MEASUREMENT OF ATMOSPHERIC POLLUTANTS BY MEANS OF
DIFFERENTIAL OPTICAL ABSORPTION SPECTROSCOPY (DOAS)
WITH A PC PROJECTOR LIGHT SOURCE

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Abstract

The conventional point sampling of pollution species at ground stations leads to concentrations for local environments. In order to evaluate average pollution conditions, it is valuable to obtain additional information of regional concentrations measured over a certain distance, e.g., several hundred meters to several kilometers. Differential Optical Absorption Spectroscopy (DOAS) based on a continuous or pulsed light source provides a useful tool for monitoring pollutant trace gases and aerosol particles in the lower atmospheric boundary layer.

In this paper we report the results of three recent campaigns: Seoul city in South Korea (August 2007), Chiba city (November 2007) and Nagano city (August 2008) in Japan. By means of the DOAS and average concentrations of NO2 and aerosol particles in the troposphere have been measured. The long-path, continuous measurements have actually been performed, and the resulting concentrations of pollutants are compared with the data from nearby ground-based monitoring stations. We measured optical thickness due to NO2 absorption and aerosol extinction in the lower troposphere (atmospheric boundary layer) using nearly horizontal optical paths in a height range of 20–130 m from the ground level.

Key words: DOAS, PC projector flashlight, air pollution, urban

1. Introduction

In recent years, the atmospheric pollution in Japan, as a whole, has been improved as compared with situations couple of decades ago. However, we still have problems in places such as urban roadside areas, where the environmental standards have not been achieved. In this respect, efforts are required for monitoring anthropogenic air pollution, especially the combustion products such as nitrogen oxides and particulate matters.

The main pollution species in urban areas in Japan is the nitrogen dioxide (NO2) and suspended particulate matter (SPM), usually referred to as PM10. In the monitoring of such species, conventional point sampling at ground stations leads to concentrations for local environments. It is also valuable to obtain additional information of regional concentrations measured over a certain distance, e.g., several hundred meters to several kilometers. Differential optical absorption spectroscopy (DOAS) in the visible and near-UV region is more suitable to monitor horizontally averaged concentrations of pollutants (Edner et al., 1993).

In the conventional long-path DOAS method, a continuously emitting light source is employed, and the source (or occasionally a retroreflector) is placed at a certain distance from the observation site. The use of aviation obstruction lights makes it possible to employ a simple detection system that consists of a telescope and a compact CCD spectrometer. Our group in Center for Environmental Remote Sensing, Chiba University (CEReS) reported a novel DOAS method that is based on a white flashlight source and a compact CCD spectrometer (Yoshii et al., 2003, Fuqi et al., 2005). In Japan, it is mandatory for tall constructions (higher than 60 m) to operate highly illuminant (more than 2×10^6 cd) white flashlights during the daytime that are detectable in every direction from several kilometers away.

The purpose of this study is to demonstrate the capability of the PC projector as a DOAS light source. The long-path, continuous measurements have actually been performed, and the resulting concentrations of pollutants are compared with the data from nearby ground-based monitoring stations.

2. Study area and Experiment

2.1. Study area

We report the results of three recent campaigns: Seoul city in South Korea (August 2007), Chiba city (December 2007) and Nagano city (August 2008) in Japan. Seoul city has the area of about 605.33 km² (2006) (nearly the same as the Tokyo area) with the population of about 10.35 million (2006). The restoration of the Cheong-gye stream in the midtown has contributed to alleviate the urban pollution situations. The environmental standards of NO2 and SPM have been mostly attained in Nagano city, while heavy traffic still causes problems in the urban Chiba city area. In these measurements, we measured optical thickness due to NO2 absorption and aerosol extinction in the lower troposphere (atmospheric boundary layer) using nearly horizontal optical paths in a height range of 15 – 100 m from the ground level.
2.2. Experimental method

