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Atmospheric Measurement Techniques Discussions

Interactive comment on "Improved retrieval of nitrogen dioxide (NO₂) column densities by means of MKIV Brewer spectrophotometers" *by* H. Diémoz et al.

Anonymous Referee #1

Received and published: 19 August 2014

This paper describes an application of the updated Diémoz et al., 2013 algorithm to the Brewer #066 NO2 measurements. It presents a significant improvement over the standard NO2 algorithm ($\sim$ 200%) and I recommend publishing the paper after addressing the following questions:

1. The algorithm includes several changes compared to the standard algorithm. I believe the paper would benefit from addressing the effect of each change on the retrieved slant and vertical columns. This is important in light of application of the proposed algorithm to the historic Brewer data where no optimization of the grating position is done and only 5 wavelengths are used. Could you apply your algorithm to the 5 standard

wavelength positions? Can your algorithm be modified so the research groups without any radiative transfer calculation experience can use it? It appears that RT modeling of the polarized light accounting for Ring effect is the major limitation of the NO2 Brewer measurements in zenith direction. How different are the retrieved columns if NDACC recommended AMF are used? 2. Accuracy of the retrieval can be better demonstrated using climatological data reported by Gil et al., 2008. In general, more consistent comparison should be shown. Since all three geometries are analyzed all three should be compared (if possible) to the NDACC retrievals. It is also interesting to note that the NDACC reference instrument chosen for the intercomparison did not operate at the optimal for the intercomparison schedule. 3. Error analysis is done for the "best case" scenario: direct sun measurements over the unpolluted high altitude site assuming "constant" atmospheric conditions. It would be very beneficial to estimate the error for more polluted conditions and also for zenith sky measurements

Line-by-line comments:

p. 7370, line 8: 'but also to reprocess with higher accuracy the long-term raw datasets recorded at the existing measuring stations.' It is unclear how this can be done since the original algorithm uses five "standard" wavelengths versus proposed six wavelengths derived through an optimization process accounting for instrumental and atmospheric parameters (point 3, page 7372). Does the additional, 6th, wavelength really provide an additional degree of freedom compared to the standard algorithm?

p. 7372, point 2: Io-correction has a small effect on the NO2 column itself

p. 7372, point 3: How different is the optimized set of wavelengths for Brewer #066 compared to the standard wavelengths. Have you tried to apply your algorithm to the 5 standard wavelengths?

p. 7372, point 4: Do you ignore aerosol and cloud effect on scattering in your O2O2 RT modeling?

p. 7373, point 5: please explain how it is different from the “spectrally-flat factors” used in the standard algorithm.

p. 7373. Table 1 lists the new and standard cross sections used. Please specify temperature of the standard cross sections. What effect on the SCD do they have compared to the standard algorithm? I think you should add information on the updated cross sections as point 7 to “Compared to the standard algorithm, the new method brings several improvements:”

p. 7374, line 4: What does the “the same set of data” mean? Did you rotate the grating to a standard position to measure light intensity at the 5 standard wavelengths and then to the optimized position to measure 6 different wavelengths?

p. 7374, line 26: Could you please explain how atmospheric turbulence impacts the accuracy of the NO2 slant column measurements at AMF < 1.5 especially at the site located at 2400 m with mainly stratospheric NO2? Figure 7 shows that the random noise and wavelength shift combined with very small NO2 absorption are mainly responsible for the large errors at small SZA that would interfere with the Langley calibration.

I suggest moving discussion of the accuracy of the proposed algorithm results from Section 5.1 into 5.3, and focus solely on the intercomparison between SCD derived from the two algorithms. I think it will be very valuable to understand which of the improvements has the largest impact on the SCD.

p. 7376. Which of the three instruments described by Gil et al., 2008 was used for intercomparison? Two of them operate at SZA between 45o and 94o. Is it possible to compare VCD from the Brewer perpendicular polarization zenith measurements with the NDACC VCD measurements at all SZA not just twilight?

p. 7375, line 11: Based on Figure 1 a significant number of measurements are around 0 molecules/cm2 at small SZA. This might be an indication of calibration issues leading to underestimation of SCD.

p. 7368, line 24: ‘thus impacting on’ remove ‘on’

p. 7369, line 7: Please provide a more direct reference than “http://www.woudc.org/” to the information about the number of Brewers operated in NO2 measurement mode.

p. 7369, line 24. Please explain what you mean by “no on-site service exists to track the NO2 calibration of the Brewer instruments operating worldwide”. Do you mean radiometric/wavelength calibration at the measurement site or another instrument co-located with each Brewer?

p. 7377. The paper focuses on the NO2 algorithm from the Brewer. I think the only reason to include the discussion of the supplementary linear polarization and O2O2 products if you use them for cloud and aerosol screening in you NO2 algorithm. If this is not the case I would remove section 5.4 from the paper.

p. 7379, lines 6-7: Replace “surrounding conditions do not relevantly changes compared” for “atmospheric conditions remain constant compared”

p. 7380, line 13: “aerosol Ångström exponent” is mentioned for the first time here. Please explain how it is used in your algorithm earlier in the paper.

p. 7381, line 7: Could you please explain why do you expect “the relative uncertainty of the NO2 retrievals to drop for higher VCDs of nitrogen dioxide (i.e., more polluted conditions).” Under more polluted conditions your main assumption of the constant atmospheric conditions compared to calibration period is not met. In addition, the Langley calibration itself is much harder to do correctly. NO2 effective temperature over the polluted sites can be between 260 and 280K (in summer) compared to 220 $\pm$ 20K assumed in the analysis. Effect of the NO2 effective height on AMF also will be more significant at SZA > 80°.

It is also important to mention that direct sun measurements are not impacted by Ring effect and the AMF calculation is very simple at SZA < 80° and are not impacted much by the NO2 profile. Zenith sky measurements will have a larger total error, please give

an estimate of it.

Table 1. Please include (estimate) temperatures for the cross sections used in “standard” algorithm

Table 2. Please include “standard” wavelengths

Table 3. I recommend to remove Table 3

Figure 1. Please add 2012 to the graph. This graph will be more informative if diurnal variability for selected days is shown (VCD as a function of SZA over the course of the day) where one can see retrieved changes for a single day as a function of SZA. The horizontal lines are also confusing. It is stated that they are climatological morning and afternoon results. What SZA are they representative of? You also should add spread of the climatological data.

Figure 2. I find this figure somewhat confusing. While we can see increase in NO2 due to slow photolysis of N2O5 at AMF < 10 (zenith) and < 5 (direct sun). We do not see any increase in NO2 after sunset due to NO oxidation by O3, as well as no sharp decrease in VCD after sunrise due to NO2 photolysis. The VCDs between zenith AMF -/+ 35 behave almost linearly. Please explain. A figure with AMF for different geometries (zenith 2 polarizations, NDACC zenith recommended and direct sun) on the Y-axis and SZA on the X-axis will be very valuable.

Figure 3. Please add the results of the linear regression fit: slope, intercept and R2.

I would remove Figures 4 and 5 unless you are using them to show cloud screening algorithm for NO2.

Interactive comment on Atmos. Meas. Tech. Discuss., 7, 7367, 2014.