Monitoring Cl2 using a differential absorption lidar system

Edner, Hans; Fredriksson, K; Sunesson, A; Wendt, W

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Monitoring Cl\textsubscript{2} using a differential absorption lidar system

Hans Edner, Kent Fredriksson, Anders Sunesson, and Wilhelm Wendt

Lund Institute of Technology, Physics Department, P.O. Box 118, S-221 00 Lund, Sweden.
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The single-ended differential absorption lidar (DIAL) technique has proved to be a powerful method for remote measurements of source emissions and ambient air quality.\textsuperscript{1,2} DIAL systems working in the UV and visible spectral regions are often based on Nd: YAG and dye laser technology. The readily accessible molecules with such a system are H\textsubscript{2}O, NO\textsubscript{2}, SO\textsubscript{2}, and O\textsubscript{3}. In this Letter we report on the use of a DIAL system to test the remote sensing of Cl\textsubscript{2}.

The gaseous forms of chlorine most frequently encountered in polluted atmospheres include Cl\textsubscript{2}, HCl, and the tered in polluted atmospheres include Cl\textsubscript{2}. In this area other molecules, especially SO\textsubscript{2}, have absorption significant fine structure could be observed. Unfortunately, in this area other molecules, especially SO\textsubscript{2}, have absorption bands. The absorption of SO\textsubscript{2} can be considerable, which means that care has to be taken to choose wavelengths with equal absorption cross sections for SO\textsubscript{2} if both gases are encountered in the atmosphere. This was not a problem in the present measurements on a simulated Cl\textsubscript{2} source.

The remote monitoring of Cl\textsubscript{2} was performed at an open site. To simulate a source an open-ended box of plywood, 200 x 50 x 50 cm\textsuperscript{3}, was constructed. The box was filled with gas from a chlorine tube, and the laser beam was directed through it. This constituted a way of simulating a chlorine plume without spreading large amounts of chlorine in the atmosphere. During the measurement, the box was placed...
on the roof of a building 350 m away, and the beam was aimed through the end openings. The openings were about the same size as the beam diameter at this distance. Therefore, the alignment had to be very carefully performed so that no attenuation of the beam due to the box occurred. This was possible to check since there was ~500 m of free air behind the box before the beam hit the ground. To verify the alignment a DIAL measurement was performed without chlorine before each chlorine injection.

The measurements were performed in two different ways. First, a constant inflow of chlorine was established, and the measurement was performed. The gas flow was maintained during the measurement. Figure 2 shows a DIAL recording of this kind taken with a wavelength separation of 5.8 nm. The lidar signals are displayed together with the ratio curve. The peak in the lidar curves is due to scattering from the box. The curves are the average of 140 shots at each wavelength, and the chlorine concentration was calculated to be 1.8 g/m³. Similar recordings with other wavelengths and other absorption cross sections on the same chlorine flow showed good differential absorption because of the broad absorption profile. Experiments where the flow was stopped showed that the gas blew out of the box very quickly, normally within a few seconds.

Another experiment was performed on a spreading plume. A small Cl₂ flow was let out from the top of a tall tube. The plume was scanned vertically in different horizontal directions downwind from the tube to monitor the spread of the chlorine plume. The gas fell very rapidly to the ground, and the chlorine concentration was calculated to be 1.8 g/m³. The lidar signals are displayed together with the normalized ratio curve. The peak in the lidar curves is due to scattering from the box. The curves are the average of 140 shots at each wavelength, and the chlorine concentration was calculated to be 1.8 g/m³.

