# Simultaneous retrieval of tropospheric aerosol extinction and $NO_2$ vertical profiles from MAXDOAS measurements in Beijing

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## ABSTRACT

We report on the retrieval of aerosol extinction and $NO_2$ profiles from ground-based multi-axis differential absorption spectroscopy (MAXDOAS) measurements performed in Beijing (N39°58'37" E116°22'51"), China. Measurements were made over a 10-month time period (June 2008 to April 2009) using a newly developed MAXDOAS instrument. A retrieval algorithm, based on the radiative transfer code LIDORT and the optimal estimation technique, has been designed to provide near real time information on aerosol extinction and $NO_2$ profiles. The algorithm was applied to measurements at two wavelengths (360 and 477nm). Comparison of the retrieved total aerosol optical depth (AOD) with values from a co-located CIMEL sunphotometer revealed a good correlation (R~0.8-0.9).

## 1. INTRODUCTION

China's rapid industrial development and the consequent large increase in energy consumption –primarily provided by coal-fired power plants– have resulted in an alarming deterioration of air quality. This increased air pollution is certainly problematic in megacities like Beijing, where the presence of elevated concentrations of pollutants such as, e.g., $NO_2$, $SO_2$, ozone and fine particles entails serious health risks for the population. Moreover, through long-range transport, air pollution in China has a potential impact on a global scale. Consequently, long-term ground-based measurements providing information on the total column and on the vertical distribution of tropospheric pollutants are indispensable. Such time series can be used to monitor the evolution of air pollution, to evaluate the direct effect of measures taken to improve air quality, and in addition, to validate air quality models and satellite observations.

Over the past decade, ground-based multi-axis differential absorption spectroscopy (MAXDOAS) has been shown to be a very promising tool for the automated retrieval of tropospheric pollutants. [1-4] MAXDOAS instruments are designed to perform quasi simultaneous observations of scattered sunlight for a range of different line-of-sight (LOS) directions from the horizon to the zenith, resulting in an increased sensitivity towards atmospheric absorbers such as $NO_2$ that are present in the lower troposphere. In addition, it has been demonstrated that aerosol information can be derived from differential absorption measurements of the oxygen collision complex $O_4$. [1, 2]

Here, we report on the application of a new retrieval algorithm developed for near real time automated retrieval of aerosol extinction and trace gas vertical profiles on MAXDOAS measurements made in Beijing during the June 2008 – April 2009 period.

## 2. MAXDOAS MEASUREMENTS

To monitor air quality and support the validation of satellite observations of tropospheric pollutants, a MAXDOAS instrument, designed and assembled at BIRA-IASB, was installed on the roof of the Institute of Atmospheric Physics (IAP) a few hundred meters from the Beijing Olympic stadium. The instrument was operated continuously from July 2008 to April 2009.

The BIRA-IASB MAXDOAS instrument is a dual-channel system. The first spectrometer covers the UV region from 300 to 390 nm, while the second extends from 400 nm to 720 nm. The optical head, mounted on a suntracker, can collect direct-sun and scattered light at various elevation and azimuth angles. During the Beijing campaign, the telescope pointed north for the MAXDOAS measurements. A full scan comprised 9 elevation angles (2°, 4°, 6°, 8°, 10°, 12°, 15°, 30°, zenith) and required approximately 15 minutes measurement time.

The measured spectra of scattered sunlight are analysed using the DOAS technique based on a least-squares spectral fitting method. [5] The direct results of this fitting are the differential slant column densities (DSCDs), i.e. the integrated concentrations of the absorbers along the effective light path of the scattered photons relative to the concentrations of the absorbers in a reference spectrum. For tropospheric profiling purposes, one can eliminate the stratospheric contribution to the measured DSCD by subtracting for each scan the DSCD measured at zenith from the off-axis DSCDs. [1] The $O_4$ and $NO_2$ DSCD, needed for the aerosol and $NO_2$ profile retrieval, are both retrieved twice, in the wavelength intervals 344.7-365 nm (UV-window) and 455-500 nm (VIS-window). These wavelength regions were optimized to obtain optimal sensitivity for $O_4$ and $NO_2$ while minimizing correlations with interfering absorption structures. In addition to the $NO_2$ and $O_4$ cross-sections (xs) other trace gas cross-sections such as $O_3$, $H_2O$, HCHO, BrO, were included, in the fitting routine, along with a Ring interference spectrum and a low-order polynomial closure term.

