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Capability of Raman lidar for monitoring the variation of atmospheric CO_2 profile*

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Lidar (Light detection and ranging) has special capabilities for remote sensing of many different behaviours of the atmosphere. One of the techniques which show a great deal of promise for several applications is Raman scattering. The detecting capability, including maximum operation range and minimum detectable gas concentration is one of the most significant parameters for lidar remote sensing of pollutants. In this paper, based on the new method for evaluating the capabilities of a Raman lidar system, we present an evaluation of detecting capability of Raman lidar for monitoring atmospheric CO_2 in Hefei. Numerical simulations about the influence of atmospheric conditions on lidar detecting capability were carried out, and a conclusion can be drawn that the maximum difference of the operation ranges caused by the weather conditions alone can reach about 0.4 to 0.5km with a measuring precision within 30ppmv. The range of minimum detectable concentration caused by the weather conditions alone can reach about 20 to 35 ppmv in vertical direction for 20000 shots at a distance of 1 km on the assumption that other parameters are kept constant. The other corresponding parameters under different conditions are also given. The capability of Raman lidar operated in vertical direction was found to be superior to that operated in horizontal direction. During practical measurement with the Raman lidar whose hardware components were fixed, aerosol scattering extinction effect would be a significant factor that influenced the capability of Raman lidar. This work may be a valuable reference for lidar system designing, measurement accuracy improving and data processing.

Keywords: Raman lidar, atmospheric CO_2, minimum detectable concentration
PACC: 9265V, 7830, 8670L

1. Introduction

The Raman lidar is well established today as a key research tool in the study of numerous important areas in the atmospheric sciences. The early work of Cooney and Melfi *et al* in the late 1960s has demonstrated the technique of Raman spectroscopy in the measurement of tropospheric water vapour.[1] In 1985 Melfi and Whiteman extended the capability in both temporal and spatial resolution.[2] An operational prototype of Raman lidar instrument was prepared and demonstrated for the US Navy and is now used for scientific investigations. These measurements have demonstrated that Raman lidar can be used as a meteorological research tool which is unique in its ability for monitoring the atmospheric species in the lower atmosphere.[3] Recently, an operational prototype of Raman lidar system for atmospheric CO_2 profile monitoring has been developed by Anhui Institute of Optics and Fine Mechanics.[4−6] The Raman lidar can be used to monitor the variation of atmospheric CO_2 concentration with alternation of seasons of the year. As an important and necessary step of the whole project, the contribution of parametrization of Raman lidar system is significant to data processing and system performance improving. Conventional methods can be used to evaluate or analyse the detecting capability of Raman lidar based on the definition of signal-noise-ratio (SNR) at the photon detector output.[7,8] While the comprehensive nature of the SNR criterion

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makes it a very useful tool for assessing a given lidar system. It is also a weak point because it obscures the effects of the different components, and it is difficult to parameterize all the parameters exactly, especially the characteristics of parameters changing with wavelength. In this paper, using a new method for evaluating the capabilities of a Raman lidar system,[9] we present the numerical simulation of detecting capability of Raman lidar for monitoring the atmospheric CO_2 in Hefei, and analyse the influence of “weather” conditions for light wave propagation on SNR. The capability of Raman lidar for monitoring atmospheric CO_2 will be evaluated in the following sections.

