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Evaluation of dual differential absorption lidar based on Raman-shifted Nd:YAG or KrF laser for tropospheric ozone measurements

Z. Wang, J. Zhou, H. Hu, Z. Gong

Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, P. O. Box 1125, Hefei, Anhui 230031, People's Republic of China
(Fax: + 86-551/5591572, E-mail: HLHU@Sun.ihep.ac.cn)

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Abstract. Based on Raman-shifted Nd:YAG or KrF laser, a method of three-wavelength Dual DIfferential Absorption Lidar (DIAL) for tropospheric ozone measurements is proposed. A theoretical analysis and numerical simulations of the measurement error have been performed. The results show that this method can reduce the error in ozone measurements caused by the aerosol layer in the troposphere by a factor of ten. The proposed method is also shown to be insensitive to aerosol optical properties, and therefore, one does not need to know the wavelength dependence of aerosol scattering. The dual-DIAL with 277.1, 291.8, 313.2 nm radiation based on a Raman-shifted KrF laser can be used both during day- and night-time. The dual-DIAL with 289.0, 299.1, 316.1 nm radiation based on Raman-shifted Nd:YAG laser can only be used during night-time.

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A number of DIfferential Absorption Lidar (DIAL) systems have been developed to measure tropospheric ozone. Their DIAL wavelengths are produced by using flash-lamp/or laser-pumped dye lasers, Raman-shifted KrF (248.4nm) excimer lasers with H_2 and D_2 cells, or Raman-shifted quadrupled Nd:YAG (266 nm) lasers with H_2 and D_2 or D_2 and DH cells [1–3]. These wavelengths coincide with regions of high absorption in ozone. Because the intense Hartley band of ozone is broad, relatively large wavelength separations are required to provide adequate differences in absorption coefficients. A main problem in retrieving ozone concentrations with DIAL lies in the unknown wavelength dependence of the aerosol extinction and backscattering coefficient as spatially inhomogeneous or high loading aerosol regions may exist in the low troposphere. Under these circumstances, aerosol causes large measurement errors in the determination of ozone profiles. Several correction methods have been proposed [4–7] for remedy; they all need to make assumptions about the wavelength dependence of aerosol backscattering and extinction coefficients, and one needs to know the spatial distribution of the aerosols. As it is impossible to obtain these parameters accurately, these correction methods cannot solve this problem satisfactorily.

In this paper, based on the Raman-shifted Nd:YAG or KrF laser, a three-wavelength dual-DIAL method for tropospheric ozone measurements is proposed. Theoretical analysis and numerical simulation results show that this method can greatly reduce the effect of aerosol on tropospheric ozone measurements. It is also found to be insensitive to the aerosol optical properties. Therefore, it does not need the knowledge of aerosol optical properties, such as wavelength dependence in scattering.

1 Three-wavelength dual-DIAL method

1.1 Dual-DIAL equation

As proposed earlier, the dual-DIAL system used two pairs of wavelengths $(\lambda_{1on}, \lambda_{1off})$ and $(\lambda_{2on}, \lambda_{2off})$ [8]. However, if we let $\lambda_{1off} = \lambda_{2on}$, only three wavelengths are needed; but it is difficult to select three useful wavelengths to satisfy the condition that $\lambda_{1on} - \lambda_{1off}$ approximately equals $\lambda_{2on} - \lambda_{2off}$. To overcome this difficulty, we can introduce a constant C to keep the condition that $\lambda_{1on} - \lambda_{1off}$ approximately equals $C(\lambda_{2on} - \lambda_{2off})$. Then, the number density of ozone molecules at a range z, $N(z)$, can be written as

$$N(z) = \frac{1}{2\Delta\delta(T)}\left[\frac{d}{dz}\left(-\ln\frac{P_{\lambda_{1on}}(z)}{P_{\lambda_{1off}}(z)} + C\ln\frac{P_{\lambda_{2on}}(z)}{P_{\lambda_{off}}(2)}\right) + B' + E'\right], \quad (1)$$

