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NO plume mapping by laser-radar techniques

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Mapping of NO plumes by using laser-radar techniques has been demonstrated with a mobile differential absorption lidar system. The system was equipped with a narrow-linewidth Nd:YAG-pumped dye laser that, with doubling and mixing, generated pulse energies of 3–5 mJ at 226 nm, with a linewidth of 1 pm. This permitted range-resolved measurements of NO, with a range of about 500 m. The detection limit was estimated to 3 $\mu g/m^3$, with an integration interval of 350 m. Spectroscopic studies on the $\gamma(0, 0)$ bandhead near 226.8 nm were performed with 1-pm resolution, and the differential absorption cross section was determined to be $(6.6 \pm 0.6) \times 10^{-22}$ m^2, with a wavelength difference of 12 pm.

This Letter reports mapping of nitric oxide plumes and measurements of the ambient NO concentration using laser radar in the UV region. Path-integrated measurements made employing retroreflectors have been reported by our group.[1,2] Other such measurements of NO have been performed in the IR region using diode lasers[3,4] and CO and CO_2 lasers.[5,6] Stratospheric NO measurements have been performed using balloons.[7] Independent NO measurements in the UV region have also been performed by the National Physical Laboratory group in the United Kingdom.[8] They have also made simultaneous NO and NO_2 measurements employing two laser systems.

The nitrogen oxides NO and NO_2 (NO_x) constitute an increasing environmental problem.[9,10] NO_x compounds are emitted from industrial processes and internal combustion engines. The major part of NO_x emissions is NO, which is formed in high-temperature combustion processes. The sources are both atmospheric nitrogen and nitrogen-containing compounds in the fuel. Atmospheric nitrogen is considered to be the most important source, and therefore it is difficult to reduce NO_x emissions in combustion processes. In the atmosphere, NO is oxidized to NO_2 in a process that is believed to be the major NO_2 source in the atmosphere. In photochemical smog situations, NO_x and several organic molecules are involved in the formation of ozone, peroxyacetylnitrate, and other secondary pollutants. In the normal background atmosphere, NO and NO_2 together control the formation of ozone. Nitrogen oxides contribute to acidification and forest damage through acid rain, normally in the form of HNO_3. Furthermore, recent studies suggest that NO_x may enhance ozone-mediated leaf injury in young plants.[11] Consequently, there is a great deal of interest in precise measurements of NO_x.

The laser-radar technique[12] allows range-resolved pollution monitoring, which has several advantages over the often-used point monitors. Larger areas can be probed easily, and pollutant distributions can be determined through mapping, where several measurements in different directions are performed. Range-resolved measurements of NO_2 have been performed by several authors.[13–16] In the path-integrated measurements of NO reported earlier by our group, stimulated Raman shifting of the frequency-doubled output of the dye laser was utilized, yielding pulse energies of approximately 40 μJ, with a linewidth of 5 pm. Now substantially higher pulse energies and narrower linewidths are utilized, which make range-resolved NO measurements possible.

A mobile laser-radar system[17] was employed in the measurements. It has been used for atmospheric monitoring of SO_2 and NO_2 and recently atomic mercury.[18] A Quantel YG 581C Nd:YAG laser and a TDL50 tunable dye laser with a dual-wavelength option were used. To generate the desired wavelength, the dye laser was operated at 575 nm and the output radiation was frequency doubled with a KD*P crystal and mixed in another KD*P crystal with the fundamental 1.06 μm from the pump laser, which was operated with an intracavity étalon to reduce the linewidth. The pulse energy of the dye laser was about 100 mJ, and after frequency doubling it was 20 mJ. The resulting pulse energy in the transmitted beam at 227 nm was 3–5 mJ, with a linewidth of about 1 pm.

The $\gamma(0, 0)$ band of the NO molecule near 226 nm was used for the monitoring. The part chosen is a bandhead at 226.8 nm, where the baseline absorption is smaller than in the center of the band. This bandhead was also selected because the interference from SO_2 absorption is negligible. Spectroscopic studies of the bandhead were performed using the laser and electronics of the laser-radar system. A quartz calibration cell 29.5 cm long was filled with 0.33 atm of a calibration gas containing 900 parts per 10^6 NO + Ar and with 0.67 atm of Ar. The laser beam was split into a probe beam and a reference beam that were directed through the cell and beside the cell, respectively, and the absorption spectrum was scanned. A recording is shown in Fig. 1. For the differential absorption lidar (DIAL) wavelengths chosen, 226.812 and 226.824 nm, the differential absorption cross section was found to be $(6.6 \pm 0.6) \times 10^{-22}$ m^2. The uncertainty is due mainly to the precision of the calibration gas. The on-

