present context. Therefore we did not add it.

Also as said in my previous review, I’m not sure by which observation geometry c.f scanning limb versus changing the aircraft altitude, more information is gained to infer profiles of the targeted atmospheric parameters. Eventually this issue could be more emphasized in the manuscript, even though it may not change to overall results. Reply: Arguably the best measurement geometry would be obtained by changing the altitude and simultaneously correcting for the pitch/roll variations to maintain the telescope par-
allel to the ground. In this way the sensitivity would be maximal at the altitude of observations, as explained in the last paragraph of our conclusion. Since we wrote the article we have learnt that this has been achieved by the group of Rainer Volkammer from Boulder with their CU-AMAXDOAS and they achieve a very high vertical resolution compared to us. To our knowledge their results have not yet been published. We added a sentence to emphasize this issue in the paragraph discussing the weighting functions. "The maximum sensitivity is achieved for viewing angles close to the horizon (90° at the altitude of observations due to the enhanced light path in this layer. This indicates that the optimal geometry for profiling applications is obtained during the ascents or descents of the plane while maintaining the telescope parallel to the horizon." Technical comments: 1. Page 13526: We report airborne differential: : : :change to : : : We report on airborne Differential

2. Throughout the manuscript: Change from Prados et al. (2010) ..... to Prados et al. (2011). .. since the has not been published past year. 3. Page 13534, second paragraph: In the derivative _SCD/_x the _ in front of the x is missing. Reply: All three corrected in the text. 4.Page 13540:There remain some uncertainties regarding the absolute value of the O4 absorption cross-section and measured DSCDs are commonly corrected with ad hoc scaling : : : Comment: Probably the absolute value of the O4 absorption cross-section will never be known, since it would require to absolutely measure the O4 concentration for relevant atmospheric conditions. Instead the collision absorption cross sections for O4 are known (e.g., Greenblatt et al., JGR, 1990) including their weak T dependence (e.g. Pfeilsticker et al., GRL, 2001). Accordingly with respect to the present knowledge on the nature of O4, this sentence is totally meaningless, and! very likely it will remain so forever.

Reply: We definitely agree that O4 should not be considered as a normal molecular absorber, and this is why we use the term O4 after having defined it as an (O2)2 collision complex in the introduction. The cross-sections we use here (Hermans et al.) have been measured between 0.4 and 0.9 atmosphere i.e. under conditions similar
to those encountered in atmospheric experiments. We have removed the expression ‘absolute value of the O4 cross-section’ and wrote: ‘O4 DSCD measurements are commonly corrected with ad hoc scaling factors to retrieve extinction.’

5. Page 13544/13545: The same holds true for the second flight, indicating that a lognormal assumption on the density of probability of extinction can be useful for its retrieval : : : : change to : : : : The same holds true for the second flight, indicating that an assumed lognormal probability density function (pdf) for the distribution of extinctions is well suited.

Reply: Corrected in the text. 6. Page 13545: The drawbacks of the retrieval scheme are first a slower convergence; it takes generally two iterations for the linear retrieval and five for the logarithmic one, which may come 5 from a smaller degree of linearity in the logarithmic statement of Eq. (2) as mentioned by Schneider et al. (2006) for water vapor retrievals: : : : : change to : : : : The drawbacks of the retrieval scheme is a slower convergence of a ‘linear’ aerosol extinction pdf as compared to a ‘logarithmic’ pdf, since, it generally takes two iterations for the former as compared to latter. For water vapor retrievals (by what method?), this has also been noticed by Schneider et al. (2006).

Reply: Changed to:

“A drawback of the logarithmic retrieval scheme with respect to the linear one is a slower convergence, it generally takes five iterations for the former as compared to two for the latter. This may come from a smaller degree of linearity in the logarithmic statement of Eq. (2) as mentioned by Schneider et al. (2006) for water vapor retrievals from ground-based infrared spectra.”