As a consistency check, we compared the $O_4$ DSCDs measured on clear-sky days with low aerosol pollution (AOD<0.15) with $O_4$ DSCDs simulated using a radiative transfer model with inputs taken from retrieved aerosol profiles and ancillary aerosol information. Under these conditions, the $O_4$ DSCDs at 30° elevation angle are rather insensitive to changes in the aerosol profile or other atmospheric parameters. [3] Therefore,

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a good agreement is expected between the measured and simulated $O_4$ DSCDs. We noticed, however, that the simulated $O_4$ DSCDs at 30° elevation were systematically lower by a factor of about 0.8. Possibly, the differences are induced during the DOAS retrieval, maybe because of errors in the absolute values of the $O_4$ xs. To account for this, we applied a correction ($xs_{corrected}=xs_{Hermans}\times1.25$) to the absolute value of the $O_4$ cross-section, for both the UV and the VIS windows. A similar correction factor has been previously reported by Wagner et al. [3]

## 3. PROFILE INVERSION

We have developed a dedicated two-step inversion algorithm. First, aerosol extinction profiles are retrieved at different wavelengths. In the second step, the derived aerosol information is used as input to retrieve tropospheric trace gas profiles. In this manner, the dependence of the effective light path to the presence of aerosol in the atmosphere is properly taken into account. [1] For this first analysis of the measurements in Beijing we handled separately the DSCDs obtained in the two wavelength intervals. In this section, we give a brief description of the algorithm and discuss specific settings used for the profile inversion from MAXDOAS measurements made in Beijing.

### 3.1 General algorithm description

In the first step, the aerosol extinction vertical profile is obtained by combining the retrieved $O_4$ DSCDs for different LOS directions. In general, the length of the light path through the atmosphere and thus the observed DSCD of an atmospheric absorber depend not only on the concentration of the trace gas but also on the vertical distribution and optical properties of the aerosol present in the atmosphere. Consequently, when the vertical distribution of an absorber is well known and nearly constant –the $O_4$ concentration varies with the square of the $O_2$ monomer– DSCD measurements provide information on the aerosol optical properties. Once the aerosol extinction profiles are retrieved, the change in the observed trace gas DSCD caused by the presence of aerosol can be accounted for, and the tropospheric trace gas profiles can be obtained from the measurements. [1]

For the inversion we used the optimal estimation method. [6] Herein, the aerosol extinction vertical profile **x** is retrieved given an *a priori* profile $\mathbf{x_a}$, the measurements **y** (here, a set of DSCDs for different LOS directions), their respective uncertainty covariance matrices ($\mathbf{S_a}$ and $\mathbf{S}_\varepsilon$), the matrix **K** of weighting functions, and a forward model operator **F** usually implemented as a numerical model. Aerosol extinction profile retrieval is a non-linear problem, and the inverse solution has to be determined iteratively:

$$x_{i+1} = x_i + \left(S_a^{-1} + K_i^T S_\varepsilon^{-1} K_i\right)^{-1} \cdot \left[K_i^T S_\varepsilon^{-1}\left(y - F(x_i)\right) - S_a^{-1}\left(x_i - x_a\right)\right] \quad (1)$$