An astronomical telescope (Meade, DS-115), with an aperture diameter of 115 mm and a focal length of 910 mm, is employed to focus the image of a point light source located at a far distance. The image is formed near the eyepiece location (the eyepiece itself is removed from the telescope) where the entrance slit (1 mm high and 5 μm wide) of a CCD spectrometer (Ocean Optics, USB2000) is placed. The CCD consists of 2048 elements and is sensitive in a wavelength range of 200-800 nm, resulting in an average resolution of 0.3 nm/pixel. This CCD spectrometer is composed of a fixed grating and a linear CCD array with a mechanically stable, crossed Czerny-Turner design. No moving parts are incorporated, resulting in high reliability and compactness (89 mm wide × 63 mm deep × 34 mm high). The CCD gate duration is set at 300 ms in the experiment. Between successive gate periods, there exists a time lag of 7 ms, in which each spectral data is sent to a personal computer (PC) through the universal serial bus. The data acquisition can be attained successfully even when no trigger (synchronous with the flashlight) is applied to the CCD spectrometer, though this relatively long gate time as compared with the flashlight duration causes somewhat increased amount of the background skylight.

We automatically achieved the discrimination of data with and without the flash event by comparing the light intensity integrated over a wavelength region of 400 – 450 nm, used for the present NO2 detection. The spectral difference between the flash and the no-flash events exhibits the net strobe intensity after the long-path transmission in the urban atmosphere. Fig.1 shows optical thickness of each component detected by the DOAS spectrometer. The absorption of NO2 gas species gives a structure of the order of 0.2 in terms of the optical thickness. In order to extract the NO2 concentration, contributions from molecules and aerosol particles can be subtracted by simply applying a linear fit to the background. Another important aspect from this figure is that, by measuring the light intensity, it becomes feasible to evaluate the aerosol contribution in the DOAS signal itself.

![Optical thickness of each component detected by the DOAS spectrometer](image)

2.3. NO2 and aerosol retrieval

The analysis of the DOAS spectra is based on the Beer–Lambert’s law expressed as

$$I(\lambda) = kI_0(\lambda)e^{-L\sigma(\lambda)n}$$

Where $I(\lambda)$ is the measured intensity, $k$ is the system constant, $I_0(\lambda)$ is the unattenuated reference intensity, $L$ is the path length, $\sigma(\lambda)$ the wavelength-dependent absorption cross section, and $n$ is the number density of the species averaged over the path length. The dimensionless quantity $Lc(\lambda)n$ represents the optical thickness, denoted as $\tau$.

Here we describe the algorithm developed for the retrieval of both the NO2 concentration and the aerosol optical thickness. After the background subtraction, the observed light intensity $I_{obs}(\lambda)$ can be expressed as

$$I_{obs}(\lambda) = kI_0(\lambda)T_m(\lambda)T_a(\lambda)T_{NO2}(\lambda)$$

Here $k$ is an empirically determined coefficient, $I_0(\lambda)$ is the spectrum of light source observed at a location close to the light source, $T_m(\lambda) = \exp[-\tau_m(\lambda)]$ is the transmittance of air molecules Rayleigh scattering [$\tau_m(\lambda)$ is the molecular optical thickness ], $T_a(\lambda) = \exp[-\tau_a(\lambda)]$ is the transmittance of air particles Mie scattering [$\tau_a(\lambda)$ is the aerosol optical thickness ], and $T_{NO2}(\lambda)$ is the transmittance representing the NO2 absorption. In the present algorithm involving both the NO2 and the aerosol retrieval, we first correct the observed spectrum $I_{obs}(\lambda)$ for the first three factors on the right-hand side of Eq.2:

$$I_{obs}^*(\lambda) = \frac{I_{obs}(\lambda)}{kI_0(\lambda)T_m(\lambda)}$$

Combining Eqs. (2) and (3), we obtain
\[ I_{\text{obs}}'(\lambda) = \exp[-\tau_o(\lambda) - \sigma(\lambda)C] \]

where \( C = nL \) is the column amount of NO\textsubscript{2}. Moreover, if it is assumed that the aerosol optical thickness exhibits wavelength dependence as given by the Angstrom exponent, we obtain

\[ -\ln I_{\text{obs}}'(\lambda) = B\lambda^{-A} + \sigma(\lambda)C \]

Removing the slowly varying contribution from Eq.(5), and applying the spectral matching to the rapidly varying part with the laboratory cross-sectional data, we obtain the value of \( C \). Then, substituting \( C \) into Eq.(5) leads to the value of \( B\lambda^{-A} \). In the actual data analysis, this process has to be operated in an iterative way.