2. Raman lidar for monitoring atmospheric CO_2 profile

2.1. Raman lidar measurement techniques

Raman scattering is one of the processes that occur when optical radiation is scattered from the molecules of the atmosphere. It is most useful because the vibrational Raman scattering provides distinct wavelength shifts for species.[10] The Raman lidar system for monitoring atmospheric CO_2 profile designed at our institute in Hefei is composed of a frequency–tripled Nd:YAG laser transmitter, which transmits at a wavelength of 354.7nm, an optical telescope receiver which is of quasi Cassegrain with a 35cm diameter, and various signal-processing and data-acquisition electronics.[4] Raman scattering is a weak molecular scattering process that is characterized by a shift in wavelength of the scattered beam of light relative to the incident one. In order to acquire the Raman backscattering photon effectively, we obtain data by photon counting (PC) instead of analogue-to-digital converter (ADC). The Raman shift toward a longer wavelength likely occurs at typical atmospheric temperatures and is known as the Stokes component of Raman scattering. The Raman shift wave number of atmospheric CO_2 is 1388cm^{-1} with pertinent centre shift spectrum at 371.66nm. Considering the characteristic of Raman lidar techniques, we often choose nitrogen (N_2) as reference medium for its constant proportion to dry air in low troposphere. The Raman shift wave number of atmospheric N_2 is 2330.7cm^{-1} with pertinent centre shift spectrum at 386.7nm.

2.2. Raman lidar equation

Assuming no multi-scattering, for a vibrational scattering Raman species X (CO_2 or N_2) we have the Raman lidar equation given by [11]

$$P(\lambda_X, r) = \frac{O_X(r)P_0(\lambda_L)A\xi(\lambda_X)N_X(r)[\mathrm{d}\sigma_X(\lambda_L,\pi)/\mathrm{d}\Omega]}{r^2} \times \exp\left\{-\int_0^r [\alpha(\lambda_L, r') + \alpha(\lambda_X, r')]\mathrm{d}r'\right\}, \quad (1)$$

where $O_X(r)$ is the Raman channel overlap function, and $P_0(\lambda_L)$ is the output power of the laser at the wavelength λ_L. $N_X(r)$ is the number density of molecular species X, and $\mathrm{d}\sigma_X(\lambda_L,\pi)/\mathrm{d}\Omega$ is the pertinent Raman backscatter cross section at the wavelength λ_L. $\xi(\lambda_X)$ is the total receiver optical efficiency at the wavelength λ_X. A is the area of the telescope. The exponential factor gives the two-way atmospheric transmission, $\alpha(\lambda_L, r')$ and $\alpha(\lambda_X, r')$ are the total extinction coefficients at the wavelengths λ_L and λ_X respectively, which are due to scattering and absorption by molecules, particles, and any other atmospheric constituents along the path of the laser beam. It should be noted that the atmospheric transmission function includes a term at the wavelength λ_L, for the transmission along the output path; and another for the backscattered signal at the wavelength λ_X, which has been shifted from the laser wavelength due to inelastic Raman scattering by the molecular species X.

The atmospheric CO_2 concentration, or mixing ratio, which is usually expressed by the ratio of CO_2 volume to the dry air in unit volume, can be determined by the ratio of two signals from the vibrational Raman shifts of CO_2 and nitrogen, expressed by

$$N_{CO_2}(r) = K\frac{P_{CO_2}(r)}{P_{N_2}(r)}, \quad (2)$$

where the calibration factor K is verified by comparing it with a rawinsonde balloon measurement. By taking into account Eqs.(1) and (2), we can obtain the atmospheric CO_2 concentration profile under the assumption that some parameters are independent of wavelength and so can be considered as constant.

3. The new method for evaluating the detecting capability of Raman lidar

Lidar designs vary widely depending on the specific application and various hardware components. To conduct quantitative tradeoff studies based on conventional methods for evaluating capability of lidar, a significant number of instrumental parameters and external environmental factors must be taken into account; however, it is often not clear how each system and environmental parameters may quantitatively influence the ultimate performance by traditional method. In order to specify the influence of different parameters on the capability of lidar, a new method for evaluating the capability of a general lidar system was founded by Agishev *et al*,[9] which introduces a reference test object as a key element for dimensionless parametrization. A decomposition of the total SNR into five dimensionless parameters representing the transmitter and receiver conditions, background noise, target efficiency, atmospheric conditions, and a scale length has been carried out. The SNR relation can be expressed as

$$\Psi_{\mathrm{X}} = VQ_{\mathrm{X}}W^2U^{-1}r^{-2}. \tag{3}$$

The subscript X represents different types of lidar, such as DIAL, Raman, Rayleigh scattering etc. In this paper, we just analyse the Raman lidar capability for atmospheric CO_2 measurement. V is defined as the ratio of the echo signal power P_{s0} received from the reference range R_0 for the reference atmosphere, to the threshold power P_{t0} in the absence of background noise.