For the trace gas vertical profile inversion a single inversion step is sufficient as **K** is independent of the **x**. [6] The weighting functions display the sensitivity of the measurements to changes in the vertical profile. The forward model **F** describes the physics of the measurement, and in our application is based on the linearized discrete ordinate radiative transfer model (LIDORT v3.3). [7] A major advantage of this code is that it includes an analytical calculation of the weighting functions needed for the inversion step. Consequently the algorithm is relatively fast, which is a major advantage since it allows for near real time automated retrievals An important parameter for the characterization of the retrieval is the sensitivity of the retrieved state to the true state [6]. The averaging kernels – which are the rows of the averaging kernel matrix **A**– express this relationship. They provide a measure for the vertical resolution of the measurement and the sensitivity of the retrieval to the true state at particular altitudes. The trace of **A** is the Degrees of Freedom of Signal (DFS), a measure of the number of independ-

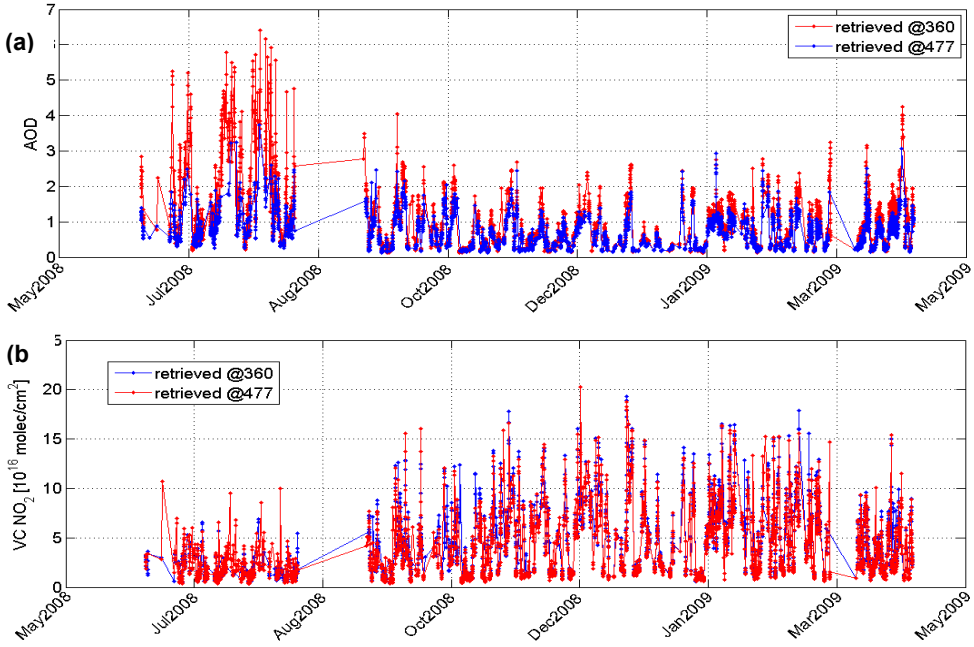


*Figure1 : Time series of the retrieved tropospheric (a) AODs and (b) $NO_2$ VCs from MAXDOAS measurements at 360 nm (red) and 477 nm made in Beijing in Beijing from June 2008 to April 2009.*

ent pieces of information extracted from the retrieval.

### 3.2 Retrieval parameter settings

With optimal estimation, the choice of the *a priori* profile and the error covariance matrices has an important impact on the results. There is a trade-off between maximizing the DFS and eliminating the occurrence of spurious oscillations in the retrieved profiles.

For the retrieval of aerosol extinction profiles, we assumed that the correction factor applied on the measured $O_4$ DSCD eliminates all systematic errors on the measurements. In this way, $\mathbf{S\varepsilon}$ is then a diagonal matrix, with variances equal to the square of the DOAS fitting error. A fixed *a priori* profile (taken from the LOWTRAN climatology) with an AOD of 0.1 was chosen. To allow for the large variations in the AOD (0.1 to 5 at 360 nm) and avoid unwanted oscillations, the $\mathbf{S_a}$ was constructed as follows: The diagonal element corresponding to the lowest layer, $S_a(1,1)$, is set equal to the square of 0.2 times the maximum partial column of $\mathbf{x_i}$. The other diagonal elements decrease linearly with altitude up to $0.2\times S_a(1,1)$. The non-diagonal terms, introducing a correlation of the profile at different altitudes, were added as Gaussian functions, see Ref. 8, with a correlation length of 0.05 km. Using these settings, the mean DFS for the aerosol extinction profile retrievals was about 1.9.

