Method to Monitor Atmospheric Atomic Mercury by a Differential Absorption Lidar System

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Abstract: A differential absorption lidar (DIAL) system has been developed for atomic mercury concentration monitoring and applied in ambient air monitoring in Guangzhou, China. A 24-hour measurement on background atomic mercury has been performed.
OCIS codes: Remote sensing and sensors(280.0280); DIAL, differential absorption lidar (280.1910)

1. Introduction

Remote-sensing techniques have been widely used for atmospheric studies, and light detection and ranging (lidar) is particularly useful. The differential absorption lidar (DIAL) variety can be used to monitor trace constituents and to measure gas concentrations range-resolved, and map the flux over large distances [1-4].

Mercury is a severe environmental pollutant related to coal combustion, mining and industrial activities [5-7]. It has well-known and severe adverse effects on humans. Almost all atmospheric mercury occurs in atomic form which poses both special challenges and possibilities. Apart from the pollution side there are also other extremely interesting aspects of atomic mercury. With its very high vapor pressure already at room temperature it constitutes an important geophysical tracer gas [8]. So atomic mercury concentration and flux monitoring is very important both in environmental analysis and geophysical research.

In this paper, we describe mercury monitoring by using a newly developed different absorption lidar system based on a Nd:YAG laser pumped narrow-band dye laser. The basic principle utilized in DIAL is that the gas to be detected has a known absorption “fingerprint”. Pulses at two different wavelengths are transmitted intermittently, one of which is absorbed by the gas of interest (on-wavelength), and the second one is very close to the on-wavelength, but is not absorbed or with weaker absorption (off-wavelength). The gas concentration is then retrieved from the slope of the ratio between the backscattering light intensities from the on- and off-wavelength pulses. The ratio between the on- and off-wavelength backscattering signals is given by [9]:

$$\frac{I(\lambda_{on},R)}{I(\lambda_{off},R)} = \exp\left\{-2\int_0^R N(r)\left[\sigma(\lambda_{on})-\sigma(\lambda_{off})\right]dr\right\} \qquad (1)$$

Here, $I(\lambda_{on},R)$ and $I(\lambda_{off},R)$ are the backscattering light intensities for the on- and off-wavelength, respectively, while $\sigma(\lambda_{on})$ and $\sigma(\lambda_{off})$ are the corresponding absorption cross-sections. $N(r)$ is the gas concentration at distance r, and R is the detection distance.

2. Method

The DIAL system used in the measurements has been thoroughly described in [10]. Here only a brief introduction is given. The system, consisting of laser system, telescope system, detection unit, data acquisition and system controlling unit, is illustrated in Fig. 1. A Nd:YAG laser (Spectra-Physics, Quanta-Ray PRO-290-30EH) pumped dye laser (Sirah, DSCAN-D-18) with function of double wavelength switching (20 Hz) is used in the DIAL system. A Newtonian telescope equipped with folding mirror controlled by a stepping motor servo system serves for light transmission and collection. The light signal gathered by the telescope is first detected by a photomultiplier tube, (Hamamatsu, R7400) and is then fed to a transient digitizer (Licel, TR 40-160). The sampled data are transferred to the computer via a TCP/IP protocol for analysis. The DIAL system is controlled by a LabVIEW-based program, including equipment controlling and data analysis. A delay generator (Quantum, 9528) is used to synchronize all the equipment during the DIAL measurements. Light from the laser passes a reflector and an expander, and propagates to the atmosphere. The backscattered light gathered by the telescope is focused on a pinhole which blocks out sky radiation. Then the light goes through a lens and becomes parallel. A narrow-band interference filter isolates the backscattered laser light and blocks out sky radiation.

For mercury monitoring, the on- and off- vacuum wavelengths used for the mercury DIAL measurements are 253.729 nm and 253.744 nm, respectively, the conversion frequency between on- and off- wavelength is 20 Hz. The peak value of the atomic mercury absorption cross-section is found to be 2.5×10^{-14} cm^2•$atom^{-1}$.