$$\Delta\delta(T) = [\delta_{\lambda 1on}(T) - \delta_{\lambda 1off}(T)] - C[\delta_{\lambda 2on}(T) - \delta_{\lambda 2off}(T)], \quad (2)$$

$$B' = B_1 - CB_2, E' = E_1 - CE_2, \quad (3)$$

$$B_i = \frac{d}{dz} \ln \frac{\beta_{\lambda i\text{on}}(z)}{\beta_{\lambda i\text{off}}(z)}, i = 1, 2, \tag{4}$$

$$E_i = -2[\alpha_{\lambda i\text{on}}(z) - \alpha_{\lambda i\text{off}}(z)], i = 1, 2, \tag{5}$$

where $\lambda_{1\text{on}}, \lambda_{2\text{on}} = \lambda_{1\text{off}}, \lambda_{2\text{off}}$ are the laser wavelengths corresponding to high, medium, and low absorption of ozone, respectively; $\delta(T)$ is the temperature-dependent absorption cross section per ozone molecule, T the temperature, $P(z)$ the lidar return-signal power or photoelectron number from range z, $\beta(z)$ the total volume backscatter coefficient due to air molecules and aerosols at range z, and $\alpha(z)$ the total extinction coefficient minus ozone absorption coefficient at range z; $\alpha(z) = \alpha_a(z) + \alpha_m(z)$, $\alpha_a(z)$, $\alpha_m(z)$ is extinction coefficient of aerosol and molecule, respectively. The constant C is introduced to minimize B' and E', i.e., to minimize the effect of aerosols. For a certain aerosol and atmospheric condition, there is a optimal constant C. In the real atmosphere, aerosol and atomospheric conditions vary in time and space. So there is not a optimal constant C for all these conditions. But we can chose a average constant C for all these conditions. For a given condition, perhaps, C is not the optimal value, but this does not cause a serious effect on dual-DIAL. This point will be discussed detail in Sect. 2. The average constant C can be chosen somewhat smaller than $(\lambda_{1\text{on}} - \lambda_{1\text{off}})/(\lambda_{2\text{on}} - \lambda_{2\text{off}})$. For example, C can be chosen in the region of 0.65–0.67 for wavelengths of 277.1, 291.8, and 313.2 nm [(277.1 − 291.8)/(291.8 − 313.2) = 0.687]. Chosen wavelengths and C should also meet the following conditions (*i*) $\Delta\delta(T)$ is large enough to meet the measurement sensitivity and spatial resolution; (*ii*) the separation of the two pairs of wavelengths is not too large.

Compared with the B_1 and B_2, E_1 and E_2, B' and E' in (1) can be neglected or only be taken into account for air molecular extinction in E', which can be estimated from the profile of the atmospheric molecules. In this way, the effect of aerosols on ozone measurements will be greatly minimized. The number density of ozone can be obtained directly from the lidar return signal.

1.2 Measurement error of the three-wavelength dual-DIAL system

The measurement error of the three-wavelength dual-DIAL can be estimated by a method similar to Schotland's [9]. The measurement error comes from two sources: the statistical error in photon counting, and the system error due to the wavelength dependence of Rayleigh and Mie scattering.

The former error ε_r is calculated by

$$\varepsilon_r = \frac{1}{2\Delta\delta N(z)\Delta z}\left(\sum_j \frac{P_{1j}(z) + P_{b1j}}{P_{1j}(z)^2} + \frac{P_{1j}(z+\Delta z) + P_{b1j}}{P_{1j}(z+\Delta z)^2} + C\frac{P_{2j}(z) + P_{b2j}}{P_{2j}(z)^2} + C\frac{P_{2j}(z+\Delta z) + P_{b2j}}{P_{2j}(z+\Delta z)^2}\right)^{1/2} \tag{6}$$

j = on, off,

where P_{bi} includes the sky background signal and thermal noise.

