A climatology of NO_2 profile for improved Air Mass Factors for ground-based vertical column measurements

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Abstract

A new NO_2 profile reference atmosphere is built up from a climatological study of measurements from space, balloon and the ground, and complemented by modelling results. The reference atmosphere is used to investigate the sensitivity of zenith-sky NO_2 Air Mass Factors (AMF) to the vertical distribution of NO_2, pressure, temperature, and ozone. The study highlights periodic signatures in the AMF reflecting seasonal, latitudinal and sunrise/sunset changes of the atmospheric profiles, which need to be taken into account when retrieving NO_2 vertical columns from ground-based observations of zenith-scattered sunlight.

1. Introduction

Nitrogen dioxide (NO_2) plays a major role in photochemical processes controlling the abundance of atmospheric ozone. Within the framework of the Network for the Detection of Stratospheric Change (NDSC) [1,2], the vertical column abundance of sunrise and sunset NO_2 is monitored from pole to pole by a network of ground-based UV-visible spectrometers [3]. Slant column amounts derived from the measurement of the zenith-scattered sunlight at twilight are converted into vertical columns using an optical enhancement factor, or Air Mass Factor (AMF). AMFs are calculated with a radiative transfer model assuming the vertical distributions of NO_2 and of radiatively active atmospheric parameters. Current NO_2 AMFs are based on a basic reference atmosphere. Previous studies have highlighted the sensitivity of the AMF to the profile shape of NO_2 and of other atmospheric parameters [4,5]. In this study, an improved NO_2 reference atmosphere is built up and used to investigate possible periodic signatures in the AMF related to seasonal, latitudinal, and sunrise/sunset change of the vertical distribution of NO_2, pressure, temperature, and ozone.

2. Calculation of air mass factor

The enhancement in optical path length for an absorber through a scattering and absorbing medium is defined as the ratio of N_s, the integrated amount of absorber along the actual path of the sunlight, to N_v, the vertical column amount. Using the Beer-Lambert law, the AMF can be estimated through the calculation of the intensities I and I_o received by the ground-based spectrometer with and without the presence of absorber:

$$AMF = \frac{N_s}{N_v} = \frac{\ln\left(\frac{I_s}{I_{so}}\right)}{\ln\left(\frac{I_v}{I_{vo}}\right)} \qquad \text{(Eqn. 1)}$$

Hereafter, the intensities are calculated at 430 nm using both a full-spherical single scattering ray tracing model [5] and a pseudo-spherical adaptation of the multiple scattering Discrete Ordinate Radiative Transfer model [6]. Atmospheric parameters listed in Table 1 are assumed in a spherical atmosphere divided into 90 layers of 1-km thickness. The various databases take into account the seasonal and latitudinal variation of the atmospheric profiles.

Parameter	Description, source
NO_2	Composite climatological database described in this work and in [4]
Pressure, temperature, geopotential height	T: monthly means, derived from ECMWF analyses in [7] S/M: monthly means from the COSPAR Reference Atmosphere [9]
O_3	T: monthly means derived from ozonesondes and SBUV/SBUV-2 data [8] S/M: monthly means from the COSPAR Reference Atmosphere [9]
Aerosol	Climatological model of aerosol extinction derived from SAGE-II data [10]
Ground albedo	0.05 (ocean) and 0.75 (snow/ice)

Table 1. Radiative transfer parameters (T: troposphere; S/M: stratosphere/mesosphere).

3. Qualitative study of air mass factor sensitivity

The response of the AMF to a 50% NO_2 enhancement of 1-km thickness in a mid-latitude profile has been investigated as a function of the altitude and solar zenith angle (SZA). The AMF response is meaningful in two altitude ranges: between 20 km and 32 km, the perturbation causes the AMF to increase; for a perturbation located below 7 km, the AMF decreases. Accordingly, the present NO_2 profile database has been designed to reflect major periodic changes in stratospheric NO_2. It also includes a realistic tropospheric background representative of unpolluted NDSC sites. The behaviour of the AMF response can be explained as follows. At twilight, the scattering layer at 430 nm is located above the tropospheric NO_2 burden and moves upwards into the stratospheric burden as the SZA increases [11]. Hence, most of the zenith-scattered radiation received at the ground passes through the stratospheric absorbing layer with long slant paths but through the tropospheric absorbing layer with nearly vertical paths. The increase in absorption linked to a stratospheric perturbation is thus amplified in the slant optical path, yielding an increase of the AMF. As the altitude of the scattering layer increases with the SZA, the amplification of the response increases also with the SZA. On the opposite, the relative effect of a tropospheric perturbation is larger on the vertical column than on the slant column, decreasing the AMF.

