Improved Air-Mass Factors for Ground-Based Total NO_2 Measurements: A Sensitivity Study

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Abstract

We present an overview of existing problems in air-mass factor calculations for NO_2 vertical column measurements by UV-visible ground-based spectrometry. Sensitivity studies made using different radiative transfer models allow us to identify and to quantify sources of uncertainties in calculations, and domiment effects are multiple scattering (4%) and profile variations due to diurnal (10 %) and seasonal changes (16%).

1. Introduction

Nitrogen dioxide (NO_2) plays a major role in photochemical processes controlling the abundance of atmospheric ozone. Ground-based zenith-sky UV-visible spectrometers measure twilight slant columns of ozone and NO_2. The accuracy of vertical columns is dependent critically on the quality of the Air-Mass Factor (AMF) used to convert the measured slant columns into vertical columns. Up to now, most groups use standard AMFs for NO_2 at all locations and seasons, introducing large uncertainties into the vertical columns.

For ozone, improvements have been made in the last years and most groups are oriented toward seasonal sets of ozone AMF for high, mid- and low latitudes because of its dependence to vertical profiles of density, aerosol (in case of volcanic loading in the stratosphere), and of course ozone itself. The spread between all AMF values is usually within 6-8% at 90° solar zenith angle (SZA) [1], [2], [3].

AMFs are usually computed by radiative transfer models. For NO_2 , a special computation must be made because, for example, of its diurnal variation, of its sensitivity to polarisation and temperature dependence of its cross-sections. A number of inputs must be studied in order to identify critical parameters that have to be improved for better accuracy of the data products. This activity is strongly supported by projects involved in data banks and satellite validation like Cose (Compilation of atmospheric Observations in support of Satellite measurements over Europe) and NDSC (Network for Detection of Stratospheric Changes) [4].

The goal of this paper is (i) to present an overview of existing problems related to NO_2 AMF computation and (ii) to carry a sensitivity study using different radiative transfer models.

2. Identified problems in NO_2 AMF computation

The list of identified problems is given below.

(i) Seasonal variation of NO_2, as for ozone AMF, needs a set of seasonal profiles, for the tropospheric and stratospheric parts of the profiles [2].

(ii) NO_2 diurnal variation is also an important parameter for NO_2 AMF computation. Then, the commun Langley-Bouguer plot (i.e. the line-of-sight amount versus AMF as a straight line) used to validate radiative transfer models and measurements, is not applicable to NO_2 and needs photochemical inputs to take into account NO_2 total column and profile diurnal variation or the use of an empirical AMF [8], [9], [10].

(iii) Chemical changes of NO_2 along the paths.
(iv) The temperature dependence of the NO_2 cross-sections gives an apparent seasonal variation similar to that of stratospheric temperature.
(v) Free tropospheric background NO_2, influence of polar stratospheric clouds and influence of boundary layer NO_2 pollution are effects dependent of the site and moreever sporadic.
(vi) Refraction is now widely integrated in models and the effect is well known.
(vii) Sky light polarisation has an effect on multiple scattering contribution on NO_2 as well as on ozone AMF and is not taken into acount in most radiative transfer models. Multiple scattering for clear sky and without polarition is identified.
(viii) Overcast weather condition and its variation is difficult to be taken into account.
(ix) Surface albedo

3. Intercomparison of radiative transfer models

The radiative transfer models used here have been compared previously for O_3 AMF computation. As a first step, AMF from four different radiative transfer models (see Table 1) were compared for the case of a pure Rayleigh atmosphere. Input profiles referenced in Table 2 were selected from Observatoire de Haute Provence (OHP) in France because of data availability at this NDSC primarily station. This test can be used to detect discrepancies between the models and problems with synchronising the input. In the past, similar tests have led to widely varying results, and a number of changes have been applied to the models to improve the consistency of the results. In Figure 1, the NO_2 airmass factors from the four models are plotted arbitrarily normalised to the SAOZ standard airmass factor. As can be seen, the differences between the models are of the order of 1% and even smaller below 90° SZA. From this result it was concluded, that no further model tests are necessary at this point.

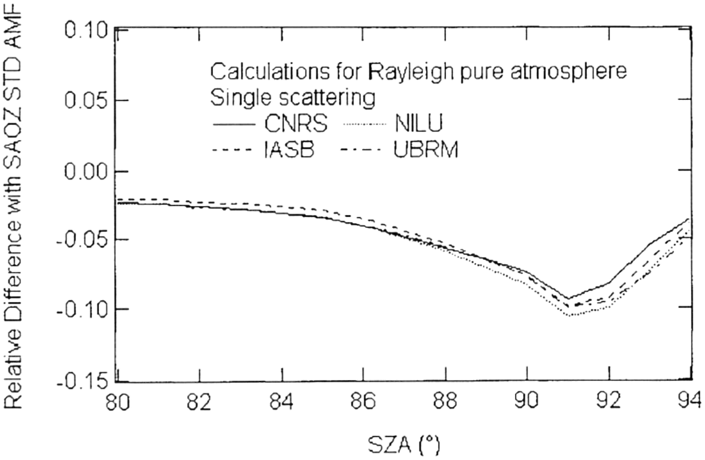

Figure 1. Normalised NO_2 airmass factors from all four radiative transfer models for a pure Rayleigh atmosphere. As can be seen, the differences are small at all SZA.