$$\begin{aligned} P_{\mathrm{s0}}(\lambda_0, R_0) &= \frac{1}{2}c\tau_{\mathrm{p}}\beta_{\pi 0}(\lambda_0, \alpha_0)AR_0^2P_0(\lambda_0)\xi(\lambda_0)T_0^2(\lambda_0, R_0), \\ P_{\mathrm{t0}} = P_{\mathrm{t}}^{\mathrm{B}=0} &= \frac{1}{2}\rho_{\mathrm{out}}^2P_{\mathrm{q}}(1+\sqrt{1+(4/\rho_{\mathrm{out}}^2)P_{\mathrm{n}}^2/P_{\mathrm{q}}^2}), \end{aligned} \tag{4}$$

where ρ_{out} is a given SNR at the photon detector output, P_{q} characterizes the quantum limit of the detector sensitivity or the threshold power. P_{n} is the power of the internal noise of the photon detector referred to as the photon detector input.

The physical signification of V is the effect of transmitter and receiver operation on a reference atmosphere according to the definition. The V-parameter is only defined for single-shot operation. However, it is essential to make shot averaging. When accumulating N lidar signals with repetition frequency f_{mod} over a time of observation T_{obs}, an increase in the SNR of $N^{1/2}$ occurs. In this case, the equivalent system parameter V^* can be defined as

$$V^* = V\sqrt{N} = V\sqrt{f_{\mathrm{mod}}T_{\mathrm{obs}}}. \tag{5}$$

Parameter Q_{X} of Raman lidar can be expressed as follows

$$Q_{\mathrm{Ram}}(\lambda_{\mathrm{R}}, \lambda_0) = \frac{\beta_{\pi\mathrm{R}}(\lambda_{\mathrm{R}})}{\beta_{\pi 0}(\lambda_0)} = \frac{N_{\mathrm{g}}\sigma_{\mathrm{R}}(\lambda_{\mathrm{R}})}{N_{\mathrm{m}}\sigma_{\mathrm{m}}(\lambda_0)}. \tag{6}$$

Parameter Q_{Ram} simply describes the backscatter efficiency of an arbitrary species relative to the molecular reference.

The third factor in Eq.(3) is a normalized atmospheric component W^2, which is determined by the transparency ratio of the atmosphere state and the standard molecular atmosphere. The normalized transparency along the sensing path for Raman lidars can be given by

$$\begin{aligned} W_{\mathrm{Ram}} =& \frac{\sqrt{T_{\mathrm{fw}}(\lambda, r)T_{\mathrm{bw}}(\lambda_{\mathrm{R}}, r)}}{T_0(\lambda_0, R_0)} \\ =& \exp\bigg\{-\frac{1}{2}\int_0^r[\alpha_{\mathrm{a}}(r', \lambda) + \alpha_{\mathrm{m}}(r', \lambda) \\ &+ \alpha_{\mathrm{a}}(r', \lambda_{\mathrm{R}}) + \alpha_{\mathrm{m}}(r', \lambda_{\mathrm{R}})]\mathrm{d}r' \\ &+ \int_0^{\mathrm{R}_0}\alpha_{\mathrm{m}}(r', \lambda_0)\mathrm{d}r'\bigg\}. \end{aligned} \tag{7}$$

Parameter U is defined as the ratio of the photon detector threshold power P_{t} to P_{t0}, defined in the presence and absence of the background noise, respectively. When the lidar operates in the nighttime, the influence of background radiation on lidar detecting capability can be neglected and the value of parameter U is unity. The parameter r is the operation range of lidar detecting.

