Remote monitoring of sulfur dioxide using a mobile lidar system

LIU Xiao-qin, ZHANG Yin-chao, HU Huan-ling, TAN Kun, YANG Gao-chao, SHAO Shi-sheng

(Laboratory of Atmospheric Optics, Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Hefei 230031, China)

Abstract: A mobile differential absorption lidar (DIAL) system, which is fully controlled by a microcomputer, has been constructed and applied in routine monitoring of atmospheric pollution such as SO_2 etc. Examples of measurements of SO_2 in Hefei and Beijing are given and results are discussed.

Key words: DIAL; Sulfur dioxide; Tunable laser

CLC Number: TN249 **Document code**: A **Article ID**: 1007-2276(2005)02-0151-04

用车载差分吸收激光雷达系统遥测二氧化硫

刘小勤，张寅超，胡欢陵，谭 锟，杨高潮，邵石生

（中国科学院 安徽光学精密机械研究所，安徽 合肥 230031）

摘要：介绍了自行研制的车载差分吸收激光雷达系统，此台雷达被用于监测大气污染如二氧化硫等。利用此台雷达在北京和合肥进行了实地测量，给出了对二氧化硫测量的典型结果并进行讨论分析。

关键词：差分激光雷达； 二氧化硫； 可调谐激光器

0 Introduction

Sulfur dioxide is one of the major air pollutants. It is harmful to human being, animal and nature environments. It may cause serious injuries to human such as pneumonia. In atmosphere sulfur dioxide combines with water and forms H_2SO_3 and H_2SO_4, then it becomes acid rain. Acid rain is responsible for the acidification of soil and water which has bad effects to human, animal and environment too.

Lidar technique applied in remote sensing of atmospheric gases has become a new efficient tool for measurements on air pollutants. It can be used monitoring a large range gases distribution for real-time measurement with high spatial resolution and temporal resolution[1-6]. Moreover, mobile system makes it easy to move from one place to another to monitor air pollution. Conventional analytical methods rely on air sampling by use of extractive methods in which the air sample is collected inside an appropriate container. It is difficult to obtain range distribution of pollutants.

In this paper, the system structure and the Lidar principle of a mobile differential absorption lidar system (AML-1) designed by us are presented. It is applied in routine monitoring of atmospheric pollution such as SO_2 etc. The field tests in Beijing and Hefei were performed by this lidar system. Measurements examples of sulfur dioxide in Hefei and Beijing are

收稿日期：2004-03-10； 修订日期：2004-05-13

作者简介：刘小勤(1967-)，女，安徽巢湖人，副研究员，硕士，主要从事实用新型激光雷达研制及激光雷达仿真软件研究。

given and results are compared with the results of ground monitor instruments.

1 Measurement theory

The principle of the differential absorption lidar technique has been described clearly before[1-3], only a brief explanation of the theory is given in this paper. This technique is based on the difference in absorption cross section of a certain molecular gas at two or more close lying wavelengths. Use of two different wavelength laser having different absorption cross sections for the gas under study: one wavelength(λ_{on}) is strongly absorbed, whereas the other one (λ_{off}) only weakly absorbed. In the case of SO_2 measurement, the DIAL is accomplished using two return signals at on and off wavelengths in the UV absorption band of SO_2. Then, the mean concentration of SO_2 is calculated for range intervals Δz according to the equation (1) :

$$\overline{N}(z)=\frac{1}{2\Delta z\Delta\delta}\times \left\{-\mathrm{Ln}\frac{p_{\lambda_{on}}(z+\Delta z)}{p_{\lambda_{on}}(z)}+\mathrm{Ln}\frac{p_{\lambda_{off}}(z+\Delta z)}{p_{\lambda_{off}}(z)}+B+E\right\} \quad (1)$$

where $B=-\mathrm{Ln}\dfrac{\beta_{\lambda_{on}}(z)\beta_{\lambda_{off}}(z+\Delta z)}{\beta_{\lambda_{off}}(z)\beta_{\lambda_{on}}(z+\Delta z)}$;

$$E=-2\{\alpha_{\lambda_{on}}(z)-\alpha_{\lambda_{off}}(z)\}$$

with $\Delta\delta=\delta_{on}-\delta_{off}.\delta_{on}$ and δ_{off} are the absorption cross section of the gas studied at wavelengths λ_{on} and λ_{off} respectively. Where $\overline{N}$ (z) is the average molecular number density of the studied gas at range z; $P_{\lambda_{on}}$ and $P_{\lambda_{off}}$ are the received power from range z with a range thickness Δz for wavelengths λ_{on} and λ_{off} respectively; β_{on} and β_{off} are the atmospheric volume backscatter coefficient; α_{on} is the atmospheric extinction coefficient due to molecule, aerosol and other gas absorption components. Because the two wavelengths are close to each other and the shooting interval between two wavelength laser is shorter, effect of aerosol and molecular is very small. The last two terms in equation (1) can be neglected. The equation (1) can be described as (2):

$$(z)=\frac{1}{2\Delta z\Delta\delta}\left\{-\mathrm{Ln}\frac{P_{\lambda_{on}}(z+\Delta z)}{P_{\lambda_{on}}(z)}+\mathrm{Ln}\frac{P_{\lambda_{off}}(z+\Delta z)}{P_{\lambda_{off}}(z)}\right\} \quad (2)$$