7. Page 13545: A second limitation lies in a probable underestimation of the errors when the retrieved value is low, as above 5 km in Fig. 10: : : :change to : : : : A second limitation comes with a likely underestimation of the errors, when the retrieved extinction is low, e.g., for altitude above 5 km (see Fig. 10)
Page 13545: This is due, as the sensitivity reduction with extinction described in Sect. 3.2, to the logarithmic behavior toward small values.

Comment: I do not understand this sentence, so please correct.

Reply: We agree that the expression ‘sensitivity reduction with extinction’ is confusing, so we just delete it without replacing it because we explain what we mean in the following sentence.

Page 13545: Three zones are distinguishable, the boundary layer with a concentration of $1.9 \pm 0.3 \times 10^9$ molec cm$^{-3}$, the lower free troposphere with around $25 \pm 1 \times 10^8$ molec cm$^{-3}$ between 1 and 4 km altitude and the higher troposphere with negligible concentrations.

Reply: Changed to: “Three zones are distinguishable: (a) the boundary layer with a NO2 concentration of $1.9 \pm 0.3 \times 10^9$ molec/cm$^3$, (b) the lower free troposphere with around $25 \pm 1 \times 10^8$ molec/cm$^3$ between 1 and 4 km altitude and (c) the higher troposphere with negligible concentrations.” We prefer not to use the expression detection limit here since it may be mistaken with the detection limit calculated in Sect. 4.3 which is valid for the boundary layer only.

Page 13546: This value appears close to our measurement but it lies inside the error bars which indicates that such low concentrations are not detected by OMI.

Reply: Changed to: “This value is close to our measurement but it lies inside its own error bars, which indicates that such low NO2 concentrations can barely be detected by OMI.”

Page 13546: For the second sounding, the lidar profile was measured at 11:40 UTC above 69.6 N, 19 E i.e. two hours and 60 km off the sounding because the clouds mentioned in the previous section disturbed the measurements at the sounding time.

Reply: Corrected in the text.


Reply: Reference added.

Page 13548: The presence of the short-lived NOx in the Arctic is usually explained from local sources, such as peroxyacetic nitric anhydride (PAN) decomposition (Stroud et al., 2003), ships (Wittrock et al., 2004) or snow photochemistry (Honrath et al., 1999). NO2 from PAN decomposition is a long-range source, and not as stated a local source. In order to verify that PAN decomposition is a local NOx source, please calculate and explicitly add the life time for PAN decomposition at relevant T!

Reply: We meant that PAN decomposition in the Arctic is a local source of NO2 since it produces NO2 in Arctic, even if PAN itself is long range transported, but we agree it is confusing so we changed the sentence: “The presence of the short-lived NOx in the Arctic is usually explained from peroxyacetic nitric anhydride (PAN) decomposition (Stroud et al., 2003), from local sources like ships (Wittrock et al., 2004) or snow photochemistry (Honrath et al., 1999)”

Page 13548: The lifetime of NO2 depends on the meteorological conditions: against which process, e.g. reaction with OH into HNO3 and than wash-out or what?
According to Evans and Jacob, GRL, 2005: “NOx is permanently removed from the atmosphere by conversion to nitric acid followed by deposition. During the day, this conversion occurs through the reaction of NO2 with OH, and during the night by hydrolysis of N2O5 in aerosols.” We added the following sentence in our text:

“The lifetime of NO2 depends on the meteorological conditions, its main diurnal sink being the reaction with OH, and at night the hydrolysis of N2O5 on aerosols (Evans and Jacob, 2005). This lifetime can however be approximated by a decreasing exponential depending on the surrounding air mass temperature.”

15. On page 13549: Interchange the word ‘higher’ with ‘larger’ and wherever it appears appropriate, e.g. where the in situ sounding vmr are the highest! where the in situ sounding vmr are the largest. …and so on.

Reply: Corrected in the text.

16. On page 13549: Notice that the simulated total NOx tracer measurements are much higher than the measured NO2 because NOx has a much shorter lifetime than the 20 days over which emissions are accumulated in the model. 