Laboratory	NILU	UBRM	IASB	CNRS
Model	DISORT [11]	GOMETRAN [12]	DISORT [11]	Flux Tracing [2]
Multiple Scattering Scheme	Discrete Ordinate	Discrete Ordinate	Discrete Ordinate	Numerical Integration

Table 1. Radiative transfer models used in the comparison.

Air Density	O3	NO2	T	Aerosol
OHP, 1 Jan 1996 See [2]	Handbook of Geophysics	OHP, 1 Jan 1996 See [2]	OHP, 1 Jan 1996 See [2]	Lidar OHP January 1996

Table 2. Input profiles for the comparison.

The objective of the second step of the intercomparison was to get an impression of the sensitivity of the NO_2 airmass factors changes in assumptions in the radiative transfer. Six different scenarios have been analysed with all models. The details of scenarios are given in Table 3, the resulting airmass factors, again normalised to the SAOZ standard airmass factor are shown in Figure 2.

SSC	SSA	MSC	MSA	OHP	STD
Single Scattering Calculations	Single Scattering with Aerosol	Multiple Scattering Calculations	Multiple Scattering with Aerosol	AMF used in June 1996 at OHP (SCUVS)	AMF used in the instrument in real time

Table 3. Identification of calculations schemes and legend to the plots in Figure 2. Airmass factors at Haute Provence (OHP) have been added as they have been used for the SCUVS intercomparison campaign at OHP in June 1996. They have to be compared to the improved airmass factors computed in this study using vertical profiles appropriate for this time and location.

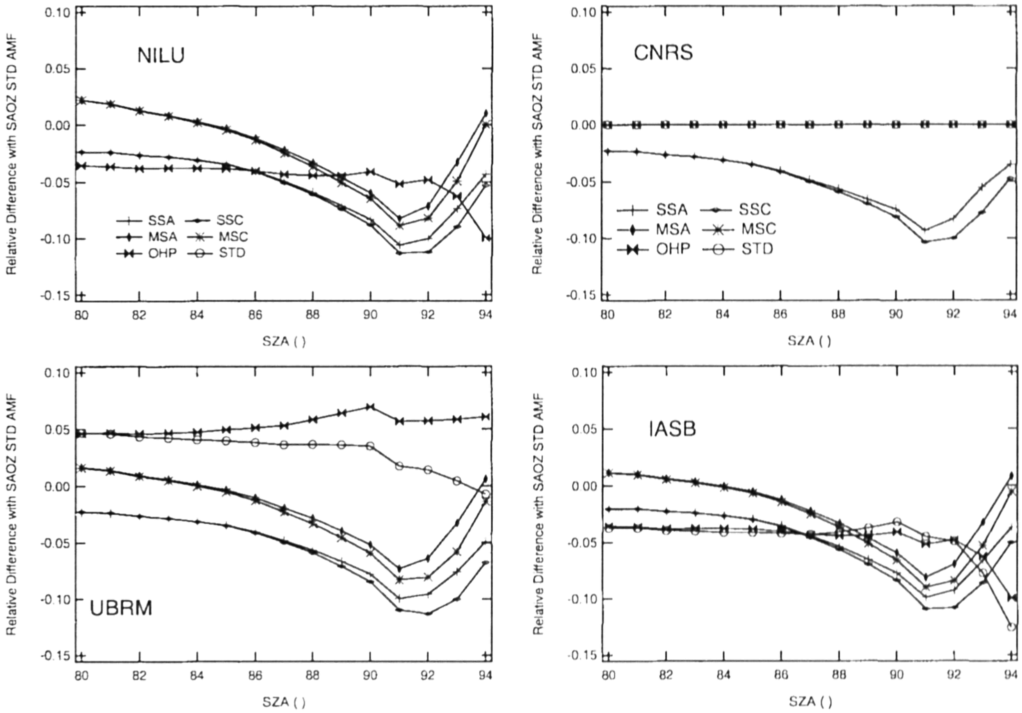

Figure 2. Influence of multiple scattering, stratospheric aerosols and profile assumptions. See Table 2 for details on the scenarios. The NILU data shown are for the SYMOCS instrument; the NILU SAOZ instrument uses the standard airmass factors.

Again, all models agree well for all scenarios. Inclusion of multiple scattering increases the airmass factors at all solar zenith angles by 4% , while accounting for aerosol scattering leads to increases at large solar zenith angles only. Changes in surface albedo were found to give a negligible effect (<1%) for twilight conditions but induce changes of up to 1.5% at high sun. All scenarios are significantly different from the SAOZ standard amf at 90°, emphasising the need for improvements in NO_2 AMF.

4. Conclusion

A sensitivity study on NO_2 AMF have been conducted in the frame of Cose activities. Results show that the spread of the NO_2 AMF at 90° SZA due seasonal variations of NO_2 profile (climatology) is 16 % , of diurnal variation is 10%, of multiple scattering 4%, of tropospheric pollution can be very large, of surface albedo and refraction is negligible. Other effects will be studied in part 2 of Cose project.

Aknowledgements

This work has been supported by the Programme de Chimie de l'Atmosphere (PNCA) in France and by the EC COSE (Compilation of atmospheric Observations in support of Satellite measurements over Europe ENV-CT98-0750) projects. Christine David has provided aerosol data from Observatoire de Haute Provence.

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