The latter system error ε_s can be calculated from the profiles of atmospheric molecules and aerosols:

$$\varepsilon_s = \varepsilon_{sB} + \varepsilon_{sE} \tag{7}$$

$$\varepsilon_{sB} = \frac{1}{2\Delta\delta N(z)\Delta z}\left(\ln\frac{\beta_{\lambda 1\text{on}}(z+\Delta z)}{\beta_{\lambda 1\text{on}}(z)} - \ln\frac{\beta_{\lambda 1\text{off}}(z+\Delta z)}{\beta_{\lambda 1\text{off}}(z)} - C\ln\frac{\beta_{\lambda 2\text{on}}(z+\Delta z)}{\beta_{\lambda 2\text{on}}(z)} + C\ln\frac{\beta_{\lambda 2\text{off}}(z+\Delta z)}{\beta_{\lambda 2\text{off}}(z)}\right), \tag{8}$$

$$\varepsilon_{sE} = \frac{E'}{2\Delta\delta N(z)} = \varepsilon_{s\text{Emol}} + \varepsilon_{s\text{Eaer}}, \tag{9}$$

$$\varepsilon_{s\text{Emol}} = \frac{-1}{\Delta\delta N(z)}\{[\alpha_{m\lambda 1\text{on}}(z) - \alpha_{m\lambda 1\text{off}}(z)] - C[\alpha_{m\lambda 2\text{on}}(z) - \alpha_{m\lambda 2\text{off}}(z)]\}, \tag{10}$$

$$\varepsilon_{s\text{Eaer}} = \frac{-1}{\Delta\delta N(z)}\{[\alpha_{a\lambda 1\text{on}}(z) - \alpha_{a\lambda 1\text{off}}(z)] - C[\alpha_{a\lambda 2\text{on}}(z) - \alpha_{a\lambda 2\text{off}}(z)]\}, \tag{11}$$

where ε_{sB}, ε_{sE} are contributing due to total backscattering and extinction, respectively; $\varepsilon_{s\text{Emol}}$, $\varepsilon_{s\text{Eaer}}$ are the contribution due to molecular and aerosol extinction, respectively.

2 Numerical simulations

For tropospheric ozone measurements, there are many suitable laser wavelengths. For example, they can be obtained from fundamental or harmonics of standard lasers or their low-order Stokes-Raman-shifted (SRS) wavelengths [1–3]. The available wavelengths in the UV spectral range are listed in Table 1. It can be seen, based on the Raman-shifted KrF or Nd:YAG laser, 277.1, 291.8, 313.2, and 289.0, 299.1, 316.1 nm can be selected as two groups of wavelengths for the three-wavelength dual-DIAL system. The corresponding constant C can be chosen as 0.67 and 0.57, respectively.

To validate the capability of the three-wavelength dual-DIAL system for tropospheric ozone measurements, the statistical error in photon counting and the system error due to aerosols are estimated by numerical simulation with the lidar equation. For comparison, the measurement error of conventional DIAL with 277.1, 291.8 nm and 289.0, 299.1 nm is also estimated. In the

Table 1. Available wavelengths in the UV region

Laser	Wavelength [nm]
KrF	248
SRS H_2	277.1, 313.2, 360.1
SRS D_2	268.4, 291.8, 319.3
SRS $H_2 + D_2$	302
Nd:YAG	
Third harmonic	355
Fourth harmonic	266
SRS (266) H_2	299.1, 341.5, 397.5
SRS (266) D_2	289.0, 316.1, 349.2
SRS (266) HD	294.2
XeBr	282