The sensitivity of a DIAL measurement on chlorine can be calculated from an estimated detectable differential absorption of half of a percent for distances up to 1 km. With the large separation of 10 nm this results in a sensitivity of 85 mg/m² corresponding to 170 μg/m³ with an absorption length of 2 x 250 m. This can be compared, for example, with the sensitivity for NO₂ monitoring, which is 10 mg/m² or 20 μg/m³ with a 2 x 250 m absorption path. The detection limit for chlorine could be lowered to 25 mg/m² if a larger wavelength separation is used. However, a large wavelength separation can cause problems. The difference in the atmospheric transmission may be so large that it gives rise to a concentration offset. In Cl₂ monitoring using the short-wavelength slope of the absorption profile the offset will be negative; i.e., the concentration is underestimated. A possible correction can be separated into a backscatter term and an extinction term. The backscatter correction will be large only for a case with strong variation in backscatter with range, e.g., a dense plume or cloud. As shown by Browell et al., it is possible to obtain the correction if the aerosol backscattering profile is calculated from the lidar return signal. It can be difficult to apply this method to the case where chlorine is present in industrial areas with interfering local particle plumes. The atmospheric extinction has to be corrected for in measurements of lower concentrations distributed over a large atmospheric volume. This correction can, according to Ref. 7, be divided into a molecular extinction correction and an aerosol extinction correction, where the molecular extinction is due to Rayleigh scattering. Using Elterman’s values the latter offset is calculated to be 0.32 mg/m³, independent of the meteorological conditions. The aerosol extinction coefficient varies with the visibility with a wavelength dependence of λ⁻α. Using α = 1 and Elterman’s values the coefficient can be estimated for different visibilities. The values will vary greatly. For a visibility of 10 km the aerosol extinction is 0.7 km⁻¹. For 4-km visibility it is 1.78 and 3.58 km⁻¹ when the visibility is down to 2 km. These extinction values yield the concentration offsets 0.36 mg/m³ (0.7 km⁻¹), 0.91 mg/m³ (1.79 km⁻¹), and 1.8 mg/m³ (3.58 km⁻¹) with a wavelength difference of 10 nm. Clearly there is great uncertainty involved in any attempt to estimate the offset. The best way to check the offset is probably to make a zero measurement before the lidar is pointed at the chlorine source. An offset should then be seen as a slope on the DIAL curve where it should be horizontal.

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References
Wind and $C_n^2$ profiling by single-star scintillation analysis: erratum

J. L. Caccia, M. Azouit, and J. Vernin

University of Nice, Astrophysics Department of the IMSP, Parc Valrose, 06034 Nice CEDEX, France.
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We recently noticed a printer's error in our paper: Eq. (1) should read

$$W(t, z) = 1.54 \lambda^{-2}(f_1^2 + f_2^2)^{11/6} \times$$

$$\int_{\lambda_{c}}^{\lambda_{c}+\lambda} C_{n}^{2}(h) \sin^{2}\pi\lambda t f_{1}^{2} + f_{2}^{2}) dh.$$  (1)

Reference


Use of LOWTRAN in transmission calculations

Daniel E. Novoseller

TRW, Inc., 1 Space Park, Redondo Beach, California 90278.
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Atmospheric transmission from ground to space may be calculated using the AFGL LOWTRAN 6 model as a function of height above sea level. The results are shown in Fig. 1 for a mid-latitude winter rural haze (5-km visibility) condition. The five curves correspond (from top to bottom) to five transmitter altitudes: 2, 1.5, 1, 0.5, and 0 km (sea level). This figure comes from a naive use of LOWTRAN and shows a severe altitude dependence. However, LOWTRAN treats the ground as being located at sea level. We have manipulated the LOWTRAN output, defining the transmission as

$$T(H) = T_0(H)T_A(0)/T_A(H),$$  (1)

where $T_0(H)$ = LOWTRAN value for transmission from $H$ to space;

$T_A(H)$ = LOWTRAN value for transmission due to aerosols from $H$ to space, and

$H$ = ground elevation above sea level.

Thus in effect we have defined the transmission from ground to space as $T(H) = T_M(H)T_A(0)$, where

$T_M(H)$ = LOWTRAN value for transmission due to molecular interactions from $H$ to space.

This is consistent with the statement that visibility is correlated to aerosol phenomena and that visibility conditions can be stated at any altitude with a similar aerosol content independent of altitude. (This ignores the effect of the humidity profile vs altitude, which is used by LOWTRAN in evaluating aerosol extinction.)

The results of the manipulation according to Eq. (1) are shown in Fig. 2 and show a far less severe dependence on ground elevation.

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