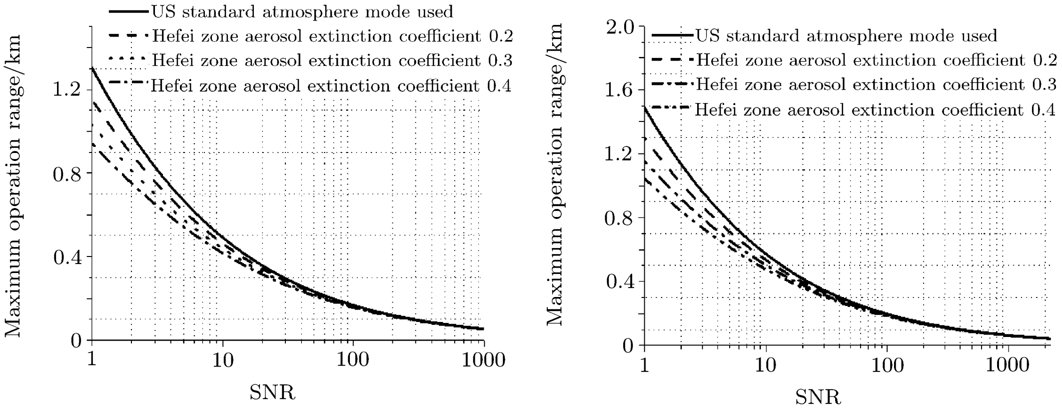
4. Evaluation of capability of Raman lidar for atmospheric CO_2 monitoring

The detecting capability of the Raman lidar includes two parts: the maximum operation range and the minimum detectable gas concentration, which can be evaluated by the above new method. According to Eqs.(4), due to variations of lidar instrumental parameters for different types of lidar (P_0, A, ξ, R_0, noise-equivalent power (NEP), f), the value of parameter V can change dramatically from 10^{-10} to 10^7. In the Raman lidar system for atmospheric CO_2 monitoring, the parameter V can reach about 7×10^5 by calculation. The value of parameter Q_X for Raman lidar ranges from 10^{-5} to $10^{-3}(N_2)$. In this Raman lidar system, the value of Q_X reaches 10^{-7} normally. The parameter U can be considered as unity when the lidar is operated in the nighttime, while parameter W indicates the influence of weather condition on the detecting capability of Raman lidar. The range of W parameter can vary dramatically for different conditions. Detailed analysis and numerical simulations of influence of parameter W on lidar capability will be presented in the following sections.

4.1. Maximum operation range of horizontal Raman lidar

Atmosphere extinction effect includes molecular and aerosol contributions, the latter changes more quickly than the former because aerosol concentration and dispersion are often changed irregularly. To determine the aerosol extinction exactly, we often need a knowledge of the exact nature of the aerosols that are responsible for the extinction.[12,13] Many researches were focused on the determination of aerosol extinction coefficient and aerosol size distribution.[14−16] In order to simulate the influence of weather conditions on the capability, different weather conditions have to be taken into account. The characteristic of atmosphere extinction can be considered as homogeneous when the Raman lidar system is operated in the horizontal direction. In this paper, the standard atmosphere mode of middle latitude in United States and the atmosphere extinction mode in Hefei are used to conduct the simulation.

Simulation is conducted at the middle latitude of standard atmosphere as in US, with Hefei aerosol extinction coefficient being 0.2, 0.3 and 0.4. The Raman lidar is operated in the horizontal direction. The graph (a) indicates the Raman lidar operated at 20000 shots accumulated, and (b) at 40000 shots. We can conclude that the maximum operation range of Raman lidar is influenced significantly by the weather conditions. The maximum difference between operation ranges can reach 0.4 and 0.5km due to different conditions for accumulating mode as shown in Fig.1. Normally, atmospheric CO_2 concentration, or the