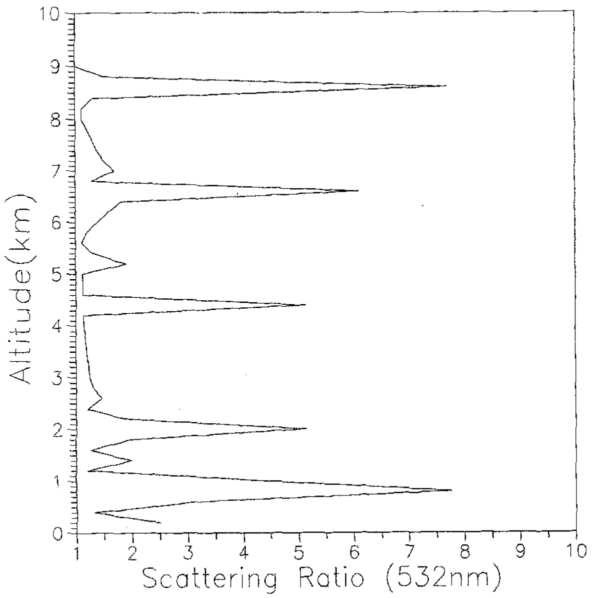

Fig. 1. Profile of the aerosol scattering ratio (532 nm)

Table 2. System parameters used for simulation

Laser wavelength [nm]	277.1 289.0 291.8 299.1 302 313.2 316.1
Laser pulse energy [mJ]	30 for all: wavelengths
Sky background radiation: [W/m^{2}sr nm]	0, 5.0×10^{-7}, 3.67×10^{-5}, 5.1×10^{-4}, 1.1×10^{-3}, 2.14×10^{-2}, 2.71×10^{-2}
Area of receiver [m^2]	0.086
Filter bandwidth [nm]	1.7
Field of view [mrad]	1.5
PMT quantum efficiency [%]	25
Total optical efficiency [%]	30

simulation, the molecular model of the atmosphere is the Elterman model [10]. An aerosol profile having several spatially inhomogeneous aerosol layers is used as shown in Fig. 1. The wavelength dependence of aerosol exinction and backscattering, $\lambda^{-\eta}$ are assumed to have $\eta = 1.0$ and 1.5, respectively. The ozone model profile used is the USA standard model [11]. The simulated lidar system is assumed to operate in the nadir and zenith direction from an aircraft flying at 5 km altitude, in the zenith direction from ground, and in a nadir direction from an aircraft flying at 10 km altitude. The lidar system parameters for simulation are shown in Table 2.

Figures 2–4 show the results of the numerical simulation. In these figures, the statistical errors are drawn as dashed lines, the total measurement errors (including the statistical error and system error due to aerosols) are drawn as solid lines and the dotted lines give the measurement errors for the conventional DIAL.

From these figures, it can be seen that the measurement error depends on the selected wavelength and the measurement time. During day-time, although the dual-DIAL signal has a larger statistical error than the conventional DIAL signal due to the sky background noise and the longer wavelength used in the dual-DIAL, its total

