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Analysis of influence of atmosphere extinction to Raman lidar monitoring CO_2 concentration profile*

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Lidar (Light detection and ranging) system monitoring of the atmosphere is a novel and powerful technique tool. The Raman lidar is well established today as a leading research tool in the study of numerous important areas in the atmospheric sciences. In this paper, the principle of Raman lidar technique measurement CO_2 concentration profile is presented and the errors caused by molecular and aerosol extinction for CO_2 concentration profile measurement with Raman lidar are also presented. The standard atmosphere extinction profile and 'real-time' Hefei area extinction profile are used to conduct correction and the corresponding results are yielded. Simulation results with standard atmosphere mode correction indicate that the errors caused by molecule and aerosol extinction should be counted for the reason that they could reach about 8 ppm and 5 ppm respectively. The relative error caused by Hefei area extinction correction could reach about 6%. The errors caused by the two components extinction influence could produce significant changes for CO_2 concentration profile and need to be counted in data processing which could improve the measurement accuracies.

Keywords: Raman lidar, atmospheric CO_2, aerosol extinction, molecule extinction
PACC: 9260T, 8670G, 8670L

1. Introduction

Monitoring atmospheric CO_2 concentration is important in predicting the 'climate change' effects due to its contribution to the planetary greenhouse effect. Burning of carbon-based fuels has led to a steady increase in the concentration of atmospheric carbon dioxide (CO_2) during the last 100 years, consequently the global average temperature has increased about 0.5 °C.[1] The ultimate objective of scientists is to achieve stabilization of greenhouse gas concentrations in the atmosphere at a level that would prevent dangerous anthropogenic (man-made) interference with the climate system. Lidars are powerful tools in the domain of environmental quality monitoring.[2] Lidars make use of different optical techniques such as Raleigh and Raman scattering, differential absorption, Doppler and resonance scattering. Raman technique has an important application in lidar fields for its simple conformation advantage.[3−6] In this work, extinction profiles of standard atmosphere and 'real-time' Hefei area mode were used to conduct correction and the errors caused by atmosphere extinction for CO_2 concentration profile were analysed.

2. Principles of Raman lidar technique

Lidars are valuable instruments for atmospheric research because they provide an active remote sensing technique and have advantages of high spatial and temporal resolution that can probe atmospheric regions inaccessible to other instruments. Raman scattering is one of the processes that occurs when optical radiation is scattered from the atmosphere molecules. It is most useful because the vibrational Raman scattering provides distinct wavelength shifts for the specific vibrational energy states of the molecule of species. The signals are collected by a telescope and

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processed by wavelength selective optics such as filters or spectroscopy. A single wavelength lidar system emits short laser pulses into the atmosphere and measures the power P of the light backscattered into its receiver as a function of distance r. The power $P_p(r)$ of Raman back-scattered signals is described by the lidar equation

$$P_{\mathrm{p}}(r) = \frac{C_{\mathrm{A}}(\lambda_{\mathrm{p}})N_{\mathrm{P}}(r)\beta^0_{\mathrm{P}}(r)}{r^2} \times \exp\Big[-\int_0^r [\sigma(r',\lambda)+\sigma(r',\lambda_{\mathrm{p}})]\mathrm{d}r'\Big], \quad (1)$$

where $C_{\mathrm{A}}(\lambda_{\mathrm{p}})$ is the lidar system constant, λ_{p} is the wavelength of the backscattered light received, r is the altitude from which the return signal is scattered, $\beta^0_{\mathrm{p}}(r)$ is the pertinent Raman backscattering cross section at shifted wavelength λ_{p}, $\sigma(r,\lambda)$ and $\sigma(r,\lambda_{\mathrm{p}})$ are atmosphere extinction coefficients at wavelength λ and λ_{p} respectively. $N_{\mathrm{p}}(r)$ is the number density of pollutants. Equation (1) is undetermined, as $C_{\mathrm{A}}(\lambda_{\mathrm{p}})$, $\beta^0_{\mathrm{p}}(r)$, $\sigma(r,\lambda)$ and $\sigma(r,\lambda_{\mathrm{p}})$ are unknown and cannot be solved without additional assumptions or calibrations. Upon examination of the equation, it becomes apparent that the Raman techniques which use the ratio of the signals at two wavelengths will greatly simplify the measurements of various parameters. The Raman signals of nitrogen (N_2) and oxygen (O_2) are often used as reference signals due to their steady mixing ratio. After introducing the lidar equation of reference gases, we can get the equation