(a) at 20000 shots

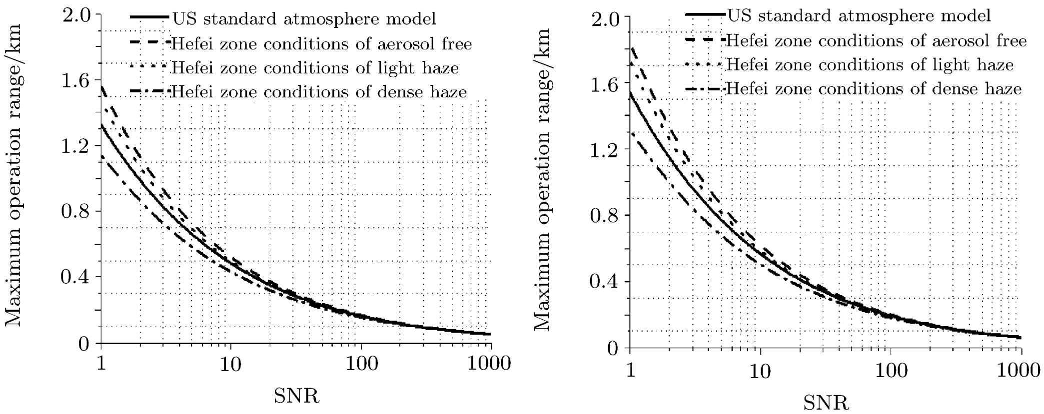
(b) at 40000 shots

Fig.1. Maximum operation range of Raman lidar operated in the horizontal direction as a function of SNR under different weather conditions in the absence of background radiation.

mixing ratio can reach 300ppmv. It should be noted that the minimum detectable concentration in Fig.1 is within 30ppmv. For a given lidar system, with the increase of the value of minimum detectable CO_2 concentration, a larger maximum operation range can be reached, while the measurement accuracy would be reduced.

4.2. Maximum operation range of vertical Raman lidar

The simulated results shown in Figs.1 and 2 allow a quantitative estimation of the influence of weather conditions and signal averaging tradeoffs. The maximum operation range in vertical direction is larger than in horizontal direction because at low altitude in the atmosphere a mass of aerosol is concentrated. It should be noted that the minimum detectable concentration obtained from Fig.2 is within 30ppmv. In Fig.2 simulation is conducted at the middle latitude of standard atmosphere in US, and Hefei conditions of light and dense haze. The graph (a) is obtained for the Raman lidar operated at 20000 shots accumulated, while (b) at 40000 shots. Introducing an ideal condition that at low altitude there has no aerosol concentration, where extinction effect is caused just by molecules, we have the maximum operation range under the conditions assumed can reach the high-point as shown in Fig.2. Therefore, the aerosol concentration or extinction characteristic is the principal factor influencing the maximum operation range for a given lidar system.

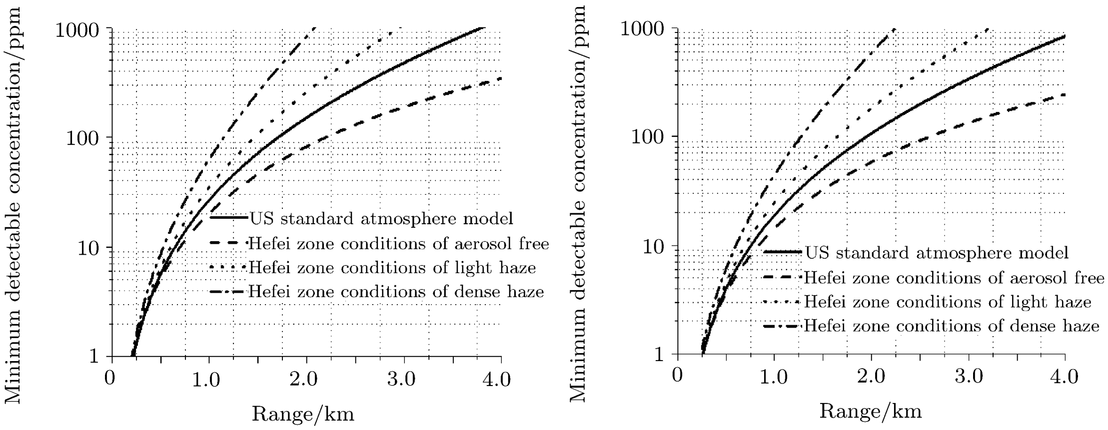


(a) at 20000 shots (b) at 40000 shots

Fig.2. Maximum operation range of Raman lidar operated in the vertical direction as a function of SNR under different weather conditions in the absence of background radiation.