2 Mobile system for SO_2 DIAL measurements

A mobile lidar system (AML-1) was constructed in 2000 at Anhui Institute of Optics and Fine Mechanics China which can be used to measure four kinds of pollutants in atmosphere, Aerosol, SO_2, NO_2, and O_3. It was composed of the laser and transmitter part, the three-dimension scanner, the detection part, and the data acquisition part. Fig.1 is a schematic diagram of the system which has been described in detail before [4]. Some basic parameters for the lidar system are given in Tab.1. Here will only discuss the details which are of special interest in applications of the DIAL technique for measurements on SO_2.

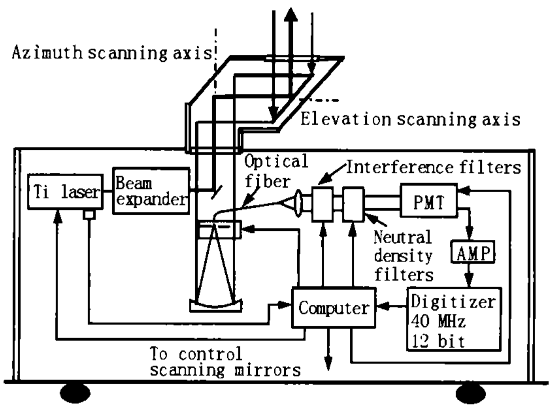

Fig.1 Schematic diagram of the mobile lidar system

The tunable Ti:saphire laser which made by Germany Elight company has been used in AML-1 lidar system. It is a flashlamp pumped Titanium Sapphire laser with SHG/THG frequency conversion, and provides pulsed UV radiation over continuing ranges 375~400/250~290 nm. In the case of SO_2 mea- surement, the laser system is employed for producing wavelengths 286.3 nm and 286.9 nm to the wavelengths on and off,respectively.The accuracy of two wavelen - gths are emphasized because this two laser wavelengths are very close. Wavelength calibration use an argon Galvatron which is hollow-cathode discharge lamp filled with argon and is a more precise wavelength calibration. The calibration routine is simple, for all operation control by computer. Calibration must be run through before each measurement to ensure accuracy of wavelength for the intended measurement. Calibration accuracy and stability are ±0.1 nm.After calibration, the laser is operating at 20 Hz and the laser wavelength is alternated between two laser pulse.

Tab.1 Basic parameters of the AML-1 lidar system

Transmitted pulse energy	1~4 mJ
Pulse repetition rate	20 Hz
Laser beam divergence	≤3.5 mrad
Diameter of the receiving telescope	300 mm
Bandwidth of the interference filter	7.5 nm
Quantum efficiency of the photomultiplier tube	26%

The two laser beams are generated by one laser and transmitted through the same optical components before leaving the system, and transmit and receive telescope are coaxial. The beam is expanded twelvefold by a beam expander before being transmitted into the atmosphere. The beam divergence is <0.3 mrad after expansion. An iris has been used which reduces the background light by decreasing the telescope field of view and also geometrically compresses the signal dynamic range. The view of iris can be changed from 0.2 mrad to 1.7 mrad according to the atmospheric condition and energy of laser. A slight misalignment of the two beams or a small difference in the intensity distribution may give distance dependence since the overlap function of the beam and the telescope field of view may change drastically. In SO_2 measurements it is possible to carefully adjust the beam direction and avoid this problem with our system. Interference filters, 7.5 nm bandwidth with a transmission of 25~40% are used to suppress the background skylight.

The optical signals are detected by a photomultiplier(EMI9214b). A fast digitizer which has a dynamic range of 4096 steps (12 bit) and the maximum sample rate of 40 MHz is used for data acquisition. Thanks for computer technique development, sample and save per pulse return signal, average signal, process averaged signal and display results can be done on-line. The software of data process and display has also been designed. When the backscattered signal from atmosphere has been obtained by digitizer in the computer, the data processing software will be run automatically, the final image will be displayed and the final result including data and images will be stored in database respectively for later research and analyses.

3 Results and discussion

The atmosphere backscattered signals are obtained with 15 m spatial resolution. In a typical measurement 5000 lidar return signals for each wavelength are averaged. This averaging is needed to improve the signal-to-noise ratio, and also improves the dynamic range that can be detected by the system. SO_2 concentration profile was calculated according to DIAL equation (2).

