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Laser monitoring of atmospheric NO using ultraviolet differential-absorption techniques

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Atmospheric NO was detected in a long-path absorption experiment using a frequency-doubled dye laser, twice Raman shifted in H₂ to 227 nm. Apart from measurements employing a distant retroreflector, the potential of range-resolved lidar measurements was investigated.

The oxides of nitrogen NO and NO₂ are important air pollutants, which together with SO₂ are responsible for the increasing acidification of certain regions. NO is formed in all high-temperature combustion and is emitted from stationary sources as well as vehicles. In the atmosphere, NO is further oxidized to NO₂. Nitrogen oxides are important gases in atmospheric chemistry and sensitively control, e.g., the concentration of tropospheric and stratospheric O₃.¹ In connection with pollution measurements, the total concentration of NO and NO₂ (NO_x) is normally measured by using conventional point monitors. Remote-sensing techniques have many advantages compared with point monitoring. NO₂ absorbs in the blue spectral region and was the first pollutant to be measured remotely by using the differential-absorption lidar (DIAL) method.^{2,3} DIAL monitoring of NO₂ employing Nd:YAG laser-pumped dye lasers is quite effective (see, e.g., Ref. 4).

The first electronic transition in NO occurs at short UV wavelengths (the γ system; A ²Σ⁺ – X ²Π, Δ*v* = 0 at 226–227 nm). Lidar work in this wavelength region is much more difficult because of low laser-pulse energies and limited atmospheric transmission resulting from increasing Mie and Rayleigh scattering. However, in the present paper such measurements are reported. Anti-Stokes shifting of the output of a frequency-doubled dye laser, pumped by a Nd:YAG laser, was used. Previously, NO was remotely monitored by using a cw diode laser,⁵ a cw CO laser,⁶ and a frequency-doubled, pulsed CO₂ laser⁷ on vibrational-rotational transitions around 5 μm.⁵ *In situ* measurements of stratospheric NO concentrations have been performed in balloon-borne experiments by Patel *et al.*⁸ using a spin-flip Raman laser in the IR region. The electronic transitions are advantageous compared with the vibrational transitions because of higher absorption cross sections and less interference from other absorbing molecules.

The setup used in the present experiment is similar to the one described in a recent paper on mercury DIAL experiments.⁹ A Quanta-Ray DCR-1A Nd:YAG laser pumping a PDL-1 dye laser was used. Tunable radiation at 279.5 nm was produced in 20-mJ pulses at 10 Hz by frequency doubling in KD*P of the rhodamine 6G dye-laser output. By second anti-Stokes shifting in a

RS-1 Raman shifter filled with 10-atm H₂ ($\nu_{\text{vibr}} = 4155 \text{ cm}^{-1}$), 0.1-mJ pulses in the 227-nm region were produced. The UV pulses were transmitted into the atmosphere, and backscattered light was collected with a 25-cm-diameter Newtonian telescope equipped with an EMI 9558 QA photomultiplier tube. Optical filtering was performed with an interference filter of 20% peak transmission. Signal detection followed, performed by an averaging transient-digitizer system¹⁰ or a PARC 162 boxcar integrator. Part of the primary UV-laser light was split off to a 30-cm reference cell filled with 1000 parts in 10⁶ (ppm) of NO in 1 atm of nitrogen.

In Fig. 1 an example of a atmospheric-backscattering recording at 226.8 nm is shown. The lidar system was directed toward a topographic target (brick wall) at a distance of 420 m. The recording is an average of 1800 transients. As can be seen, a useful lidar signal can be obtained even with a very low laser-pulse energy at these short wavelengths, although it is sensitive to atmospheric conditions.

A strong signal enhancement can be obtained by using a retroreflector. This is illustrated in Fig. 2, in

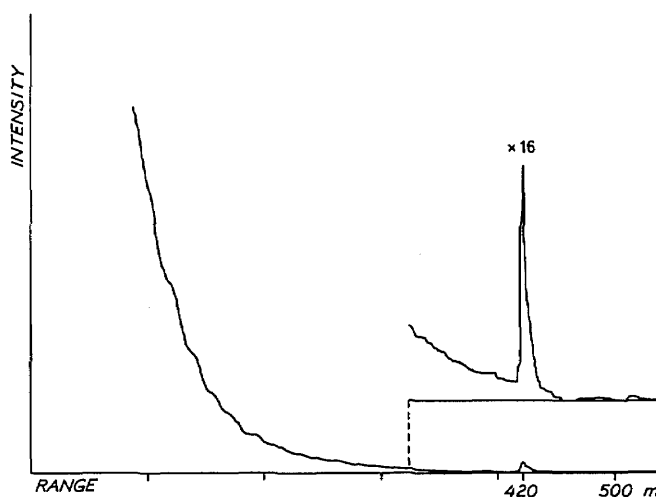


Fig. 1. Lidar recording obtained as the average of 1800 transients at 226.8 nm. Atmospheric backscattering as well as the echo from a topographic target at 420 nm is shown.