4.3. Minimum detectable concentration by using horizontal Raman lidar

Minimum detectable gas concentration is a key factor for a lidar system. In the following sections, the new method mentioned above will be applied to the estimation of the minimum detectable concentration of the atmospheric CO_2 using a Raman lidar system, under conditions of light haze, dense haze and US atmosphere mode. The minimum detectable concentration by the Raman lidar system is shown in Fig.3, with the Raman lidar is operated in the horizontal direction. Figure 3(a) is for the Raman lidar operated at 20000 shots accumulated, while 3(b) at 40000 shots. The simulation is carried out under the condition of neglecting the background radiation.

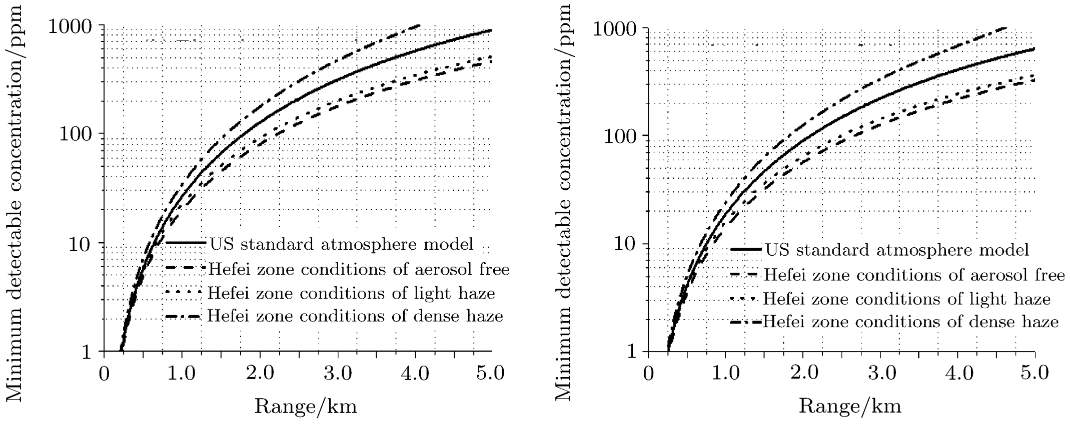


(a) at 20000 shots (b) at 40000 shots

Fig.3. Minimum detectable CO_2 concentration by Raman lidar operated in the horizontal direction as a function of W parameter under different weather conditions in absence of background radiation.

4.4. Minimum detectable concentration by using vertical Raman lidar

An obvious difference in minimum detectable concentrations can be observed from Figs.3 and 4, primarily due to the aerosol scattering components at low altitude. Simulation is conducted at the middle latitude of standard atmosphere of US, and Hefei conditions of light and dense haze etc. In Fig.4 the Raman lidar is operated in vertical direction, with Fig.4(a) for the Raman lidar operated at 20000 shots accumulated, and 4(b) at 40000 shots. When the aerosol scattering extinction is removed and the molecular scattering is examined, the difference in minimum detectable concentrations can be neglected as shown in Fig.5. The relative error is within 5% from the ground to 3km altitude. The aerosol scattering extinction is important for determining the minimum detectable concentration, when a molecular profile analysis is applicable.



(a) at 20000 shots (b) at 40000 shots

Fig.4. Minimum detectable CO_2 concentration by Raman lidar operated in the vertical direction as a function of W parameter under different weather conditions in absence of background radiation.

