A wide-range ultraviolet lidar system for tropospheric ozone measurements: Development and application

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A KrF-laser-based ozone lidar system is described which operates in the wide vertical range between 0.1 and 12 km. The wavelengths used for the measurements are generated by efficient stimulated Raman scattering which is optimized by using a KrF laser in an oscillator-amplifier configuration. Two receiving telescopes are used to divide the considerable dynamic range of the backscattered signal which covers more than eight orders of magnitude. The ozone density errors are smaller than $7.5 \times 10^{-6}$ m$^{-3}$ (i.e., 3.1 ppbv near the ground) which is comparable to those of standard ozone monitors. A further improved performance is expected in the future. A first annual series with about 580 individual measurements was carried out in 1991 and is briefly discussed.

I. INTRODUCTION

The gradual increase of the tropospheric ozone concentration has recently given rise to research programs for the verification of the detailed mechanisms governing the balance of this harmful constituent in the lower atmosphere. The existing network of ground-based monitoring stations for complete and long-term measurements of ozone and a variety of its precursors has been extended. Nevertheless, agreement has been reached that, due to the meteorological and chemical conditions near the ground, these stations, in general, cannot give a fully representative picture of the tropospheric ozone distribution. The density maxima are frequently observed in the upper elevations of the planetary boundary layer. Their localization as well as a mapping of the temporal variation of ozone in the boundary layer can only be achieved by vertical-sounding methods. Such measurements may also help to determine the seasonal variation and the long-term concentration increase of ozone in the free troposphere.

Vertical sounding of atmospheric trace gases and meteorological quantities traditionally means ascents of suitable sondes with balloons. Balloon-based ozone measurements, however, suffer from several shortcomings. First, the long preparation times and the considerable costs of the launches restrict the number of flights to a few per week. Frequent measurements of the diurnal variations thus become problematic. Second, the chemical sensors of typical ozone sondes are slow thus preventing the vertical resolution desirable for measurements in the vicinity of altitudes with strong concentration gradients. To compensate for the long response time by a slower rise of the balloon is prohibited by the often enormous lateral drift during an ascent. Finally, a careful calibration of the sensors is necessary.

These problems do not occur in the case of lidar measurements. Almost continuous measurements are possible during a single day with only moderate costs. The instrument does not require repeated calibrations, the ultimate accuracy, reached for a clear atmosphere, being determined by that of the known absorption cross sections and by the detection noise. A high vertical resolution can be achieved with height intervals even below 50 m, at least for moderate distances. Full position control is ensured by the light-beam axis and the light-pulse traveling time.

Ozone monitoring systems utilizing the differential-absorption lidar (DIAL) technique have successfully been used for measurements in the stratosphere for more than a decade (see, e.g., Refs. 1–15). Most of these systems are based on powerful XeCl excimer lasers with emission at 308 nm (as first emphasized on by Uchino et al.16) as well as a reference wavelength of 353 nm generated by stimulated Raman shifting the 308 nm radiation in hydrogen. This technique is established for precise measurements between 10 and 50 km above the ground.4,6–15

For many years, tropospheric ozone measurements with the DIAL method have been carried out more or less on a demonstrational basis or in short campaigns (see, e.g., Refs. 17–25). Several of these systems have even applied frequency-doubled dye lasers, the frequently low pulse energies of which have not always permitted optimum specifications for the lidar operation. Only very recently have there been stronger efforts to develop fully operational DIAL instruments for long-term observations of ozone in the lower atmosphere. In view of the importance of the applicability of such systems in field campaigns there is some major demand for a robust design, easy handling, and also low costs. Fixed-frequency lasers are, therefore, preferred to the more sophisticated tunable ones, which usually require excitation by expensive pump lasers. The first of these new ozone lidar systems have been based on frequency-doubled Nd:YAG lasers26–28 The range of the system in Ref. 26 is restricted to the upper troposphere, but also includes the lower stratosphere. Recently, systems with Raman-shifted excimer lasers have also been under development at several places.