4. Composite climatology of atmospheric NO_2

(i) Climatological model of stratospheric NO_2

A model has been designed to describe the seasonal, latitudinal and sunrise/sunset variation of stratospheric NO_2 on a climatological basis. It relies on sunrise and sunset profiles measured from 1995 through 1998 by the UARS Halogen Occultation Experiment (HALOE, [12]). HALOE NO_2 data version 19 cover altitudes spanning from above the stratopause down to 20 km. Data prior to 1995 are not considered here to avoid any perturbation of neither NO_2 nor its measurement due to Pinatubo aerosols. As shown in Plate 2 of Figure 1, the occultation measurement mode of HALOE results in a sparse temporal/latitudinal sampling. To improve the accuracy of the model, climatological characteristics of major stratospheric NO_2 variations are derived from long-term observations of total (stratospheric) NO_2 from a network of ground-based UV-visible spectrometers (Figure 1, Plate 1). The climatological characteristics determine the type of function to be used for the least-square fitting of low-pass filtered HALOE NO_2 profile time-series (Figure 1, Plate 2). The resulting climatological model provides stratospheric NO_2 as a function of latitude, altitude and time.

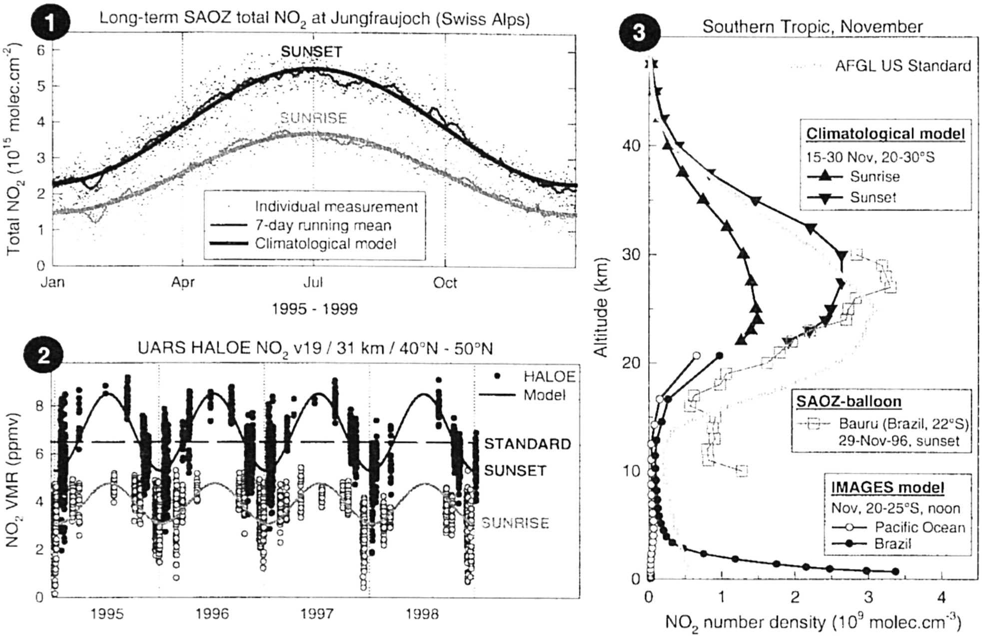

Figure 1. Construction of the composite NO_2 profile database: (1) ground-based determination of climatological characteristics of stratospheric NO_2; (2) at each altitude, fitting of HALOE data inspired from the ground-based characteristics; (3) use of IMAGES model results and SAOZ-balloon observations to complete the profile down to the ground. US Standard Atmosphere [16] values are depicted for comparison.

