

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

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Received and published: 21 July 2014

The paper describes a new algorithm to retrieve NO₂ columns from a MKIV Brewer spectrometer. This algorithm reduces significantly the uncertainty compared to the standard algorithm. If such new algorithm could be applied to all existing MKIV Brewers, then a very useful, unique long term record of NO₂ columns could be obtained. In my opinion, this possible re-processing of historical datasets is the most important aspect of the paper. In order to improve the manuscript I suggest therefore a more detailed analysis of these key-questions:

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- Is the algorithm suggested in the paper the best choice?
- How does the uncertainty change between using the optimized wavelengths as in Brewer #66 and the standard wavelengths as in all the other MKIV Brewers?

On the other hand I suggest removing section 5.4 (O₂O₂ columns and degree of polarization from zenith sky data). These ‘supplementary products’ are very interesting and deserve their own paper, but do not really fit into this manuscript.

Is the algorithm suggested in the paper the best choice? The algorithm presented in the paper is a ‘Brewer type’ algorithm, where a weighted sum of the (log of the) measurements at each wavelength is used. The weights are determined to minimize the effects of Rayleigh scattering, aerosols, noise, and ozone absorption. However the weights chosen in this work do not minimize the effect of wavelength shift. As seen in figure 6, the wavelength shift and the noise are by far the dominant contributions to the total uncertainty. While the latter can be reduced by averaging data, the former is a more systematic effect and therefore the main driver for data accuracy. Directly after a wavelength calibration with the internal Hg-lamp, the wavelength uncertainty in the Brewer measurements is typically very small (~0.001nm). However it tends to increase with time, until it is ‘reset’ by another Hg-lamp routine. This gives an overall wavelength uncertainty of up to 0.02nm (the number used in the paper). Is it possible to include the wavelength shift in the weights, i.e. requiring the weighted sum over the $d\lambda/d\lambda$ to be zero? Is it possible that a spectral fitting algorithm, where several parameters (including the NO₂ slant column and the wavelength shift) are retrieved simultaneously from the data be more suitable? I do not expect the authors to develop other algorithms for comparison, but these questions should be addressed.

Uncertainty Line 202 states that ‘...Although the discrepancies are very low and far below the uncertainties of both instruments...’. This is not totally obvious to me. Section 6 is an excellent analysis of the uncertainty in the vertical NO₂ columns from direct sun measurements, but the data in section 5.3 are zenith sky measurements during twilight.

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I suggest that the uncertainty analysis in the manuscript should be done separately for the uncertainty in the slant columns and the uncertainty in the air mass factors. In this way it is valid for both, direct sun and zenith sky observations. If the values at the left side of figure 6 are the uncertainty of the slant columns and we assume an air mass factor of 10 or more for the twilight measurements, then the 0.02DU discrepancy for the data in section 5.3 is not 'far below the uncertainties of both instruments'. I also suggest that the comparison to the NDACC instrument (section 5.3) should be done at the basis of the slant columns. This would reduce the possible reasons for differences, since point 2 (line 207) and point 4 (line 217) would not apply in that case.

Data reprocessing I suggest extending the last paragraph of the paper by addressing the following questions: What steps are needed to reprocess the historic data of a MKIV Brewer? How does the 'non-optimized' grating position affect the data quality? Are individual weights for each Brewer needed? How well do the dispersion and the ND filter attenuation have to be known?

Minor comments

- The authors use the term 'air mass enhancement factor'. Isn't the standard way in literature just to call it 'air mass factor'?
- Line 7: 'with deviations of less than 0.02 DU', Add: 'in the vertical column amount from zenith sky data during twilight.' But a better way would be to analyze the uncertainty separately for slant columns and air mass factors as mentioned above.
- Line 7: 'easily implementable generalization...'. It should be noted that this technique is only 'easily implementable' at very clean sites with basically no tropospheric NO₂.
- Line 10: 'drift of nitrogen dioxide' -> 'drift of stratospheric nitrogen dioxide'
- Line 52: 'or the zenith sky' -> 'or the sky'
- Line 88: 'If the weighting coefficients are properly chosen, the last sum is cancelled out' -> 'is minimized'. Only for a given set of values for the other parameters (the ones

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used in the determination of the weights) it cancels out.

- Line 107: if W is the slit function, then equation 2 should have $W(\lambda-\lambda_0)$. If instead what is meant is the filter function (the 'flipped' slit function) than the integral can go over $W(\lambda_0-\lambda)$.
- Line 107: What a-priori value for X (NO₂) is being used?
- Table 2: I assume the old coefficients were calculated for the standard NO₂ grating position and the new weights with the optimized grating position. So can the old and new weights really be compared?
- Line 162: 'whilst atmospheric turbulence at midday is large'. Please explain in more detail. Do you have a reference?
- Line 170: 'agree well with both the expected stratospheric VCD for the clean site of Izana and the climatological values reported by Gil et al.': what is the difference between 'expected VCD' and 'climatological values'?
- Figure 3: the caption should say what the data are. I assume it is vertical NO₂ column amounts.
- Line 201: 'Deviations of 0.01–0.02 DU may be noticed between the series' What does this refer to? The difference between the data and the linear fit?
- Line 323: 'the accuracy of the measurements is expected to improve in more polluted conditions' I don't understand this. Do you refer to the relative uncertainty, which would decrease with higher column amounts?

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

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