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Interactive Comment

Interactive comment on "Retrieval of nitrogen dioxide stratospheric profiles from ground-based zenith-sky UV-visible observations: validation of the technique through correlative comparisons" *by* F. Hendrick et al.

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General Comment:

The authors are to be congratulated on a concise and well-written paper that sets out anew the compelling case for ground-based UV-visible practitioners to be inverting NO2 profiles from their data. The comprehensive validation and recalculation of how errors map on to the inverted profile should now allow us all to move forward with a standard error budget to which we can all refer. The determination of independent information (trace of A) and the eigenvector analyses are particularly important, and the authors are to be congratulated on a useful climatology of stratospheric NO2 profiles from satellite measurements and a careful assessment of their variability, which can be used for a priori profiles in the retrieval process.

Important specific comment:

However, this success has led to the exposure of what I believe is a conceptual mistake concerning the smoothing error, also to be found in our own earlier work (Preston et al. 1997). The mistake is now obvious because the variability of NO2 from the new climatology is much larger than our earlier guess. This results in a smoothing error that is at least 5 times the sum of all other errors, often over 10 times their sum (Hendrick et al. 2004, Figure 1). This surely cannot be correct.

The formalism for the covariance of the smoothing error on p2875 is correctly quoted as Ss = (A-I)Sx(A-I)T, where A is the matrix of averaging kernels and Sx is the covariance matrix of the true NO2 profile. The authors, like us, then equate Sx to the covariance matrix of the a priori profile, Sa. From the variability of satellite measurements, together with the correlation of values between adjacent altitudes, the authors have for the first time been able to derive an accurate value of Sa, where the diagonal elements are the squares of the variabilities and the off-diagonal elements are the correlations.

However, equating Sx to Sa cannot be correct, as the true profile cannot have a standard error as large as the variability the authors show from NO2 climatology, which is 50% at 25 km (Hendrick et al. 2004, Figure 1), even though Rodgers (2000) justifies it: ŞBecause the true state is not normally known, we cannot estimate the actual smoothing error... What is required is a description of the statistics of the error, which must be calculated from ... some appropriate ensemble of states, which may or may not be that described by ... Sa.Ť

Rodgers (2000) goes on to a more subtle discussion of the meaning of smoothing error: ŞMany remote observing systems cannot see spatial fine structure, the loss of which contributes to smoothing error. To estimate it correctly, the actual statistics of the fine

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structure must be known.Ť This argues that the smoothing error is not concerned with the variability of NO2 (the diagonal elements of Sa), only with size of fine structure, or wiggliness, of likely true profiles (the off-diagonal elements). If the diagonal elements of Sa were set to zero, might this then provide a better estimate of smoothing error Ss? The smoothing error in Figure 1 of the manuscript would certainly have more reasonable values.

Fortunately, this knotty philosophical problem can be bypassed. Rodgers (2000) exposes another conceptual mistake that we have all been making for many years: Şit may be better to abandon the estimation of the smoothing error, and consider the retrieval as an estimate of the smoothed version of the state, rather than an estimate of the complete version of the state.Ť In the manuscript, the validations compare retrieved ground-based profiles with smoothed balloon-borne and satellite profiles, so the retrieval is indeed considered an estimate of the smoothed version of the state. This is also true of most literature discussions of profiles from remotely-sensed measurements, necessarily so if the profile is shown at altitudes spaced by the vertical resolution of the retrieval. Is it appropriate in the manuscript and most of the literature to abandon the concept of smoothing error?

Other Specific comments:

1. In the validation exercise, the algorithm adopted for smoothing the high-resolution profiles to the lower resolution of the ground-based inversions (p2880 line5) is excellent if the high-resolution profile extends over a large altitude range. However, when its range is limited to 20 km or less, as in the Figures 6 and 8, the algorithm smoothes the high resolution profile inside its range with the a priori outside it. If the a priori is less than the high resolution profile, the smoothed profile is biased downwards, as is clear in the Figures. This cannot possibly be the smoothing imparted by the retrieval scheme to true high-resolution profiles, they do not stop at 30 km. One way to proceed would be to repeat the retrieval using the previous result as the a priori. Another would be to replace the a priori in the smoothing alogorithm by the retrieved profile. Best

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might be to extend the balloon or satellite profiles by a smooth and continuous function (e.g. cubic spline) that became equal to the a priori at, say, 40 km. Certainly the current result is unsatisfactory because in most cases it makes the comparison with ground-based data look better than it should be.