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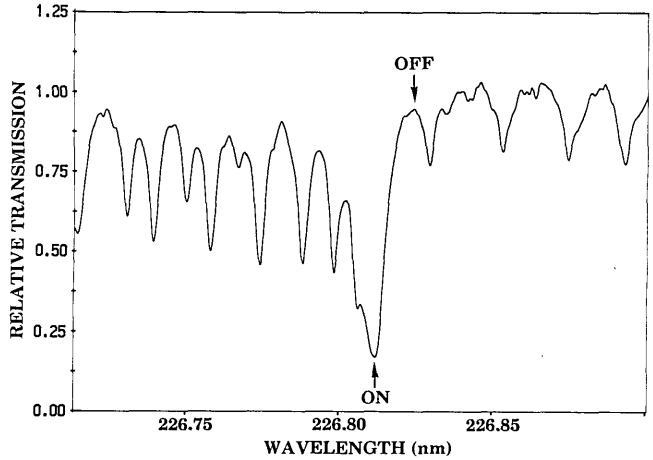

Fig. 1. Absorption spectrum of part of the $\gamma(0, 0)$ bandhead of NO with 1-pm resolution. The optical depth was 88 parts in 10^6 m^{-1}.

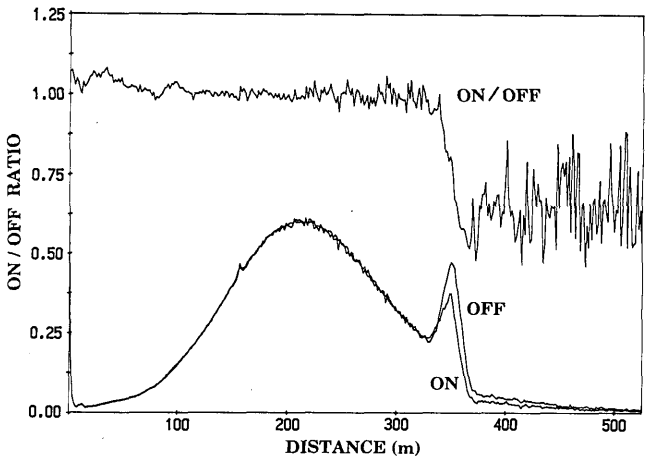

Fig. 2. On- and off-resonance curves (lower) and the DIAL curve (upper) for a NO plume. Note the strong atmospheric attenuation and the heavy nondifferential absorption in the plume.

resonance wavelength value that we use was calibrated earlier against an atomic resonance.[19]

Measurements were performed on a smokestack from a small heating plant. The beam was directed toward the plume, and by measuring in several directions it was possible to map the plume in a vertical cross section. Figure 2 shows the on- and off-return signals and the resulting DIAL curve with the on/off ratio for a single direction. The aerosol dependence, evidenced by the increased backscattering from the plume, is eliminated in the DIAL curve, where the slope in the plume is determined by the difference in absorption cross section for the two wavelengths. The atmospheric extinction at these short wavelengths seriously affects the range. For the pulse energies obtained the maximum range was about 500 m. The measurements were also affected by the fact that the plume, although invisible to the eye, attenuated both the on- and the off-resonance wavelengths heavily, as can be seen in Fig. 2. A mapping of the NO concentration in the plume was achieved by performing several measurements in a vertical scan through the plume, and the result is shown in Fig. 3. Such a scan can also be used to measure the flux of NO passing through the vertical section. The concentrations are then integrated over the area of the plume cross section, and the integrated content is multiplied by the wind velocity normal to the vertical section. Ambient monitoring was also investigated. Figure 4 shows a DIAL curve from the air in Lund, with the beam traversing the air above a road junction; 800 pulse pairs were averaged for this measurement. The NO absorption is readily apparent from the almost exponential attenuation of the curve. The mean concentration on this occasion was evaluated to 130 $\mu g/m^3$. The detection limit, based on the variation between several background measurements and measurements with both wavelengths off resonance, was estimated to 3 $\mu g/m^3$, with an integration interval of 350 m. Multiple scattering may constitute a problem. To investigate this, the signal was detected with an UV polarizer in the tele-

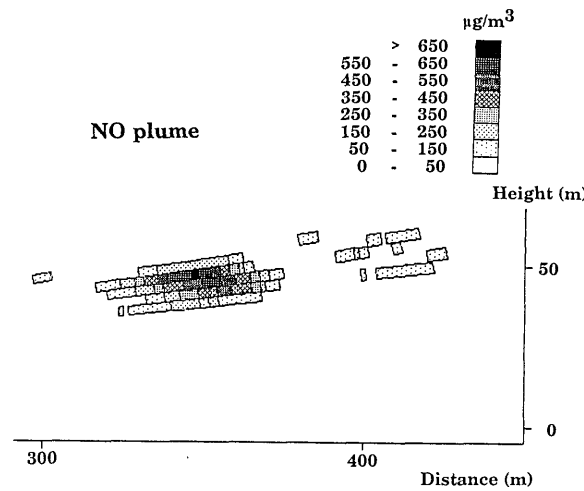

Fig. 3. NO plume mapping.