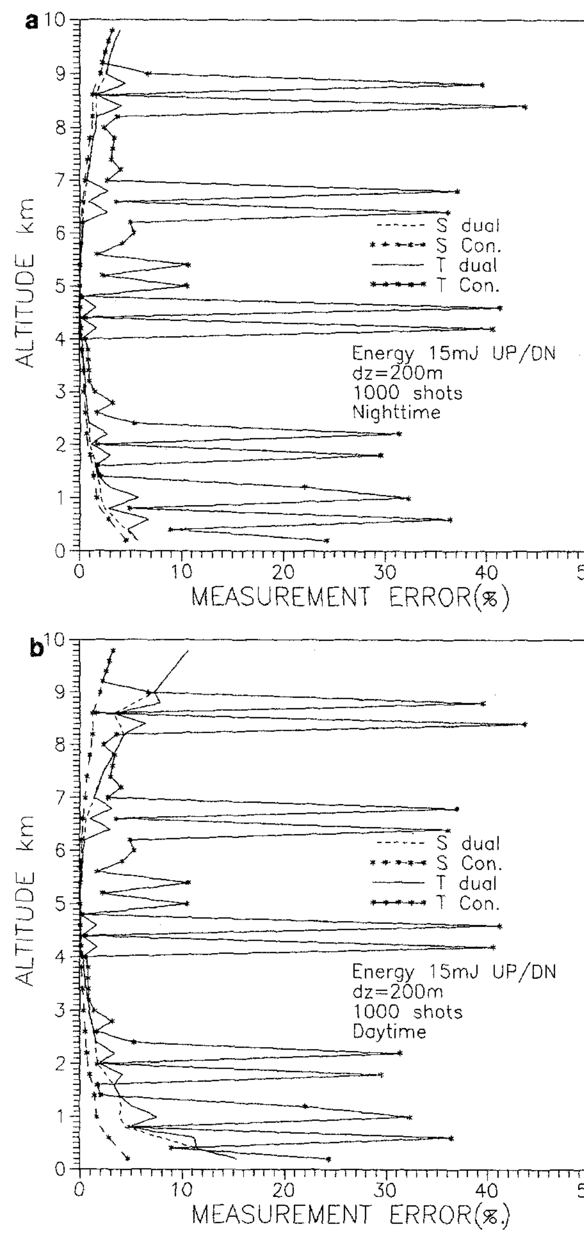

Fig. 2a, b. Simulations of measurement error vs altitude for the dual-DIAL with 277.1, 291.8, 313.2 nm and conventional DIAL with 277.1, 291.8 nm operated from an aircraft flying at an altitude of 5 km. The ozone profile is the US standard ozone profile. In the legend, *S* represents the statistical error, *T* the total measurement error, *dual* the dual-DIAL method and *con.* the conventional DIAL method: (**a**) night-time; (**b**) day-time

measurement error is smaller (Figs. 2, 3b). For the same reason, the dual-DIAL based on the KrF laser is better than that based on the Nd:YAG laser (Figs. 2, 3b). During night-time, the difference in the statistical error between the dual-DIAL and convential DIAL is very small (Figs. 2, 3a). The dual-DIAL based on the Nd:YAG laser (Fig. 3a) can yield the same measurement accuracy as the one based on the KrF laser (Fig. 2a).

In the region without the serious influence of aerosols, both dual-DIAL and conventional DIAL can yield accurate ozone profiles. However, in the spatially inhomogeneous or high loading aerosol layer, a larger system

