

Interactive comment on “The CU Airborne MAX-DOAS instrument: ground based validation, and vertical profiling of aerosol extinction and trace gases” by S. Baidar et al.

Anonymous Referee #3

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The paper describes a novel aircraft-borne optical spectrometer and some atmospheric test measurements, including a validation of the inferred data with auxiliary measured data. The employed method builds on (a) the measurement of Limb and Nadir optical spectra, (b) the retrieval of the measured atmospheric parameters i.e. of UV/vis light absorbing trace gases and the aerosol extinction by means of Differential Optical Absorption Spectroscopy (DOAS), (c) forward radiative modeling of the Limb observations (whereas for the interpretation of the Nadir measurements a surrogate method is used), and (d) a mathematical inversion technique (is it linear?, see my comment I.2.b below) to infer vertical profiles of the targeted atmospheric parameters from the set of

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Limb observations. While the strength of the manuscript is with the description of the instrument and the validation of the inferred data by quasi-independent observations, some weakness comes with the description of the steps (c) and (d). In addition, the manuscript contains some statements, which are in contradiction to other statements made in the same manuscript and/or to the generally available wisdom in field. In a revised version of the manuscript, these deficits need to be removed.

I. Major comments:

1. Since all the essentials of the used technique and methods are not all new (even though the technique is renamed in the manuscript), it should be stated early in the manuscript (e.g. in the introduction), that the AMAX technique (or variants of it) primarily builds on the well-known Limb observation technique. For your information, the Limb technique is deeply rooted in astronomy. It started in 1907 when first limb observations were performed by the famous Catalan astronomer Josep Comas Solà. The observation led him to discover the atmosphere of Titan which is unfortunately not published in modern form. Later the technique was further developed by Milne, 1921, van de Hulst, 1950, Barth et al., 1969, Barth et al., 1971, and many others (see refs below). In the 1960's, Limb observations started in the terrestrial atmosphere from aboard the Nimbus satellites (e.g., Rodgers, 1976, Haas&Shapiro, 1985, and refs therein). Then in the early 1980's the Limb technique was employed for atmospheric observations from high flying research balloons (e.g. Water et al. 1981), and later (in the early 1990s) from research aircrafts (e.g., Traub et al., 1991). Since all these applications of the Limb technique are essentially containing the same elements as the method described in the manuscript, it appears worthwhile to trace your study back to this history (c.f. also by using a proper nomenclature). This would also provide to a wider readership a better orientation to understand the methods described manuscript.

2. Very disturbing are contradictory statements made in the manuscript, for example (a) the statement made on the DOF's and that 'the result is independent from the signal to noise' et cetera, made on page 7244, line 28 and again on page 7266 line 29. This

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statement is in utmost conflict with the presented equations c.f., following your equation 4, since your $S(\epsilon)$ could essentially be determined by the signal to noise, if all other measurement errors were negligible. Therefore, the solution vector (your x) is always determined by all factors going into $S(\epsilon)$, hence the measurement error as well. So at best you can state that the signal to noise of your (DOAS) measurement is not a major contributor to $S(\epsilon)$. Here, an informed reader would like to learn however, what factors of what magnitude are contributing to your $S(\epsilon)$. For example what are the uncertainties in $F(x,b)$ (partly given in Table 5) or K , all related to your forward (RT) model? When discussing of your DOF and error budget you could also refer to Roscoe and Hill, (2002).

(b) The statements made in the manuscript regarding an 'iterative approach' are very disturbing (c.f., Page 7258, lines 8 and 24, and on page 7264 line 20) since the mathematics of your 'a posteriori' inversion scheme does not require iterations (see your equation 4). So what inversion technique did you really use? Did you apply a non-linear scheme in order to solve your equation 3, or did you just play around with parameters until you found the result 'convincing'? Explain.

3. Radiative modeling: Even though treated to be independent, the SSA and the asymmetry parameter g are not total independent parameters. This can be seen from the definition of the SSA ($k(\text{scatter})/[k(\text{scatter})+k(\text{abs})]$), the definition of the (effective) free mean path length (i.e. the length until photon are directionally randomized) which is frequently implemented in RT codes i.e. $k(\text{scatter})^{-1} = \lambda(\text{eff}) = \lambda(\text{Mie})/(1-g)$, where $\lambda(\text{Mie})$ is the free path for Mie scattering, and g is the asymmetry parameter. How these different definition of $k(\text{scatter}) = 1/\lambda(\text{eff})$ (in SSA) or $k(\text{scatter}) = 1/\lambda(\text{Mie})$ are dealt with in the used RT needs be clarified before the firm conclusion regarding the uncertainty or (cross) sensitivity of the asymmetry parameter (e.g., on page 7259 line 12, and page 7268 line 28) and its independence on the SSA and inferred extinction can be made.