2. The authors determine the amount in the reference spectrum from the retrieval itself, justifying this as avoiding a Langley plot (p2871 line14). This is a circular argument, as the use of a Langley plot is identical to the addition of the extra parameter of amount in the reference into the retrieval, if the Langley plot uses the retrieved profile in the radiative transfer model when calculating air mass factors (AMFs). The advantage of the authorsŠ method is that it avoids systenmatic errors in the reference amount that could be introduced by a Langley plot using the a priori profile for calculating AMFs. The disadvantage is that prior Langley plots can derive intercepts for a whole data series, which can then be averaged to reduce random errors in reference amount. The best of both techniques would be achieved by averaging the reference amounts from the authorsŠ method over the whole data series, then re-retrieving with this average as a forced reference amount.

3. The authors introduce systematic errors in the measurements into the error budget (p2875 line22). But unless these are of a pseudo-random nature with Gaussian distribution, they cannot be lumped in with random measurement errors in this way. Many systematic errors have a rectangular distribution with different upper and lower limits - how could such a distribution be included here?

4. The number of independent pieces of information is determined from trace of A, the matrix of averaging kernels. The manuscript contains several examples of the dependence of its maximum value on various retrieval parameters, including a rather ambiguous statement about the upper limit of solar zenith angle (SZA) (p2878 line18). This is a critical point - could the authors please give a definitive statement or, better, a figure or table? Also important is the dependence of trace of A on the sampling - it presumably increases if the measurements go from intervals of 1° to 0.5° SZA, but

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does it further increase if they are at intervals of 0.25° SZA?

Technical comments:

A. The authors have an unusual notion of an ill-posed problem (p2872 line20). They state that there are more elements in the state vector x than in the measurement vector y. If true, the problem would be underdetermined, the equivalent of 3 simultaneous equations in 5 unknowns, and there is no unique solution. But this is not usually true in ground-based zenith sky measurements - the state vector should have few elements (6 altitudes between 5 and 45 km), whereas spectra are measured at intervals of 0.5° SZA over a range from 88° to 94° SZA (12 measurements). Hence the problem is overdetermined, and there would be conflict but for noise on the measurements, which if large enough also results in the lack of a unique solution. Either problem would usually be thought of as ill-posed.

B. The authors rightly set the tropospheric amount in the a priori profile to a negligibly small value (p2873 line3). But their explanation is only true if xa is defined as concentration (molec cm-2) not mixing ratio (ppbv). Although the Figures show that xa is indeed concentration, they are introduced later in the paper. For clarity, the definition should be given at this earlier point.

C. The authors beg many questions by introducing an Şideal observing systemŤ (p2874 line5), a phrase that could lead to many hours of debate. The system described is one where each measurement corresponds exactly to one element in the state vector, so inversion is unnecessary.

D. The description of the balloon-borne measurements in sections 7 and 9 as being Şsolar occultation techniquesŤ is rather odd considering that only measurements in ascent are used in the manuscript (occultation refers to the sun passing behind the earth, sunrise or sunset). Furthermore the ascent measurements could well be much finer than the 1 km vertical resolution quoted.

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E. The authors make repeated points about whether the satellite data is inverted with chemical correction or not, but do not say the likely size of the correction. In fact at the higher altitudes of the validation data it is small, almost certainly less than 1% at sunset (Roscoe & Pyle 1987).

F. The authors see a consistent seasonal pattern in the comparison of ground-based and POAM results (p2882 line11, and Figure 10), which my eye cannot see. They state the mean overestimation by POAM is 6% in spring and 12 % in summer, but in midsummer 1998 (June) the overestimation averages about 9%; and in spring 1999 (April) all values exceed 12%. Only the year 2000 exhibits anything like the pattern described in the manuscript.

G. In the caption to Figure 5, the use of ŞŞfitŤŤ is obscure and its quotes do not help; and the statement about error bars is ambiguous - is the full length 5e14 or are they ś5e14, and are they one or two-sigma?

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