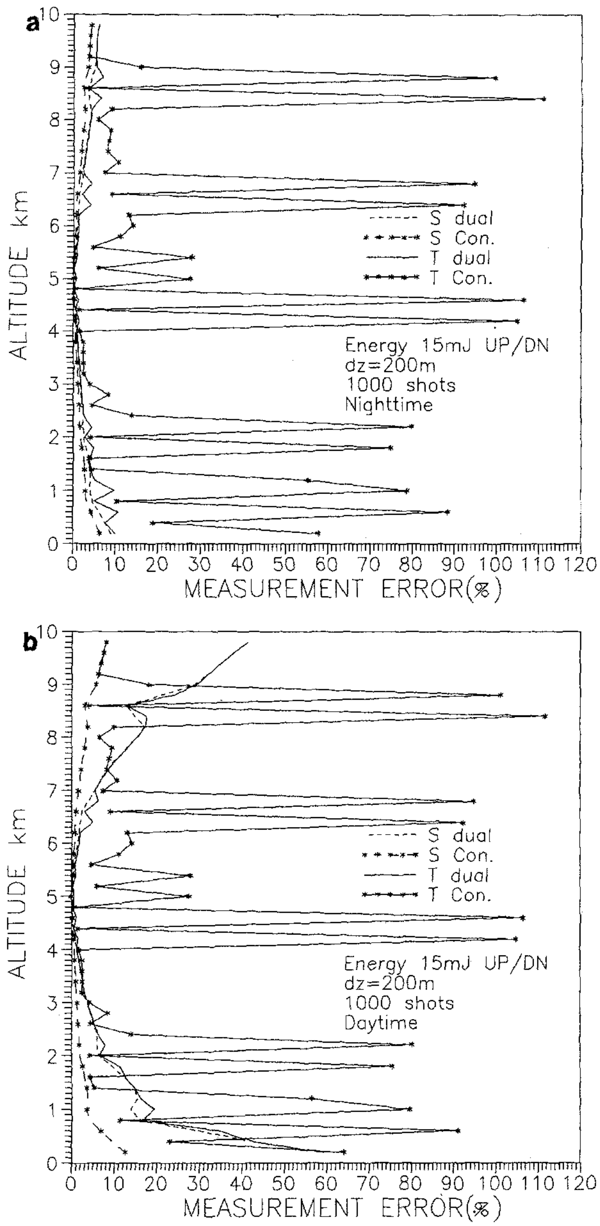

Fig. 3a,b. Simulations of measurement error VS altitude for the dual-DIAL with 289.0, 299.1, 316.1 nm and conventional DIAL with 289.0, 299.1 nm operated from an aircraft flying at an altitude of 5 km. The ozone profile is the USA standard ozone profile. (**a**) night-time; (**b**) day-time (other notations as in Fig. 2)

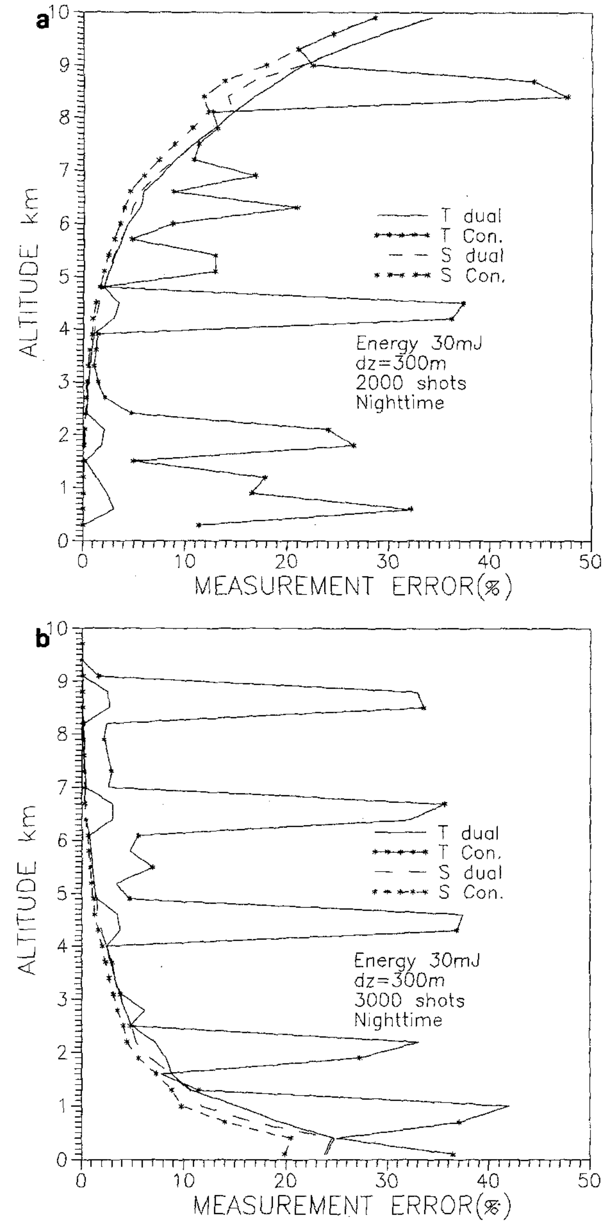

Fig. 4a, b. Same as Fig. 2 except the lidar is operated at: (**a**) ground; (**b**) an altitude of 10 km above ground

error is introduced in the conventional DIAL (Figs. 2–4); sometimes it can be over 100%. But, the effect of these aerosol layers on the dual-DIAL is much smaller and its measurement error is reduced by five-to ten-fold over the conventional DIAL, even during day-time.