The drawback of the use of aviation obstruction light as a DOAS source is that in accordance with the regulation, the light intensity is diminished at dusk and dawn, and during the nighttime, blinking red lights replace the flashlights. Thus, the DOAS measurement is limited to the daytime. Moreover, the measurement cannot be carried out where no obstruction flashlight is situated. Alternatively, here we propose the use of a commercially available PC projector as a white-light source. This is relatively inexpensive, yet the possibility of unattended, continuous operation is quite suitable for the DOAS measurement. This light source is portable, and it can be used during both daytime and night time.

3. Result

3.1. DOAS measurement with a projector flashlight in the restoration zone, Seoul

The lamp height is about 130 m above the ground level. According to the regulation, the light intensity is diminished at dusk and dawn, and during the nighttime blinking red lights replace the flashlights. Thus, the DOAS measurement is limited to the daytime, around 5 a.m. to 7 p.m. during the summer. For the measurement in Seoul, we made use of projector as a light source. The lamp height was about 70 m above the ground level. The DOAS measurement with the light source of projector was used during the daytime to nighttime. In the east direction 1.4 km from the source, a DOAS system was installed in a Cheong-gye stream at Cheong-gye-3 ga.

Power consumption of the projector is 220 W, and the emission covers the visible wavelength. For the observations in Seoul measurement parameters were set up at about 30 m height. The projector flashlight was set up at about 70 m height. The projector flashlight was used as a light source and the distance of the light source to observation point was 1.4 km. The ground data were obtained every hour. The observed values of NO\textsubscript{2} ranged from 30 to 60 ppb, and a reasonable agreement between the two methods suggests the well-mixed condition of pollutants in the lower atmospheric boundary layer (Fig. 2).

![Fig. 2 The DOAS measurement in comparison with the ground sampling data in Seoul](image)

3.2. DOAS measurement with a projector flashlight in Chiba and Nagano

For the measurement in Chiba and Nagano, we made use of projector as a light source during the daytime to nighttime. The projector flashlight was used as a light source and the distance of the light source to the observation point was 2.7 km. The projector flashlight was placed at about 30 m height in Chiba University. Power consumption of the projector is 220 W, and the emission covers the visible wavelength. For the observation in Chiba measurement was set up at about 90 m height. The DOAS method allows to acquire data every five minutes, which is much more frequent than the data (every one hour) provided from the Ministry of the Environment Atmospheric Environmental Regional Observation System (AEROS). Therefore, the data observed by DOAS and Ground system data (Fig. 3, 4, 5). A reasonable temporal correlation was found between the result of the long-path measurement using DOAS method and the data of NO\textsubscript{2} concentration from the ground station near each optical path in Chiba city. Moreover, a reasonable correlation was found between the aerosol optical thickness using DOAS method and the SPM concentration observed from a ground station in these cities.
Fig. 3  The DOAS measurement in comparison with the ground sampling data in Chiba

Fig. 4  Comparison between aerosol optical thickness and SPM concentration in Chiba

Fig. 5  The DOAS measurement in comparison with the ground sampling data in Nagano

4. Conclusions

Although ground sampling stations provide hourly data on urban air pollutants, the DOAS technique yields highly frequent data (about 5 min) averaged over a relatively long path of a few kilometers. A conventional PC projector was successfully used as a DOAS light source, making it possible to observe the data for 24 hours. By focusing on the light intensity received by the detector, we can employ the DOAS data to retrieve the aerosol concentration along the horizontal light path.

References