The field tests have been done by using the AML-1 lidar system in Beijing and Hefei. Here some results have been selected and compared with results of local ground monitors which were measured by local environmental protection department with conventional chemical methods.In Fig.2,a typical dial measurement of SO_2 in the ambient air is shown. The three continual vertical profiles of SO_2 concentration have been obtained in Hefei on 2001/11/06. The average value obtained by a local ground monitor at same place and same time is given for comparison. It can be seen from the figure that three curves have same trend. The fluctuation among them is not more than 10%. The results are stability. The mean values of three curves are 6.76 ppb、5.52 ppb and 5.39 ppb respectively cohering with mean value 6.47 pbb of local ground monitor.

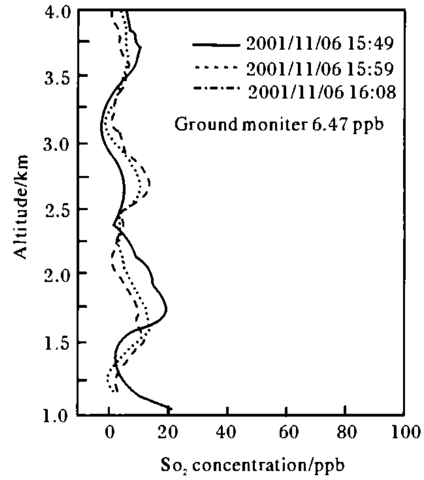

Fig.2 SO_2 profiles obtained by AML-1 in Hefei

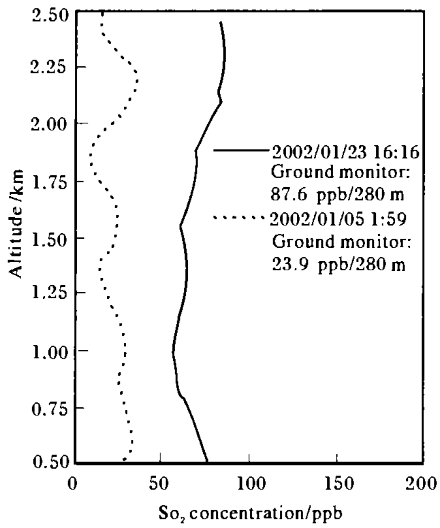

Fig.3 SO_2 profiles obtained by AML-1 in Beijing

The Fig.3 shows the two vertical profile of SO_2 measurement in Beijing on 2002/01/23 and 2002/01/05. The results obtained by local ground monitor at 280 m tower are also presented in this figure. The dot line in this figure was measured on 11:59 2002/01/05. The SO_2 concentration of that time fluctuated around 20 ppb and consistent to value 23.9 ppb obtained by the local ground monitor at a distance of 280 m. The real line of the figure was measured at time 16:16 on 2002/01/23. As can be seen the remote sensing results are fully compatible with the reference value. The SO_2 concentration of that day was higher than 2002/01/05 verified by AML-1 lidar measurement results and local ground monitor value 87.6 ppb.

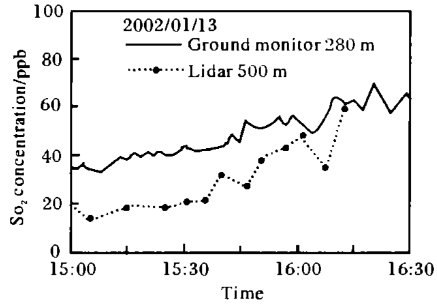

Fig.4 Time variation of the SO_2 concentration over Beijing

The time variation of SO_2 concentration over Beijing is shown in Fig.4 from time 15:00 to 16:15 on 2002/01/13. Evaluation of the backscattered return signals from the atmosphere at 500 m as well as a local ground monitor at distance of 280 m are shown in the figure. As can be seen the remote sensing results have a same trend with the reference values. The relativity of them is good. The difference of values between them may be caused by different altitude. From the figure, after the time half and fifteen, SO_2 concentration of two curves both begin rising, for the measurement place near a high way the traffic becomes busy from that time.

The SO_2 measurement errors from the experimental results are estimated considering the following sources of error: (1) The uncertainty different absorption cross section. (2) The statistical error. (3) The existence of interfering species in the atmosphere. (4) Instrumental error. The estimation was performed according to the method described in reference paper[7-9]. The total error of AML-1 in the SO_2 measurement with a range resolution of 300 m estimated to be <7 ppb.

The lidar system described in this paper has been proved efficient and useful for SO_2 pollutants probing.