Comparing the measurement results of systems operated at different altitudes, it seems that one good way to measure tropospheric ozone is to operate the system in the nadir and zenith direction from an aircraft flying at 5 km altitude. The ground-based system operated in zenith direction has to increase the measurement time to improve the measurement accuracy at higher altitude.

In the numerical simulation, calculations were also made to investigate the sensitivity of the dual-DIAL to aerosol optical properties and the constant C. In the calculations, the spatial resolution is 0.1 km, and the backscatter ratio $R_{\lambda 2\mathrm{off}}(z)$ equals 1.0. In Fig. 5, the terms of $N(z)\varepsilon_{\mathrm{sB}}$ for the three-wavelength dual-DIAL with 277.1, 291.8, 313.2 nm and conventional DIAL with 277.1 and 291.8 nm are plotted as a function of aerosol scattering ratio $R_{\lambda 2\mathrm{off}}(z+\Delta z)$ for 4 values of the wavelength dependence of aerosol backscattering $\eta(\beta(\lambda) \propto \lambda^{-\eta})$. It is apparent that, compared with conventional DIAL, the three-wavelength dual-DIAL minimizes the effect of aerosols on the ozone profile more than ten times and $\varepsilon_{\mathrm{sB}}$ also change slightly with the change of η and $R_{\lambda 2\mathrm{off}}(z+\Delta z)$. So the ozone profile obtained by dual-DIAL measurements is insenstive to the inhomogeneity of aerosols and the aerosol optical properties. In Fig. 6, the term of $N(z)\varepsilon_{\mathrm{sB}}$ for the three-wavelength dual-DIAL with 277.1, 291.8, 313.2 nm

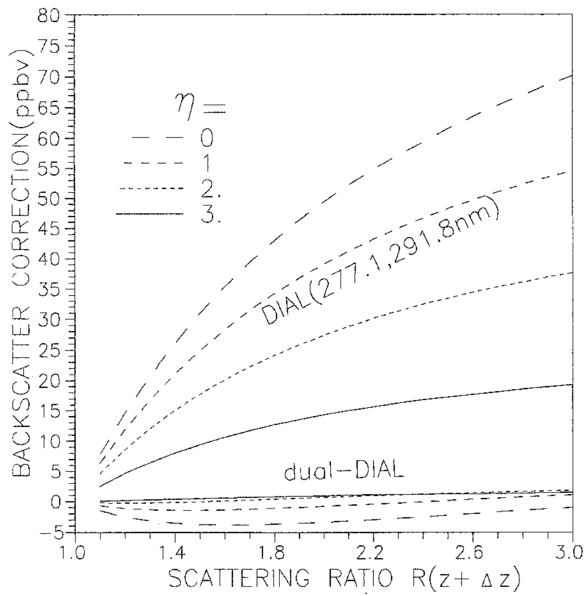

Fig. 5. Backscatter correction $N(z)\varepsilon_{sB}$ in the dual-DIAL with 277.1, 291.8, 313.2 nm and conventional DIAL with 277.1, 291.8 nm as a function of scattering ratio $R_{\lambda 2\text{off}}(z + \Delta z)$ for 4 values of wavelength dependence of aerosol backscatter, $R_{\lambda 2\text{off}}(z)$ equals 1.0