$$N_{\mathrm{p}}(r) = \frac{N_{\mathrm{R}}(r)C_{\mathrm{A}}(\lambda_{\mathrm{R}})P_{\mathrm{P}}(r)}{C_{\mathrm{A}}(\lambda_{\mathrm{P}})P_{\mathrm{R}}(r)} * \frac{\beta^0_{\mathrm{R}}(r)}{\beta^0_{\mathrm{P}}(r)} \times \exp\Big[\int_0^r [\sigma(r',\lambda_{\mathrm{R}})-\sigma(r',\lambda_{\mathrm{P}})]\mathrm{d}r'\Big], \quad (2)$$

where the subscript R indicates reference gases such as N_2 or O_2, the physical implications of parameters in Eq.(2) are similar to parameters in Eq.(1). The value of $N_{\mathrm{p}}(r)$ could be obtained from Eq.(2) after calibrating various parameters. Normally, the value of parameter $\beta^0_{\mathrm{p}}(r)$ and $\beta^0_{\mathrm{R}}(r)$ can be ascertained by experiment or theoretical calculation. $N_{\mathrm{R}}(r)$, $\sigma(r,\lambda_{\mathrm{R}})$ and $\sigma(r,\lambda_{\mathrm{p}})$ can be obtained from standard atmosphere mode or radiosonde.

The third harmonic (355 nm) of a Nd:YAG laser was used to provide night-time measurement. After expansion and reflection, laser beams are conducted into atmosphere, the signals including elastic scattering signals (Mie and Rayleigh scatter signal at 355 nm wavelength) and Raman non-elastic signals (N_2 , CO_2 and other gases Raman scatter signals at 355 nm wavelength) are collected by telescope. Raman scattering signals of CO_2 (371.66 nm) and N_2 (386.66 nm) provide rude concentration profiles of CO_2 and N_2 in the lower atmosphere. The collected signals are introduced into narrow band filters via fibre and into photoelectron multiple tube and amplifier sequentially, at last photons could be counted by photon counter. At the same time, an energy meter records the variety of laser intensity in order to eliminate the influence of energy fluctuation.

3. Data processing and error analysed

3.1. Data processing without extinction correction

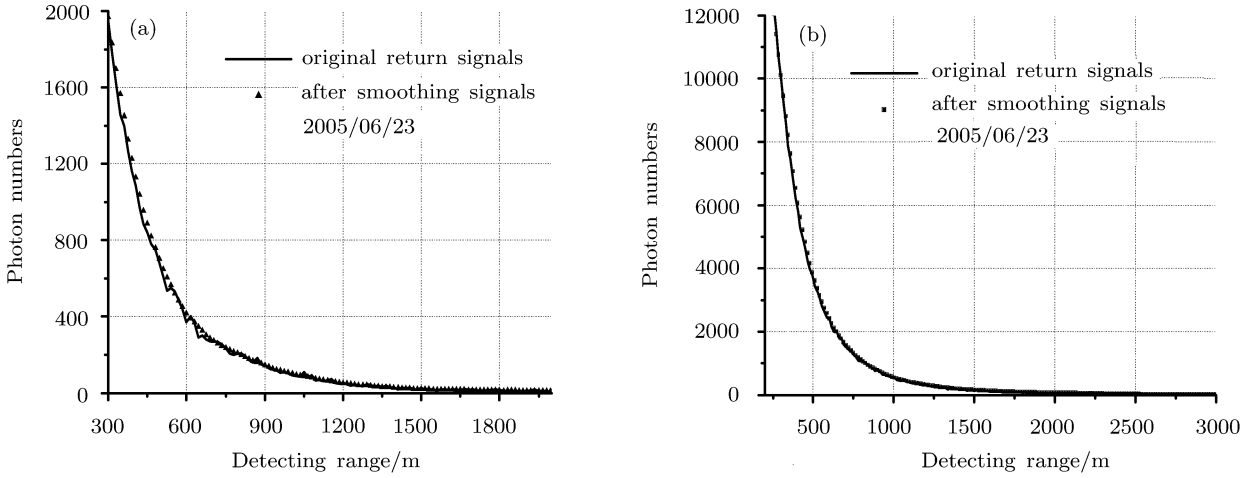


Fig.1. Return signals for CO_2 and N_2 at Hefei someday.