Reference:

[1] Schotland R M.Some observations of the vertical profile of wave vapor by a laser optical radar[A]. Ann Arbor Proc 4th Symposium

(下转第 163 页)

0.97203(通道 2)。测量结果如图 5 所示。

由以上数据可以看出，所研制的模块在量程范围内的标准误差约为 150 ps，且有较好的线性度，通道 1 的线性度优于通道 2。在 3.0~4.0 μs 范围内，测量误差 Δt 靠近零，而在其两侧 Δt 符号相反，这是因为设置 TDC 对 20 MHz 时钟的分频因子为 64，这样参考时钟周期 T_{ref}=3.2 μs，所以才会出现这种情况。另外也可以看出测量的结果存在明显的量子化效应，这一方面与最终输出的结果数据只有 15 bits 有关，最后一位的权重为 195 ps，所以测出的结果以 195 ps 跳变；另一方面 TDC 采用的是延迟线插入法，测量的结果必定是离散的，其最高时间间隔分辨力只能达到一个 LSB(最高为 250 ps)，这是其极限误差。此外，测量结果的跨度约为 588 ps，这主要是因为 TDC 和恒温晶振采用同一电源隔离器供电，而恒温晶振的功耗较大势必对供电电压的稳定性造成影响，从而影响 TDC 测量的精度。而且本模块采用的是两层 PCB 板制作，因此在信号抗干扰上还有待完善。

3 结 论

采用 TDC 专用芯片开发的模块，不仅线路简单，控制和使用也非常方便，利用计算机总线，便于集成。研制的时间间隔测量模块达到了机载激光测深系统所要求的测距精度，并已经用于系统中。在以后的改进中，可以采用更高时间分辨率的专用时间数字转换芯片，对晶振和时间数字转换芯片分开供电，并进一步提高供电电压的稳定性，在电路制作上可以采用四层或者更高的制作工艺，提高线路的抗干扰能力。研制的模块可用于任何脉冲激光测距和时间间隔测量应用中。

参考文献：

[1] Thmoas R W L,Guenther G C.Water surface detection strategy for an airborne laser bathymetry[A].SPIE:Ocean Optics X[C].1990,1302. 597-611.

[2] 陈卫标，陆雨田，褚椿霖，等.机载激光水深测量的精度分析[J].中国激光，2004，31(1):101-104.

[3] 霍玉晶，陈千颂，潘志文.脉冲激光雷达的时间间隔测量综述[J].激光与红外，2001，31(3):136-139.

[4] Acam-messelectronic gmbh.Am Hasenbiel27-D-76297,Stutensee-Blankenloch-Germany [EB/OL].http://www.acam.de/Documents/English/DB_GP1_e.pdf,2001-2-12.

[5] NATIONAL instruments,PCI-6503 User Manual [DB/OL].http://www.ni.com/pdf/manuals/320938c.pdf.

[6] Karen Hazzah；孙喜明.Windows Vxd 与设备驱动程序权威指南.第二版[M].北京：中国电力出版社，2001.

(上接第 154 页)

on Remote Sensing of Environment[C]. University of Michigan,1996,273-283.

[2] Fredriksson K,Galle B,Nystrom K,et al. Lidar system applied in atmospheric pollution monitoring[J].Appl Opt,1979,18:2998-3003.

[3] Egeback A L,Fredriksson K A,Hertz H.DIAL techniques for the control of sulfur dioxide emissions[J].Appl Opt,1984,23(5):722-729.

[4] Yinchao Zhang, Huh Huanling, Kan ten, et al. A mobile lidar system for air pollution measurement[A].SPIE 3rd International Asia-Pacific Symposium on Remote Sensing of the Atmosphere, Ocean, Environment, and Space[C].Hangzhou, SPIE, 2002.

[5] Browell E V,Ismail S,Shipley S T.Ultraviolet DIAL measurements of O_3 profiles in regions of spatially inhomogeneous aerosols[J].Appl Opt,1985,24(17):2827-2841.

[6] Fredriksson K,Galle B,Nystrom K, et al. Mobile lidar system for environmental probing[J].Appl Opt,1981,20(24):4181-4189.

[7] Tetsuo Fukuchi, Naohiko Goto, Nemoto K,et.al.Error analysis of SO_2 measurement by multiwavelength differential absorption lidar [J].Opt Eng,1999,38(1):141-145.

[8] Takashi Fuji,Tetsuo Fukuchi,Nao hiko Goto,et al.Duel differential absorption lidar for the measurement of atmospheric SO_2 of the order of parts in 109[J]. Appl Opt,2001,40(6):949-956.

[9] Takashi Fuji,Tetsuo Fukuchi,Nianwen Cao,et al.Trace atmospheric SO_2 measurement by multiwavelength curve-fitting and wavelength optimized dual differential absorption lidar[J].Appl Opt,2002,43(3):524-531.