In fact I recommend to parameterize the model with respect to the SSA for $g = 0$ and for

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the final discussion/inter- comparison rescale (see above) the inferred extinction with an actual or assumed typical g value for urban aerosols. Here for the aerosol retrieval it could also very helpful to increase the information content by not only using information from measured O_4 slant column but also from relative radiances.

4. The statements made on the T dependence and 'pressure effects?' of the O_2-O_2 absorption bands (page 7258, line 8 to 23 page 7263, line 10 to 25) are scientifically incorrect, hence not useful. First from O_2-O_2 collision experiments in the laboratory, the nature, structure, orientation dependent as well as the thermally averaged well-depth ($D_e(O_4) = -(1130+80) \text{ J/Mol}$) is well known (Aquilanti et al., 1999). Second, the nature of the O_2-O_2 absorption bands is well understood from theoretical studies (e.g. Biennier et al., 2000). Third, the weak T -dependence of the O_2-O_2 absorption bands found in the laboratory (e.g. Long and Ewing, 1973) as well as in the atmosphere (e.g. Pfeilsticker et al., 2001) can concisely be reconciled to the nature of the O_2-O_2 interaction. So there is no need to speculate on (a) either pressure effects, (b) an alpha factor for which you fortunately infer $\alpha = 1$, or (c) the T -dependence of the O_2-O_2 absorption. In fact I suppose that studies claiming an $\alpha \neq 1$ are subject to deficits in correctly dealing with the RT, for example neglecting the dependence of the SSA on g , and $k(\text{abs})$ et cetera.

II. Other comments:

(a) I strongly recommend to inter-change section 3.3. with 3.4 and 3.5, because the way you deal with your Nadir observations is a 'poor man' (or approximate) version of the inversion methods introduced in section 3.4 and 3.5. Also, I recommend to strictly separate the Nadir and Limb observations (in the nomenclature as well as by sections) in order to make clear what result is obtained from what observation geometry.

(b) The statements made anywhere in the manuscript on advantage of an EA angle control need to be fine-tuned. First, an active control of EA is certainly an advantage in maximizing the DOF of the measurements, if EAs are carefully chosen in order to

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maximize the DOF. Here a sensitivity study could help a lot to demonstrate what set of EAs is most relevant to increase the DOF in aircraft-borne Limb observations. Further, as far as I know, predecessor instruments had always means to learn the actual EAs, although most of them were not actively controlling the EAs. So the statements regarding the active control of the EAs made throughout the manuscript need to be accordingly fined-tuned.

Moreover, the averaging kernels shown in Figure 11 do not really indicate that for these measurements the instrument was actually scanning through a series of EAs. Rather the AK pointed that the observations were made for a more or less stable EA (= 0° degree) during aircraft ascents and descents which is nice, but also not new. Also I found it rather disturbing how uncertainties in the EAs are discussed, in particular the coarse delta(EA) resolution (x-axis) shown in Figure 3. Here a higher resolution version would be helpful, and additionally a more concise explanation needs to be given in section 2.5 why the actual pointing error is smaller than expected based on the (Gaussian) sum of the individual errors.

III. Minor comments:

Here I summarize typos, improvements to the English, missing information, references and other stuff.

- Page 7246, line 15: 'the first true et cetera' see my comment above
- Section 2.3: Please provide information somewhere on the dispersion pixel/FWHM, since then the magnitude of over, or under-sampling for the detection of each gas can be assessed.
- Page 7250, line 28: clinometer → inclinometer
- Page 7251, line 2: It not clear whether systematic errors should and can be Gaussian added. Explain.
- Section 2.6. Please provide information on the change in pressure within the cabin

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and how the change in p and hence change the refractive index within the spectrometers affect your wavelength recording.

- Page 7253, line 2: It is great to have a separate O3 monitor aboard, but why then the O3 data are not used to be compared with retrieved O3 profiles?
- Page 7255, line 21: McArtim may simulate much more than stated here. In fact in the math following your equation (4), you use c.f., computed Jacobians from McArtim.
- Page 7259, line 19: Delete this sentence because it is not well based on facts (see e.g. Kritten et al, 2010, Merlaud et al., ...)
- Section 4.2: Validation of NO2 vertical column → Validation of the Nadir measured NO2 vertical column. ... since actually you do not infer the column by integration of an inferred NO2 profile (see my comment above II.a)
- Section 4.4.1, fourth line. ... between the measured and modeled O4 SCD. A better wording were ... between the measured and best-guessed (see above) or inferred ...
- Page 7264, line 26: Error contribution in retrieved extinction due EA uncertainty → The error contribution in retrieved extinction due to EA uncertainty. ... and then ... It illustrates
- Page 7265, line 223. $1 \text{ ppb} = 2.46 \times 10^{10} \text{ molecules cm}^{-2}$ reconsider the correctness of the dimension cm^{-2} ?
- Page 766, line 1: ...error is slightly smaller in the FT, where aerosol extinction presents less of a limitation, ... a limitation of what, and less with respect to what?
- Page 7266, line 29: Reconsider the statement with respect to the comment I.2.a.
- Page 7288, line 1: though the presence of elevated layer was observed as well → though the presence of an elevated pollution layer was observed as well. ... or → though the presence of elevated pollution layers was observed as well
- Figure caption 7: mention here ... Nadir observations