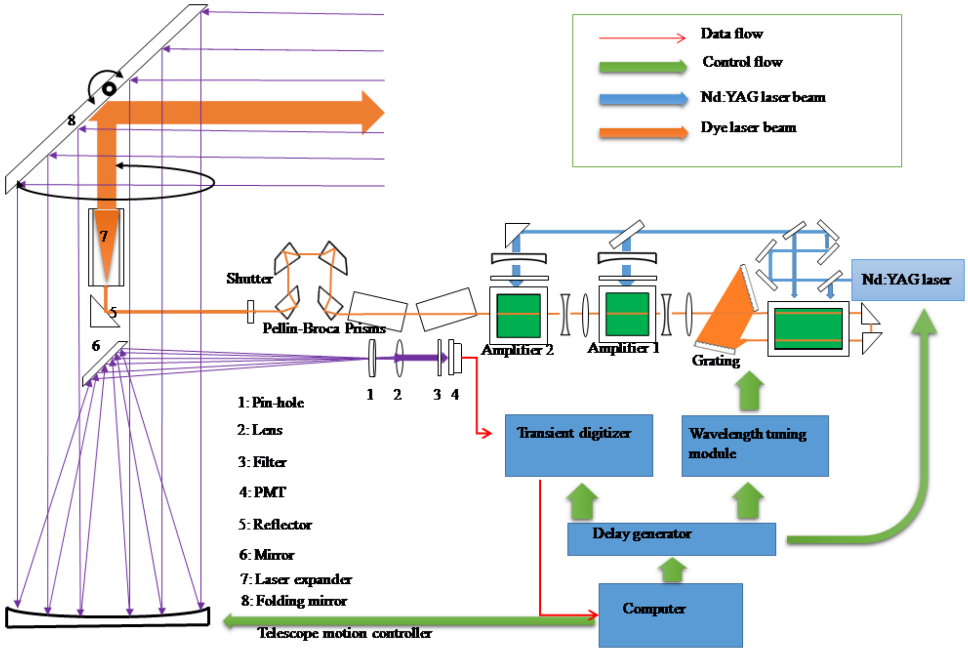

Fig. 1. Scheme of differential absorption lidar (DIAL) system.

3. Results and Discussion

A 24-hour mercury monitoring measurement campaign in Guangzhou city was performed. During this period, the temperature stayed constant at 14±1˚C. Fig. 2 presents an individual differential absorption curve taken for a laser beam directed at 18 degrees elevation. According to Eq. (1), the mercury concentrations could be calculated. Fig. 3 presents the mercury concentration of the campaign. The mercury concentration is analyzed in two different ranges, for low altitude from 60 m to 120m (corresponding distance from 150 m to 345 m) in Fig. 3 (b), and for the higher altitude from 120 m to 205m (corresponding distance from 345 m to 600 m) in Fig. 3 (a). It is found that the average mercury concentration for the low altitude is 12±1 ng/m^3, while for the high altitude it is slightly lower, i.e., 8±2 ng/m^3. The signal-noise-ratio of the differential absorption curve determines the error of the measurement of mercury concentration. The errors of the mercury concentration of low altitude are generally smaller than ±1 ng/m^3, and they are much larger for the high altitude.

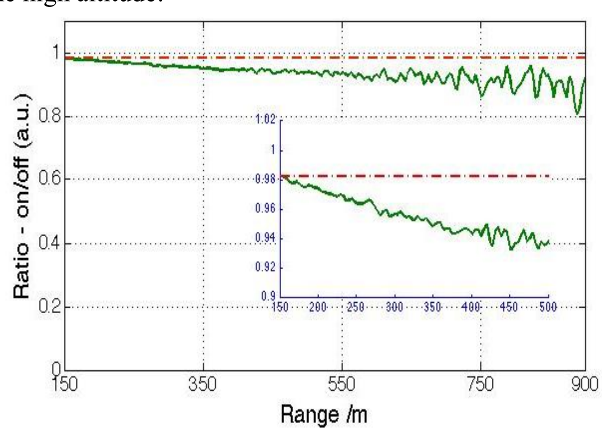

Fig. 2. Differential absorption curve for mercury measurement; recording time was at January 11th, 2013, from 21:00 to 22:00;

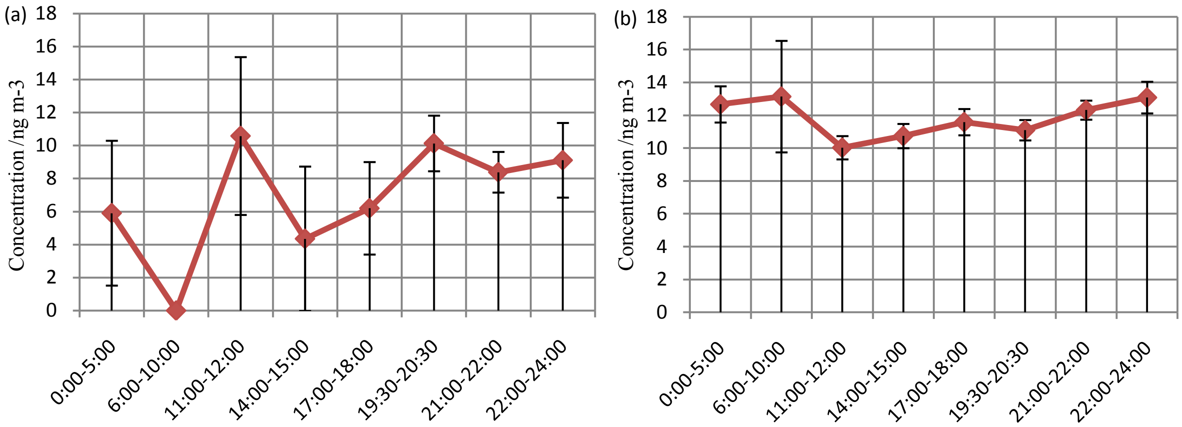

Fig. 3. Mercury concentration variation during the 24-hour recording; Low altitude data(60 m - 120m) are presented in (b), and higher altitude values (120m- 205m) are given in (a).The error bars give the evaluated errors of the mercury concentrations.

This preliminary measurements show that the differential absorption lidar system performance is good to monitor mercury concentration with some height resolution. As for low altitudes (below 120m), the average concentration was about 12 ng/m^3 in Guangzhou, a quite low value related to the low temperature in the winter, reducing degassing of metallic mercury pollution. Further measurements of mercury concentrations for different weather conditions are planned.

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