Original signal smoothing and background-subtracting and signal retrieving constitute the lidar data processing process. The Raman lidar system measuring CO_2 concentration profile often makes use of the adjacent average smoothing method as shown in Fig.1. Normally, additional assumptions are introduced based on Eq.(2). Firstly, we assume that the ratio of $\beta_{\rm p}^0(r)$ to $\beta_{\rm R}^0(r)$ is the constant value of 1 considering that two wavelengths are so close to each other. Secondly, we neglect the influence of atmosphere molecular and aerosol extinction.[7,8]

The results of CO_2 concentration profile was obtained as shown in Fig.2. The results could give an imprecise indication of CO_2 concentration profile in a way. Some potential factors which would influence the measured accuracies exist, such as the atmosphere extinction and other gases' interference. To improve the measurement accuracies, we need to take into account the molecular and aerosol extinction influence which are considered in the following parts.

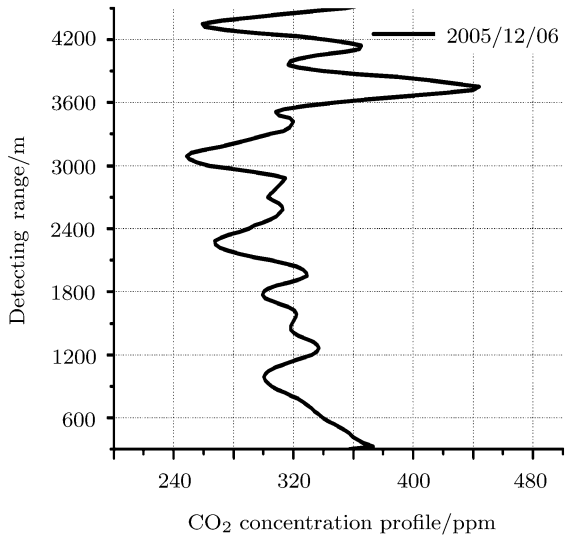


Fig.2. CO_2 concentration profile someday.

3.2. Data processing with extinction correction

In previous works, the influence of molecule and aerosol extinction was often neglected for the reason that the double Raman excited wavelengths were too close to each other.[7,8] The previous approach which neglected the influence was deemed reasonable, but some researches recognized the importance of considering the difference of transmission function of two Raman scattered signals during the data processing.[9,10] In this part, the extinction influence of two components for Raman lidar data processing was simulated. The standard atmosphere mode and a real aerosol extinction profile of the Hefei area were used to simulate the retrieving process. The errors caused by neglecting the influence were also estimated.

3.2.1. . Data processing with standard atmosphere mode correction

The extinction that occurs at a certain wavelength is due to both scattering and absorption from both aerosols and molecules in the atmosphere and is a function of wavelength. The total extinction coefficient is therefore given by aerosol and molecular contributions:

$$\sigma(r,\lambda_{\rm R}) = \sigma_{\rm m}(r,\lambda_{\rm R}) + \sigma_{\rm A}(r,\lambda_{\rm R}),$$
$$\sigma(r,\lambda_{\rm P}) = \sigma_{\rm m}(r,\lambda_{\rm p}) + \sigma_{\rm A}(r,\lambda_{\rm P}), \qquad (3)$$

where the subscripts m and A indicate molecule and aerosol contributions respectively. Normally, to derive the extinction coefficient profile, the middle latitude standard atmosphere mode of America is most commonly used. The relationship of two components' volume backscatter and laser wavelength and altitude is given by the relationships