Notice that the simulated measurements of total NO2 are much larger than the actual measured NO2 because NOx has a much shorter lifetime than the 20 days over which emissions are accumulated in the model. Comment: I do not understand the second half of the sentence, neither form the English nor from its meaning. So please explain and reformulate.

Reply: We changed to:

“Notice that the simulated total NO2 tracer measurements are much larger than the measured NO2, because the model accumulates emissions over 20 days without considering removal processes.”

17. Page 13549: with known errors in the emission inventories used (Prank et al., 2010): 

Comment skip ‘used’!

Reply: Corrected in the text.

18. Page 13550: In particular, emissions in Nikel in the western Kola Peninsula, which the observed air mass traversed, are too low and appear erroneously attributed to Murmansk in the inventories (Prank et al., 2010): 

In particular, NOx emissions transported from Nikel which is located on the western Kola Peninsula are apparently too low and appear erroneously attributed in the inventories to emissions of Murmansk (Prank et al., 2010).

Reply: Corrected in the text.

19. Page 13550: For both soundings, the free troposphere extinction matched a layer with enhanced CO, indicating pollution transport, with rather different absolute values however. The small extinction detected in the first sounding is explained from back-trajectories as a mix between stratospheric and polluted air from Northwestern Europe, while the higher extinction seen the next day originated mostly from central Europe: 

For both soundings, the extinctions inferred for the free troposphere match layers of enhanced CO which indicates pollution transport. The magnitudes are however much different. The small extinction detected in the first sounding is explained from back-trajectories indicating a mixture of stratospheric air and polluted air transported from Northwestern Europe, whereas for the second sounding the air masses mostly originated from central Europe. 

Reply: Corrected in the text.

Referee #2 comments (comments received and published: 30 June 2011) General comments:

1. The authors tend to cite the most recent works, not giving an appropriate credit to the pioneering researchers.

Reply: We added a reference for the satellite uv-vis limb measurements (SME) which has a longer history than airborne limb doas measurements (see 1st reply to referee 1).
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.

Reply: The retrieval scripts were done in Matlab from scratch using Rodgers book with useful advices from colleagues duly acknowledged if not coauthors. We used the Gauss-Newton algorithm for the iterations. We agree this was not clear enough in the paragraph so we replaced “e.g with the GN algorithm” by “in our case with the GN algorithm”. We also added: “Both soundings are retrieved independently.” in the next paragraph. Considering Sa we used a diagonal matrix with element $Sa(i,j) as \beta (linear case) or \beta ln(xa(i,j))2 (logarithmic case), then we tune \beta to optimize the dofs. This was added in the text. Being more accurate about the differences between the logarithmic and linear retrievals involve doing more simulations, we have started to work on that but this would be out of the scope of this paper. The reference spectrum was also added in Sect 4.1.

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”; 
Reply: We prefer to avoid acronyms.

p. 13526, line 11: We present results from two soundings ..
Reply: Corrected in the text
exchange Differential Optical Absorption Spectroscopy (DOAS) on line 18 (p. 13527) with DOAS on line 15;
Reply: We prefer our formulation since it explains DOAS at the beginning of a paragraph, DOAS has been explicated in the abstract already
p. 13527, line 27: referred hereafter as O4, which has strong absorption bands;
Reply: Corrected in the text
p. 13528, line 2: The vertical resolution retrieved from the ground-based measurements remains however poor;
p. 13529, line 16: 100 um 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);
Reply: All four corrected in the text
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.”;
Reply: In this sentence record is synonym to measurement, i.e. an average of spectra during 30 seconds, we agree it is not clear and thus have modified the sentence.

p. 13530, line 17: compared;
p. 13530, line 18: between 0.02 and 3 um;
p. 13531, line 7: Remove “mainly”;

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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.

The DSCDs obtained with the DOAS analysis depend on the light path, which is different for every single observation due to the telescope scanning and the variations in azimuth and altitude when the planes perform a vertical sounding.

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).