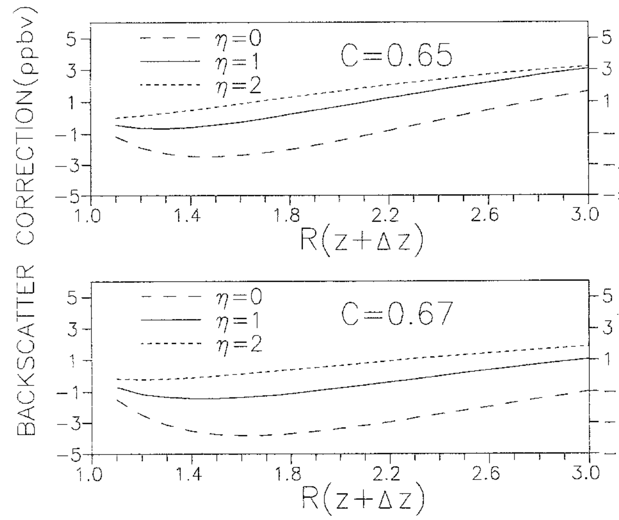

Fig. 6. $N(z)\varepsilon_{sB}$ of dual-DIAL with 277.1, 291.8, 313.2 nm as a function of aerosol scattering ratio $R_{\lambda 2\text{off}}(z + \Delta z)$ for different C and η

is plotted as a function of aerosol scattering ratio $R_{\lambda 2\text{off}}(z + \Delta z)$ for different C and η. Figure 6 shows that the effect of aerosols changes slightly with the change of C in the used region of C. As the effect of aerosols on dual-DIAL is insensitive to the variation (near the chosen C) of C, the constant C for a group of wavelengths can be easily chosen.

3 Conclusion

We have presented a three-wavelength dual-DIAL method based on the Raman-shifted Nd:YAG or KrF laser. Theoretical analysis and numerical simulation results show that this method can greatly reduce the effect of aerosols on ozone measurements in the troposphere and it is also shown to be insensitive to aerosol optical properties. Therefore, it does not need aerosol profiles and make any other assumption about the aerosol optical properties which are usually difficult to obtain. The ozone profile can be obtained directly from the lidar return signals.

For practical use, two groups of laser wavelengths, 277.1, 291.8, 313.2 and 289.0, 299.1, 316.1 nm, have attractive advantages. These wavelengths can be produced by Raman shifting of the KrF laser or Nd:YAG laser.

When the above-mentioned three laser wavelengths are simultaneously transmitted into the atmosphere to measure tropospheric ozone, the spatially inhomogeneous or high loading aerosol layer can be found by using the Bernoulli method [4]. In these layers, the dual-DIAL method is used to process the experimental data. In the other region, the conventional DIAL method can still be used for it has a small statistical error. In this way, the accurate ozone profile can be obtained in the whole troposphere.

The dual-DIAL based on the KrF laser can be used during both day- and night-time except for low ozone-density distribution during day-time. The dual-DIAL based on the Nd:YAG laser can be used during night-time. Of course, in addition to the above two groups of wavelengths, depending on the available system, other groups of wavelengths can also be chosen for measuring tropospheric ozone.

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References

1. U.N. Singhm Z. Chu, R. Mahon, T.D. Wilkerson: Appl. Opt. **29**, 1730 (1990)
2. W.B. Grant, E.V. Browell, N.S. Higdon, S. Ismail: Appl. Opt. **30**, 2628 (1991)
3. D.A. Haner, I.S. Mcdermid: IEEE J.QE-**26**, 1292 (990)
4. E.V. Browell, S. Ismail, S.T. Shipley: Appl. Opt. **24**, 2827 (1985)
5. W. Steinbrecht, A.I. Carswell: J. Geophys. Res. **100**, 1215 (1995)
6. A. Papayannis, G. Ancellet, J. Pelon, G. Megie: Appl. Opt. **29**, 467 (1990)
7. V.A. Kovalev, J.L. McElroy: Appl. Opt. **33**, 8393 (1994)
8. Z. Wang, H. Hu, J. Zhou: In *Proc. 17th Intl Laser Radar Conference*, Sendai, Japan (1994) p. 54
9. R.M. Schotland: J. Appl. Meteorol. **13**, 71 (1974)
10. L. Elterman: *UV, Visible, and IR Attenuation for Altitudes to 50 km*, Report AFCRL-68-0153 (AFCRL, Bedford, MA (1968)
11. A.J. Krueger, R.A. Minzner: J. Geophys. Res. **81**, 4477 (1976)