$$\beta_{\rm m}(r) = 1.54\times10^{-3}\Big(\frac{532}{\lambda}\Big)^4\exp\Big(-\frac{r}{7}\Big),$$
$$\beta_{\rm A}(r) = \Big\{2.47\times10^{-3}\exp\Big(-\frac{r}{2}\Big) + 5.13\times10^{-6} \times\exp\Big[-\frac{(r-20)^2}{6^2}\Big]\Big\}\Big(\frac{532}{\lambda}\Big), \qquad (4)$$

where $\beta_{\rm m}(r)$, $\beta_{\rm A}(r)$ indicate molecule and aerosol volume backscatter coefficient, the extinction–backscatter ratio of molecule and aerosol are $8\pi/3$ and 50, respectively. More details of the two components' extinction coefficient can be shown by Fig.3. We currently use molecule and aerosol extinction profiles at the wavelengths of 386.68 nm and 371.66 nm to evaluate the influence. The extinction profiles of two components can be obtained according to the conversion relations Eq.(4). We define $Q_{\rm m,A}\ (\lambda,\lambda_{\rm R},r)$ as molecule and aerosol total transmission function at N_2 Raman shifted wavelength, which can be expressed as follows:

$$Q_{\rm m,A}(\lambda,\lambda_{\rm R},r) = \exp\Big[-\int_0^r[\sigma_{\rm m,A}(r',\lambda)+\sigma_{\rm m,A}(r',\lambda_{\rm R})]{\rm d}r'\Big]. \qquad (5)$$

With the same approach, molecule and aerosol total transmission function at CO_2 Raman shifted wavelength can be shown as

$$Q_{\mathrm{m,A}}(\lambda, \lambda_{\mathrm{P}}, r) = \exp\Big[-\int_0^r [\sigma_{\mathrm{m,A}}(r', \lambda) + \sigma_{\mathrm{m,A}}(r', \lambda_{\mathrm{P}})]\mathrm{d}r'\Big]. \quad (6)$$

It is easy to find that the atmospheric transmission function includes two terms, which account for the fact that photons are transmitted into the atmosphere at laser wavelength λ and return at Raman-shifted wavelength λ_{p} and λ_{R} for pollutants and reference gases. Thus the above Eqs.(5) and (6) can be simplified by neglecting the former term as shown by the following equations:

$$Q'_{\mathrm{m,A}}(\lambda_{\mathrm{R}}, r) = \exp\Big[-\int_0^r \sigma_{\mathrm{m,A}}(r', \lambda_{\mathrm{R}})\mathrm{d}r'\Big],$$
$$Q'_{\mathrm{m,A}}(\lambda_{\mathrm{P}}, r) = \exp\Big[-\int_0^r \sigma_{\mathrm{m,A}}(r', \lambda_{\mathrm{P}})\mathrm{d}r'\Big], \quad (7)$$

where the subscripts m and A indicate molecule and aerosol particulate contributions respectively. According to the above analysis, the simulation results were obtained as shown in Fig.4. The simulation with standard atmosphere mode correction indicated that molecule and aerosol extinction could influence the retrieving results differently.

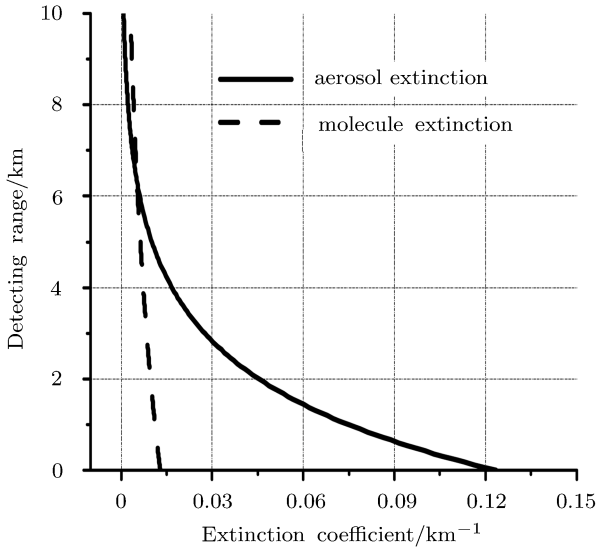


Fig.3. Aerosol and molecule extinction of standard atmosphere mode of middle latitude of American at 532 nm.