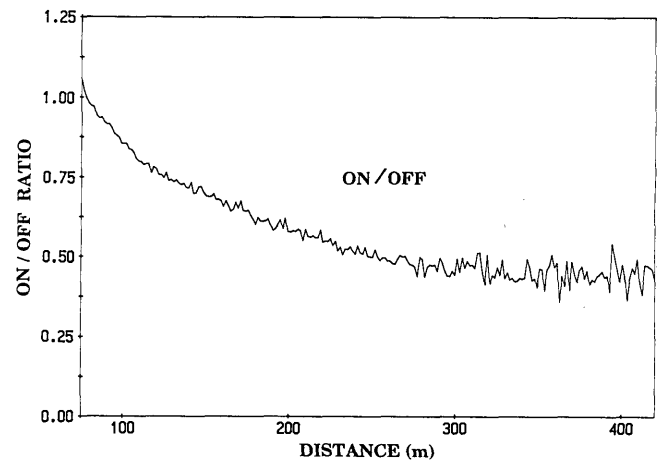

Fig. 4. DIAL curve for ambient air. The mean concentration was evaluated to 130 $\mu g/m^3$ by fitting a straight line to the log of the on/off ratio.

scope before the detection. Since multiple scattering may induce depolarization of a polarized beam,[12] detecting the signal at maximum transmission through the polarizer and then rotating the polarizer 90° to detect at minimum would be a way of detecting depolarization. It was found that the signal was strongly polarized.

The atmospheric extinction coefficient was estimated to be 3.5–4.0 km^{-1} from measurements made off the NO bandhead at 227 nm for normal visibilities. From mercury lidar measurements at 254 nm, a value of 1.2–1.6 km^{-1} was obtained.[18] Existing theory for Rayleigh scattering and empirical formulas for the Mie scattering part[12,20–22] predict a value of about 1.3 km^{-1} at 254 nm. If absorption by ozone is included[21,23] at a concentration of 10 parts per 10^9 (a typical background concentration), this would contribute 0.3 km^{-1} to give a total extinction coefficient of 1.6 km^{-1} at 254 nm, in agreement with the earlier observed values.[18] For 227 nm, however, available Rayleigh and Mie formulas predict an extinction coefficient of 1.6–2.3 km^{-1} for average seeing conditions (visibility 3–10 km), which is much less than the 3.5–4.0 km^{-1} that was estimated from the measurements. The strong increase in attenuation is due instead to the Herzberg dissociation continuum, a very weak continuum ranging from approximately 240 nm down to 190 nm, where the Schumann–Runge bands definitely make the atmosphere opaque. In the literature concerning the absorption cross sections of the Herzberg continuum, many different cross sections are found.[24–29] The discrepancies are due to a pressure-dependent absorption involving two oxygen molecules.[25] Different pressure dependencies have been proposed, and some authors assume a dependence on the nitrogen/oxygen ratio. We have used the attenuation coefficients for oxygen absorption and Rayleigh scattering given by Cheung *et al.*,[29] which are the most recent values. For 227 nm the oxygen extinction is 1.77 km^{-1} for 0.2-atm oxygen. This, together with Rayleigh and Mie extinction values for 3–10-km visibility and allowance for 10 parts in 10^9 of ozone, yields a total atmospheric extinction coefficient of 3.4–4.9 km^{-1}, in reasonable agreement with the experimental value of 3.5–4.0 km^{-1}.

Range-resolved measurements and mapping of NO have been shown to be possible by using the laser-radar technique. An obvious refinement of the measurement technique is to adapt it for simultaneous measurements of NO and NO_2. As pointed out in Ref. 1, this can be done with only one laser source if suitable wavelength pairs are chosen. The UV radiation must then be generated by direct doubling of a blue wavelength. The new nonlinear material beta-barium borate is attractive in this context. Beta-barium borate permits efficient doubling down to 200 nm. Preliminary spectroscopic studies have been performed.[19,30] Studies of the conversion of NO to NO_2 in a plume and investigations of the diurnal cycles of NO_x would also be of considerable interest.

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