The TOMS V8 climatology depends on the total column, see McPeters, JGR, 2007 ('For the TOMS retrievals, it was important to include a total ozone dependence, making this in effect a four-dimensional climatology (latitude–month–altitude–ozone)'). We replaced 'uses as input' with 'depend on'.
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);

Reply to the last two comments: 9 SCDs are measured within 30 sec at a single elevation angle, during which the azimuth vary by 30°. We reformulated the whole paragraph to make it clearer:

“The observation geometry is not constant during a measurement due to the circular flight pattern and the 30s accumulation time. In particular the relative azimuth angle varies by up to 30°. To overcome this problem, we calculate each SCD at a single telescope angle as a weighted mean of 9 intermediary SCDs equally distributed in the 30s measurement interval. The weights correspond to the different radiances calculated in the respective intermediate SCD geometries, defined by the corresponding orientations and altitudes of the aircraft, telescope angles and solar positions.”

Considering the assumption that the aerosols are equally distributed in the azimuth range, it is imposed by the radiative transfer model, in which the atmospheric parameters are entered as a profile and not as a 3d field.

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 $\frac{\Delta \text{SDC}}{x}$.”

Reply: We agree that it is redundant but it helps to follow anyway.

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;

Reply: The telescope scanning angle is not the same as the angle between the horizon and the telescope, since there is the plane’s roll which is varying. We added: “This happens when the telescope angle compensates the plane’s roll” to make it clearer.

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

Reply: Actually I am referring respectively to Fig.1 and Fig.4 in the papers of Zhou and Wittrock. I am comparing these figures to my figure 4.

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.”

Reply: We changed the text to:

“Ground-based MAX-DOAS measurements are most sensitive close to the ground, corresponding to quasi horizontal pointing, when the light path enhancement is maximum. The box AMF, around 20, is then comparable to airborne limb measurements. But this sensitivity decreases rapidly with altitude contrary to our airborne set-up which enables to look the horizon from any altitude reachable by the plane. As a result, the airborne 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 ($\pm 5$, $\pm 1$) collected at 6km altitude?

“O4 and NO2 DSCDs presented in the following are relative to their respective columns in the same spectrum, selected at the top of the as0831 sounding, near 6 km altitude.”

Reply: We use the same reference spectrum for all telescope angles. We added: “The telescope angle was then 0°” to make it clearer.

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 ab-
solute 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."

Reply: We agree and we modified our sentence (see reply to the first referee on a similar remark.)

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

p. 13548, line 4: Please replace “The second sounding:” with the following “HYPLIT 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”

Reply: All five corrected in the text

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

Reply: Corrected in the text.

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

Reply: We have tried to follow the instructions from ACP (“Fig.” should be used when they appear in running text followed by a number unless they come at the beginning of a sentence).

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.

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Reply: We added: “when the plane was in the marine boundary layer” in the caption and the other information requested in the text to keep the caption short.

“The spectrum was recorded at 9h57 UTC when the aircraft was flying at 0.43 km altitude inside the marine boundary layer during the flight as0831 (8 April 2008). The solar zenith and relative azimuth angles were then respectively 64° and 223°. The telescope angle was -1° which corresponded to a range between -10° and +20° around the horizon, due to the plane’s roll variations during the 30s of the measurement.”

Fig. 4: How were 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.

Reply: As mentioned in the text, these weighting functions are typical and we have selected a few of them to make with various angles to show how the sensitivity was varying with the telescope zenith angle. They do not thus represent the maximum sensitivity at each layer. It was not possible to plot all the weighting functions since it would have been overloaded. We prefer to keep the cm unit for the x-axis since the weighting functions are related to the length of the photon path in the layers.

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

Reply: Added in the caption

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

Reply: There were some inconsistencies in our text between scaling factor on the DSCD and on the cross-sections, which are supposed to have an inverse relationship. As we dropped the expression ‘absolute O4 cross section’ in response to a previous comment, we only refer here to a scaling factor applied to the DSCDs.

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).

Reply: Corrected in the text. I have also changed the caption of Fig. 11 accordingly.
Fig. 11. Only averaging kernels and retrieved profiles are shown (no weighting functions) Reply: Corrected in the text

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