It is easy to see from Fig.3 that the molecule extinction coefficient is smaller than that of aerosol, but the error caused by the molecule extinction is larger than that caused by aerosol influence, probably because the former extinction changed nonlinearly with the wavelength: its change accords with λ^{-4} relations and decay follows the exponent relations. The value of errors caused by molecule extinction could reach about 8 ppm. It could lead to evident change for signal retrieving with molecule extinction correction in comparison with that without correction. At the same time, the influence of aerosol extinction must be taken into account during the data processing, it also could lead to evident change for the result with correction. The value of error caused by the aerosol extinction could reach about 5 ppm. The relative error caused by two components could reach 10 ppm and should not be neglected for the reason that CO_2 concentration usually reaches 350–400 ppm.

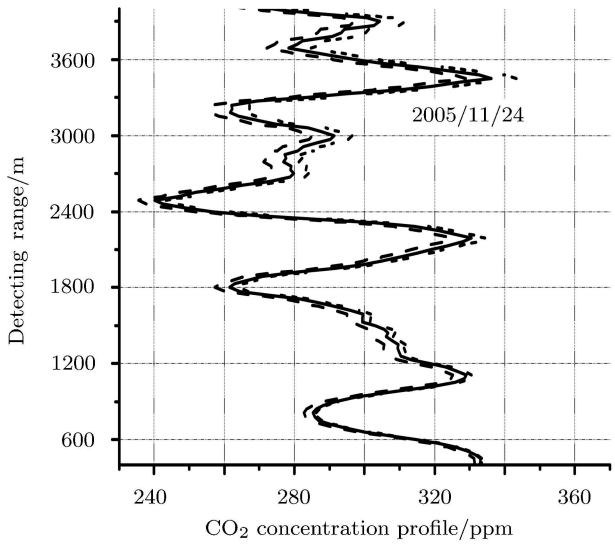


Fig.4. Simulation with standard atmosphere mode considering aerosol and atmosphere molecule extinction. –– Result without extinction correction, —— Result with aerosol extinction correction, ----- Result with molecule extinction correction.

3.2.2. . Data processing correction at Hefei area

Atmosphere extinction often changes with different area and climate conditions even for the same area and it often has different extinction profiles following season changes.[11,12] Volcano eruption and human activities usually change the aerosol concentration and its extinction coefficient. To improve the measurement accuracy of Raman lidar measuring CO_2 concentration, we need to make use of the 'real-time' extinction profile. In this part, the atmosphere extinction profile of Hefei area from AML-2 (Air Monitoring Lidar -2) was used to conduct correction. Figure 5 indicates the

different extinction profiles for different times someday. It is easy to find that extinction profiles changed evidently at different times and had distinct difference in comparison with the standard atmosphere mode. The Raman lidar we used currently could not implement the function of collecting extinction signals (Mie and Rayleigh scattering) and needs to be improved in future works. Figure 6 indicates that the error caused by atmosphere extinction correction could reach about 13 ppm. Figure 7 indicates that the relative error with Hefei area extinction correction could reach about 6%, and the relative error with standard atmosphere mode correction could reach 5%, and the relative error with Hefei area aerosol extinction correction could reach 4.5%.

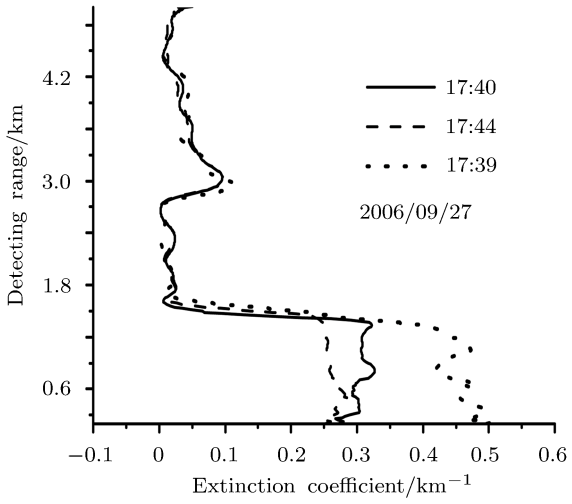


Fig.5. Extinction profile of Hefei area.