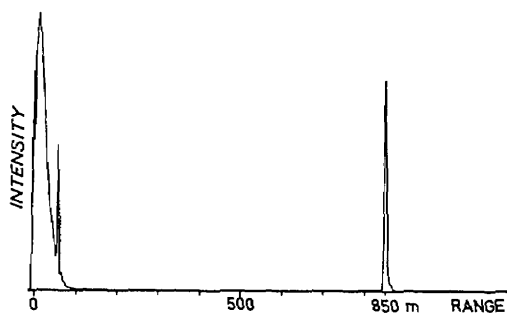


Fig. 2. Lidar recording at 226.8 nm with prompt signal at 0 m (scattering in telescope), atmospheric backscattering with "grow-in" of laser and detection lobes, an echo from an intermediate folding mirror at 70 m, and the echo from a 12.7-cm-aperture corner-cube reflector. The curve was obtained by adding 1000 individual transients.

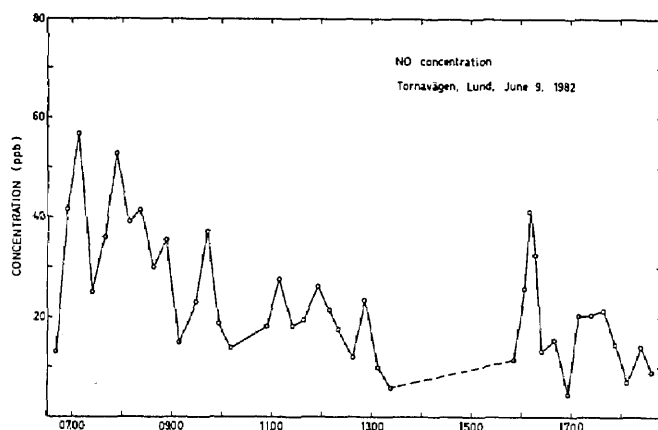


Fig. 3. Recording of the average NO concentration over a pathlength of 2 m \times 850 m about 10 m above a street in Lund. Each dot represents the average of several measurement values.

which a recording of 1000 averaged transients is shown. A 12.7-cm-aperture aluminized retroreflector (Edmund Scientific No. 72273) was used at a distance of 850 m. The echo at 70 m is due to a 50-cm \times 50-cm first-surface aluminum mirror used to fold the optical path. The mirror, used at a 45° incidence angle, directed the laser light toward the retroreflector in a path 10 m above a street in Lund. The average concentration of NO above this street was measured by differential absorption employing wavelengths close to the NO band head at 226.8 nm. An on-resonance wavelength of 226.80 nm was used, whereas 226.82 nm and sometimes 226.78 nm served as off-resonance wavelengths. In measurements on our NO reference cell, the differential-absorption cross section at the used laser linewidth of 4 cm was determined to 5.8×10^{-3} (ppm \cdot m) $^{-1}$ and 4.4×10^{-3} (ppm \cdot m) $^{-1}$ for the first and second line pairs, respectively. In the remote NO measurements, the retroreflector echo was gated in with the boxcar averager, and its height was recorded with a 5-sec time constant on a strip-chart instrument. The average laser power was simultaneously recorded. In the measurements, the signal levels were recorded for about 1 min at alternat-

ing wavelengths. A differential absorption of up to 50% was recorded for the 1700-m path length. In Fig. 3 the average NO concentration during a day is recorded. The circles represent mean values of 4–6 recordings during 10–15 min. A correlation with busy traffic hours can be noted. Because of a rather strong wind, the NO level is strongly influenced not only by the particular street but also by the nearby area, in particular, a highway. A good consistency of values was obtained between measurements employing the two wavelength pairs. Possible interference from SO₂ was excluded by determining that SO₂ has no significant differential absorption in the wavelength region used.

The present investigation shows that UV-lidar techniques can be extended to NO monitoring. By sum generation of doubled dye-laser radiation and residual 1.06 μ m of radiation from the pump laser, at least 30 times higher pulse energies should be attainable. Range-resolved measurements utilizing atmospheric backscattering should then be fully possible. Clearly, fast switching between on- and off-resonance wavelengths is then especially highly desirable. This can be performed in the Swedish mobile lidar laboratory,⁴ and such measurements are planned.

In long-path absorption measurements using a pulsed laser, advantage of intermediate mirror echoes (as illustrated in Fig. 2) can be taken for partially range-resolved measurements. By surrounding the telescope with a few retroreflectors, multiple-pulse bounces between a reasonably close retroreflector and the lidar system should be obtainable for increased path length and consistency checks.

Wavelengths for NO DIAL measurements can also be produced at low efficiency by direct frequency doubling of blue light in KPB. The possibility of performing simultaneous measurements of NO and NO₂ with a single lidar system is of particular interest for studying the atmospheric transformation of NO to NO₂.

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