(ii) Extension through the troposphere

As illustrated in Plate 3 of Figure 1, the climatological model is extended down to the ground using monthly means of tropospheric NO_2 profile provided by the 3D chemical transport model of the global troposphere named Intermediate Model of Global Evolution of Species (IMAGES, [13]). NO_2 profiles measured by the SAOZ-balloon experiment [14] at middle and high northern latitudes in various seasons and in Brazil, provide useful information at altitudes where the accuracy of both HALOE data and IMAGES results might degrade. The resulting composite NO_2 climatology has been validated against ground-based UV-visible measurements of the NDSC, and HALOE and ERS-2 GOME [4,15] satellite overpass data.

5. Periodic signatures in the air mass factor – Conclusions

Figure 2 depicts the global picture of the seasonal, latitudinal and sunrise/sunset variation of the NO_2 AMF at 90° SZA while Figure 3 concentrates on the NDSC Alpine station at the Jungfraujoch. In general, the AMF based on the US Standard Atmosphere [16] would yield an underestimation of the retrieved vertical columns in the northern hemisphere and an overestimation at southern middle and high latitudes. The bias would exceed 10% in denoxification conditions in polar winter. The AMF exhibits a seasonal variation, which grows from a few percent in the tropics, up to about 5-6% at middle latitudes and 8-10% at high latitudes. The dawn-to-dusk photochemical variation of stratospheric NO_2 affects the AMF by a few percent, sunset AMFs being generally larger than sunrise AMFs.

To conclude, periodic signatures in the NO_2 AMF revealed by the present study demonstrate the need to reflect changes of the atmospheric profiles in order to improve the accuracy of NO_2 vertical columns retrieved from ground-based zenith-sky UV-visible measurement. The impact of tropospheric NO_2 on the AMF should be investigated further.

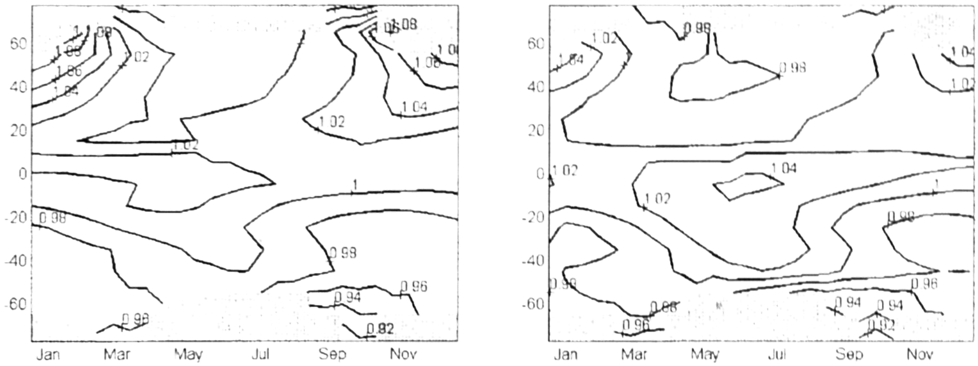

Figure 2. Seasonal, latitudinal, and sunrise/sunset variation of NO_2 air mass factors: ratio of NO_2 AMFs at 90°SZA calculated with the US Standard and with climatological NO_2 profiles, for a sunrise (left) and a sunset (right) stratosphere and a clean troposphere.

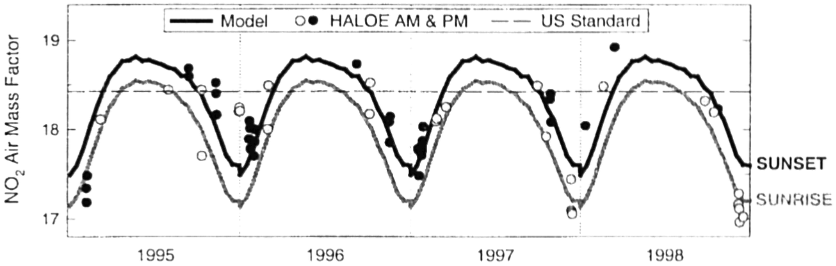

Figure 3. Seasonal and sunrise/sunset variation of the zenith-sky NO_2 air mass factor at 90° SZA calculated with: (i) individual overpass HALOE NO_2 data acquired within 500 km around the Jungfraujoch (Swiss Alps, 47°N); (ii) the present composite climatological model for 40°-50°N; and (iii) the US Standard atmosphere.

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