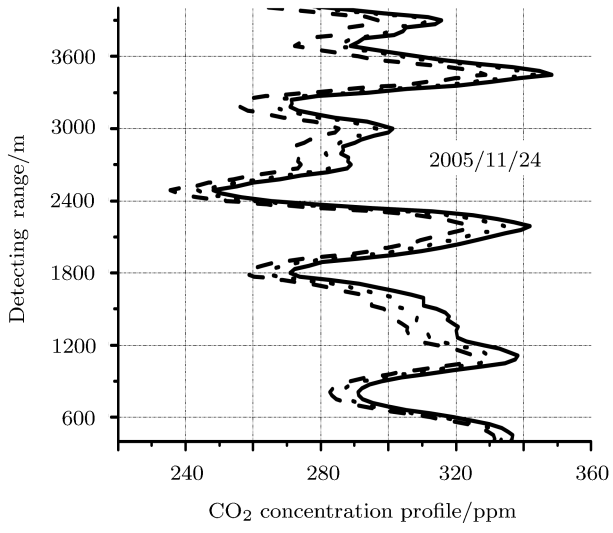


Fig.6. Simulation correction with Hefei area extinction. —— Result with Hefei area extinction correction, – – Result without extinction correction, ····· Result with standard atmosphere mode correction.

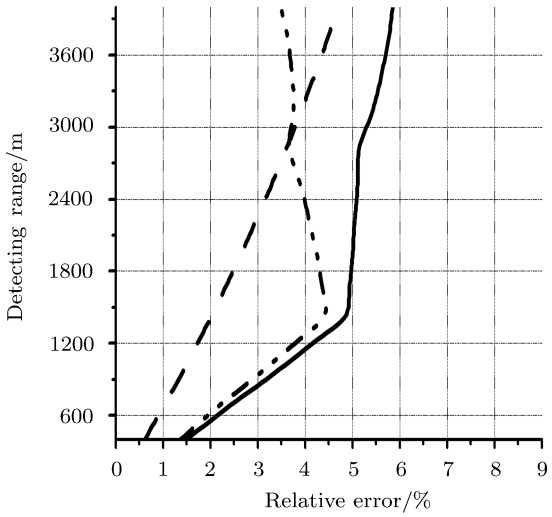


Fig.7. Relative error after considering extinction correction. —— Relative error with Hefei area extinction correction, – – Relative error with standard atmosphere mode correction, ····· Relative error with Hefei area aerosol extinction correction.

4. Conclusions and planned future work

In this paper, the principle of Raman Lidar technique measuring tropospheric CO_2 concentration profile is presented. At the same time, the analysis of influence of atmosphere extinction was also presented. Standard atmosphere of the middle latitude of America was used to conduct correction and obtained the conclusion that the error caused by molecule extinction correction could reach about 8 ppm which is slightly larger than the error caused by aerosol extinction correction. The Hefei area extinction profile was used to simulate the correction due to the disagreement with standard atmosphere mode, variability of atmosphere extinction profile following alternating seasons, and influenced by human activities. Simulated correction which used Hefei area extinction profile was different from the former correction. To improve the measurement accuracies, we need to improve the current Raman Lidar compositions to collect the Mie and Rayleigh scattering signals which could be used to retrieve extinction coefficient in real-time mode. High signal-to-noise ratios and measurement

stability are needed for the measurement of Raman lidar CO_2 concentration profile.

Measuring the tropospheric CO_2 concentration profile from space has many potential error sources. These include possible interference from other trace gas species, the effects of temperature, clouds, aerosols and turbulence in the path, changes in surface reflectivity, and variability in dry air density caused by changes in atmospheric pressure, water vapour and topographic height. We have only analysed the atmosphere extinction influence and the others need to be accounted for in future works.

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