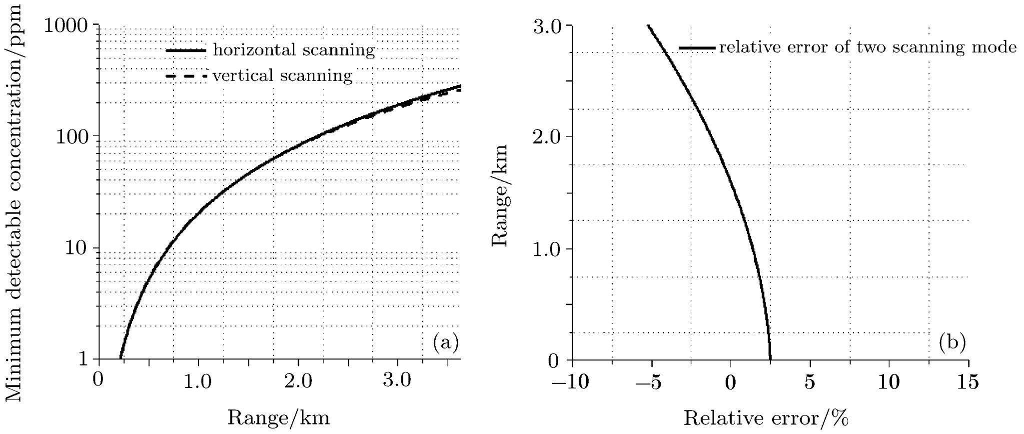


Fig.5. (a) Minimum detectable CO_2 concentration by Raman lidar under the conditions of neglecting the influence of aerosol scatting and background radiation in different operation directions. (b) The relative error between the two directions within the range of 3km.

5. Discussion and conclusions

In this paper, numerical simulation results are presented, obtained using a new method for evaluating the capability of a Raman lidar system in monitoring the concentration of atmospheric CO_2. The influences of weather conditions on the capability of the Raman lidar are described for horizontal and vertical directions. It is easy to see that the capability of the Raman lidar under 'good' or appropriate conditions is high. For a Raman lidar system, the maximum difference of the operation ranges caused by weather conditions alone can be about 0.4 to 0.5km if other parameters are kept constant, and the minimum detectable concentration is 30ppmv. The range of minimum detectable concentration caused by the weather conditions alone can reach about 20 to 60ppmv in horizontal direction at 20000 shots at a distance of 1km. Some other typical parameters of the Raman lidar are shown in Table 1. It can be concluded that the capability of the Raman lidar operated in vertical direction is better than that in horizontal direction due to the distribution of aerosol in lower atmosphere.

Table 1. Capability of the Raman lidar under different weather conditions obtained by simulation.

		horizontal direction		vertical direction	
pulse number		20000	40000	20000	40000
max difference of operation range/km		0.4	0.5	0.4	0.5
the range of min detectable concentration/ppmv	1.0km	20–60	15–45	20–35	14–25
	1.5km	45–260	30–180	45–90	30–60

Figure 6 shows the capability of the Raman lidar under weather conditions of light and dense haze. The stability of the former is better than the latter, which are shown by dashed and solid lines respectively. We believe that simple qualitative design issues such as the use of a more powerful laser transmitter, a larger aperture of receiving telescope, and a more sensitive photon detector will obviously be helpful for achieving a greater operation range and better retrieval accuracy.

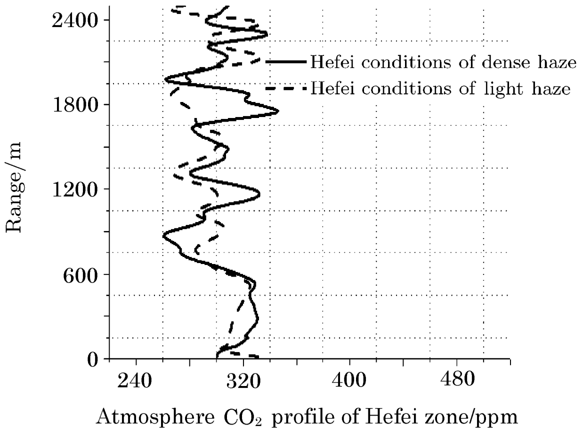


Fig.6. Atmospheric CO_2 profile in Hefei under different conditions: light haze and dense haze.

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