The $O_4$ DSCDs calculated using the forward model not only depend on the aerosol extinction profile but also on aerosol single scattering albedo and phase function. To obtain a good estimate of these optical aerosol parameters, we used the inversion products (size distribution and refractive index) reported on the AERONET website [9] from a CIMEL sunphotometer located next to the MAXDOAS instrument in Beijing. The AERONET AODs were used for a first validation of the aerosol extinction profiles retrieved from the MAXDOAS measurements.

For the $NO_2$ profile inversion, the $\mathbf{S_\varepsilon}$ was constructed in a similar way as that for the aerosol profile retrieval. To account for the large variability in the $NO_2$ tropospheric column densities (VCs) we used the "geometrical approach" to obtain a first estimate. In this approach the DSCDs measured at an elevation of 30° are converted into VCs using simple geometrical considerations based on the following equation [10]:

$$VC_{tropo} = \frac{DSCD(30°) - DSCD(zenith)}{AMF(30°) - AMF(zenith)} = \frac{DSCD}{\sin^{-1}(LOS) - 1} \quad (3)$$

This first approximation assumes that the $NO_2$ layer is located below the scattering altitude, and it neglects the effect of the presence of aerosol on the air mass factor (AMF). The *a priori* profile is chosen to be an exponential function with a scaling height of 0.5 km and a total column equal to the VC obtained using this "geometrical approach". The diagonal elements for each layer are equal to 0.1 times the partial *a priori* column. For the non-diagonal elements, Gaussian functions with a correlation length of 0.2 km were used. The mean DFS thus obtained was 2.2.

## 4. RESULTS AND DISCUSSION

Aerosol extinction and $NO_2$ profiles were retrieved from MAXDOAS spectra measured in Beijing during