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- Insert Figure 10: Measured and modeled. The latter expression is rather confusing because it is unclear whether O₄ it is forward modeled (e.g. using the result of Fig. 10B), inferred or even retrieved

- Reference and Introduction: In the public available literature there is a larger series of previous publications on Nadir Imaging (DOAS) measurements which need to be mentioned) since you deal with Nadir observation. A selection could be

o Heue, et al., Direct observation of two dimensional trace gas distributions with an airborne Imaging DOAS instrument, *Atmos. Chem. Phys.*, 8, 6707-6717, doi:10.5194/acp-8-6707-2008,2008.

o Krings et al., Quantification of methane emission rates from coal mine ventilation shafts using airborne remote sensing data, *Atmos. Meas. Tech. Discuss.*, 5, 7383-7429, 2012, doi:10.5194/amtd-5-7383-2012.

o Thorpe et al., Point source emissions mapping using the Airborne Visible/Infrared Imaging Spectrometer (AVIRIS) see http://www.geog.ucsb.edu/~akthorpe/documents/Andrew_K_Thorpe_2012_SPIE.pdf

Some useful references referred to above:

Aquilanti et al., Molecular Beam Scattering of Aligned Oxygen Molecules. The Nature of the Bond in the O₂-O₂ Dimer, *J. Am. Chem. Soc.*, 121, 10794-10802, 1999.

Biennier et al., Structure and ro-vibrational analysis of the [O₂(1Δg)v = 0]₂-[O₂(3Σg⁻)v = 0]₂ transition of the O₂ dimer, *J. Chem. Phys.* 112, 6309 (2000).
Barth, C. A., Planetary ultraviolet spectroscopy, *Appl. Opt.*, 8, 1295, 1969.

Barth et al., Mariner 6 and 7 Ultraviolet Spectrometer Experiment: Upper Atmospheric Data, *JGR*, 76, 10, 2213, 1971.

Long, C. A., and G. Ewing, Spectroscopic investigation of van der Waals molecules¹, . Infrared and visible spectra of (O₂)₂, *J. Chem. Phys.*, 58, 11, 4824 - 4834, 1973

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Milne, E.A. *MNRAS* 81: 361-375, 1921.

Haas, I. S., R. Shapiro, "The Nimbus Satellite System: Remote Sensing R&D Platform of the 1970s," *Monitoring Earth's Ocean, Land, and Atmosphere from Space - Sensors, Systems, and Applications*, Progress in Astronautics and Aeronautics, AIAA, Vol. 97, pp. 71-95, 1985.

Pfeilsticker et al., First Atmospheric Profile Measurements of UV/visible O₄ Absorption Band Intensities: Implications for the Spectroscopy, and the Formation Enthalpy of the O₂-O₂ Dimer, *Geophys. Res. Lett.*, 28, 4595 - 4598, 2001

Rodgers, C. D., Retrieval of atmospheric temperature and composition from remote measurements of thermal radiation, *Rev. Geophys. Space Phys.*, 14, 609-624, 1976.

Roscoe, H.K.; Hill, J.G.T., Vertical resolution of oversampled limb-sounding measurements from satellites and aircraft. *Journal of Quantitative Spectroscopy and Radiative Transfer*, 72 (3). 237-248, 2002.

Traub, W.A, K.V. Chance, D.G. Johnson, and K.W. Jucks, Stratospheric spectroscopy with the far-infrared spectrometer (FIRS-2): overview and recent results, *SPIE*, 1491, 298, 1991

Van de Hulst, H. C., *Bull. Astron. Inst. Netherlands* 11 (410): 135, 1950.

Waters, J.W., J.C. Hardy, R.F. Jarnot, and H.M. Pickett, Chlorine monoxide radical, ozone, and hydrogen peroxide: stratospheric measurements by microwave limb sounding, *Science*, 14, 61-64, 1981.

Interactive comment on *Atmos. Meas. Tech. Discuss.*, 5, 7243, 2012.

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