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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 #1

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The present manuscript reports on aircraft-borne UV/vis limb measurements of NO2 and the aerosol extinction in the Arctic troposphere during POLARCAT in April 2008. For two examples, supporting air mass trajectory calculations provide evidence on the causes for the detected indicators (NO2, aerosol extinction and CO) for air pollution in the Arctic. I feel that the manuscript has much improved from the version I read before, but still it contains some oddities with the English. Also a reaction to some other comments which I previously rose is still missing in the revised manuscript. Overall I feel that manuscript is well suited for a wider readership in ACP, and it might be acceptable after appropriate reaction to may comments.

Major comments:

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.,

ïĂ■ Mount, G. H., D. W. Rusch, J. F. Noxon, J. M. Zawodny, and C. A. Barth, Measurements of Stratospheric NO2 from the Solar Mesosphere Explorer Satellite 1: An overview of the Results, J. Geophys. Res., 89, pp. 1327, 1984. ïĂ■ Heath, D.F., A.J. Krueger, H.A. Roeder, and B.D. Henderson, The solar backscatter ultraviolet and total ozone mapping spectrometer (SBUV/TOMS) for NIMBUS G, Opt. Eng., Vol. 14, pp. 323-331, 1975.

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.

Technical comments:

The manuscript still contains a considerable number of oddities with the English (grammar, typos, ..) though its English improved much from the earlier version. Also the meaning of some sentences is very hard to decipher, accordingly I list a number of recommendations for the correction.

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 δSCD/δx the δ in front of the x is missing. 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 been 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 for ever. 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. 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). 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) 8. 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. 9. Page 13545: Three zones are distinguishable, the boundary layer with a concentration of 1.9$\pm$0.3$\times$109 molec cm$-$3, the lower free troposphere with around 25

3$\pm$1$\times$108 molec cm$-$3 between 1 and 4 km altitude and the higher troposphere with negligible concentrations. . . .. Correct to . . .. . .Three zones are distinguishable (a) the boundary layer with a NO2 concentration of 1.9$\pm$0.3$\times$109 molec cm$-$3, (b) the lower free troposphere with around 25 3$\pm$1$\times$108 molec cm$-$3 between 1 and 4 km altitude and (c) the upper higher troposphere where NO2 concentrations were below the detection limit. 10. 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. . .. . . change to . . . The OMO NO2 measurements is close to our measurement and it lies within the stated error bars, but it also indicates that such low NO2 concentrations can barely be detected by OMI. 11. Page 13546: For the second sounding, the lidar profile was measured at 11:40UTC 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. ..change to . . .. For the second sounding, the lidar profile was measured at 69.6_ N, 19_ E around 11:40UTC i.e. 60 km and two hours off our sounding mainly since later the cloud cover prevented a co-located measurement. 12. Page 13547: Using CO/O3 correlations to trace tropospheric and stratospheric air amsses, much earlier studies exist than the two cited in the manuscript, e.g. Koike, M., Y. Kondo, S. Kawakami, H. Nakajima, G. L. Gregory, G. W. Sachse, H. B. Singh, E. V. Browell, J. T. Merrill, and R. E. Newell (1997), Reactive nitrogen and its correlation with O3 and CO over the Pacific in winter and early spring, J. Geophys. Res., 102(D23), 28,385–28,404, doi:10.1029/97JD02085. 13. 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 e relevant T! 14. 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? 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. 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 ... . .change to . . . 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. 17. Page 13549: with known errors in the emission inventories used (Prank et al., 2010). . .Comment skip ‘used’! 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 13549 attributed to Murmansk in the inventories (Prank et al., 2010). . ..change to . . . 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). 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. . ..change to . . . 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.

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