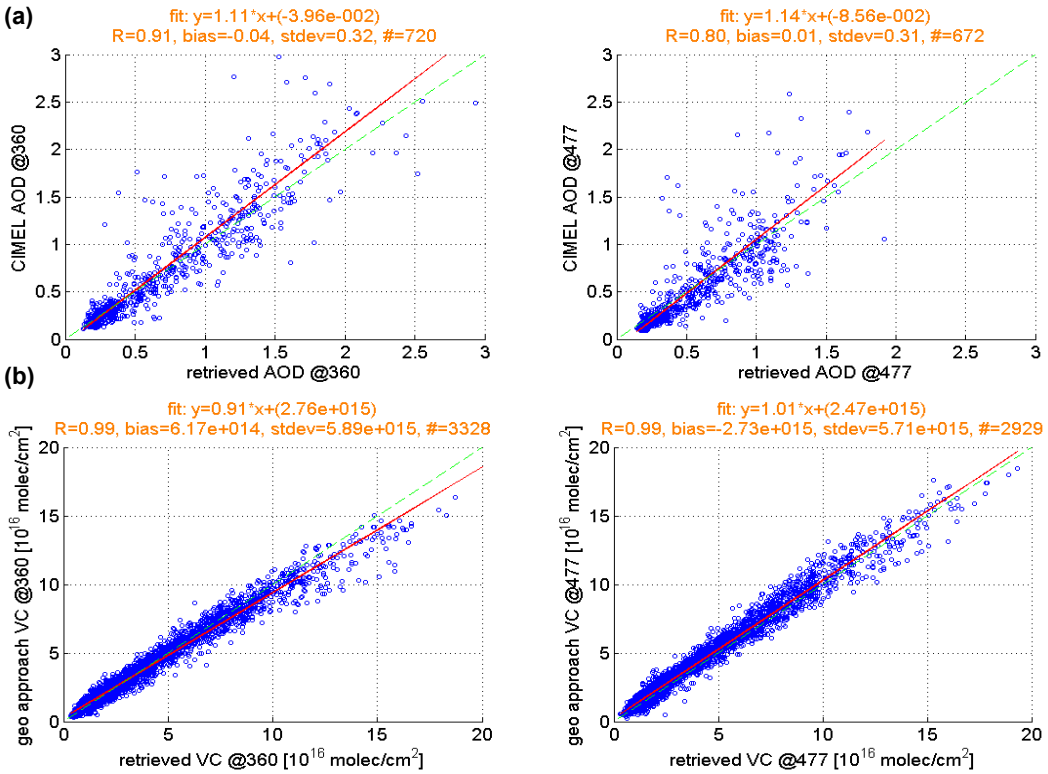


*Figure2 : Scatter plots of (a) the AODs at 360 and 477 nm retrieved from the MAXDOAS measurements versus the AODs from a co-located CIMEL sunphotometer and (b) the tropospheric $NO_2$ VCs retrieved using the profiling tool versus the VCs obtained using the "geometrical approach". Also shown are the linear regression fits and the corresponding statistical parameters (correlation coefficient R, bias, standard deviation (stdev), number of points (#)).*

the period from June 2008 until April 2009 for 2 wavelengths (360 and 477 nm). Only those profiles are retained for which the measured and simulated DSCDs agree to within 10%.

The time series of quality-checked AODs and tropospheric $NO_2$ VCs retrieved using the profiling tool at 360 and 477nm are displayed in Fig. 1. The AODs and tropospheric $NO_2$ VCs were calculated from the corresponding retrieved vertical profiles. From the AOD time series, it can be seen that during June and July 2008 the observed AODs are relatively high and display strong variation. The tropospheric $NO_2$ VCs, on the other hand, are relatively low during the June-July 2008 period. The latter result is in agreement with satellite observations and model output. [11] The period of low observed tropospheric $NO_2$ VCs coincides with the 2008 Beijing Olympic Games during which measures were taken by the Chinese government to reduce air pollution. Due to instrumental problems, no MAXDOAS measurements were taken between the 21st of August and the 16th of September 2008.

To obtain an indication of the quality of the retrievals, we compared hourly averages of the retrieved AODs with AODs measured by a co-located CIMEL sunphotometer. Figure 2a displays scatter plots for the two wavelengths. A very good correlation is obtained between our retrieved AODs and those of measured by the CIMEL instrument. The correlation coefficients (R) are 0.9 and 0.8 for the retrieval at 360 and 477 nm, respectively. It is interesting to note that the good agreement is obtained not only for those situations where the AOD is close to the *a priori* value, but also in situations where a high aerosol load has been successfully retrieved.

Tropospheric $NO_2$ VCs were compared with *a priori* VCs obtained from the $NO_2$ DSCDs measured at 30° elevation using the "geometrical approach", see Fig. 2b. The tropospheric columns correlate well. This indicates that for these conditions, the assumption made concerning the scattering altitude is valid and that consequently the geometrical approach can be used to obtain a good approximation of the tropospheric total VCs. Differences are most probably related to the influence of the aerosol, which is not taken into account in the geometrical approach.

## 5. SUMMARY AND OUTLOOK

We have presented a newly-developed algorithm for near real time simultaneous retrieval of aerosol extinction and $NO_2$ vertical profiles. We have outlined a specific approach for the successful automated retrieval of profiles for locations (such as Beijing) where a large variability of AODs and $NO_2$ VCs is apparent.

Results were presented for MAXDOAS measurements made in Beijing from June 2008 to April 2009. Retrieved AODs at 360 and 477nm were compared to AODs from a co-located CIMEL sunphotometer. Good correlations (0.8-0.9) and acceptable standard deviations (~0.3) were obtained.

These first results are very encouraging and strengthen the belief that MAXDOAS measurements can be used to provide high-quality long-term time series of aerosol optical properties and trace gas vertical profiles.

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