The system described in this article (preliminary results were published in Ref. 29) is derived from the stratospheric lidar systems developed by Rothe et al.4,6,8 It has been designed to provide accurate ozone density distributions from near to ground level up to the tropopause. It, again, uses an excimer laser as the radiation source and stimulated Raman shifting in hydrogen to generate the wavelengths to be used for the differential absorption measurements. Excimer lasers
provide substantially higher pulse energies and repetition rates than frequency-quadrupled Nd:YAG lasers and thus allow averaging times of just a few minutes.

The good experience with stratospheric systems cannot simply be transferred to the tropospheric case. There are a few important differences one has to pay attention to. First, one faces a dynamic range of the backscattered radiation of more than eight orders of magnitude which is caused by the \( \frac{1}{r^2} \) law, the exponential decrease of the backscatter coefficients as a function of the height \( r \) and the frequently enhanced extinction by aerosol layers or clouds in the lower atmosphere. This huge dynamic range cannot be handled by a single photodetector. Second, a sensitive determination of the much lower ozone densities in the troposphere requires stronger absorption, i.e., wavelengths shorter than 308 nm. The absorption cross section of ozone in the region of the unstructured\(^{30} \) Hartley band system increases continuously towards shorter wavelengths and reaches its maximum value \((1.1345 \times 10^{-21} \text{ m}^2)\) at 254.0 nm.\(^{31-34} \) The KrF excimer laser, which is used for this system, emits at 248.5 nm, i.e., next to this wavelength. The absorption by ozone at 248.5 nm is already far too strong to allow measurements up to the tropopause. The optimum wavelength range for tropospheric ozone measurements turns out to be about 275–295 nm, with an additional “off” wavelength beyond 310 nm. For the optimization of the emitted light the stimulated Raman shifting of the 248.5 nm radiation into the first and also the second Stokes order for hydrogen and deuterium had to be studied in detail. There was not sufficient experience available at the beginning of this project, in particular for the chosen KrF laser type.

In addition, for an operational lidar system one had to gather experience concerning some potential problems with the short wavelengths below 300 nm, i.e., aging by, e.g., color-center formation, the excitation of fluorescence of optical components in the receiver or signal distortions caused by excess electron emission from the detector photocathode. Furthermore, the light scattering and extinction by aerosols and clouds has to be taken into consideration, which is only a minor problem in the stratosphere except during periods with enhanced volcanic activity. The main difficulty arises from the necessity to select rather distant wavelengths for the DIAL measurements of \( \text{O}_3 \) caused by the absence of spectral structure in the Hartley band system and from the not very precisely known wavelength dependence of the light scattering by aerosols. Apart from the aerosol interference one has to consider also that of other trace gases absorbing in the same spectral region as \( \text{O}_3 \), the species to be taken most seriously being \( \text{SO}_2 \).

Finally, for ozone measurements in the lower troposphere, a reliable daytime operation has to be guaranteed because of the pronounced diurnal cycles of the \( \text{O}_3 \) density. At the reference wavelength of 313.2 nm (second Stokes order for Raman shifting in hydrogen) the scattered sunlight is not sufficiently absorbed by ozone and, thus, a rather high background light flux hits the detector. The daylight background is lowered relative to the backscatter signal by both a high transmitted pulse energy and narrow-band filtering at the receiver. Optimum filtering by using a combination of Fabry–Pérot etalons as demonstrated in Ref. 8 is not suitable for the near-range of a tropospheric lidar where a rather wide acceptance angle has to be admitted.

This work was carried out as a part of the TESLAS (Tropospheric Environmental Studies by Laser Sounding) subproject of the European EUROTRAC project. This subproject has focused on the development and testing of ozone lidar systems for tropospheric measurements which should, after their completion, be made available for routine measurements coordinated in other EUROTRAC subprojects.

The article is organized as follows: In Sec. II, we describe the generation and transmission of the DIAL wavelengths, the receiver and the data acquisition, as well as our concept for an improved receiver bandwidth reduction. Section III contains details on the data evaluation. The most important error contributions are discussed. In Sec. IV, a short description of a first annual series of ozone measurements is given, which was carried out in 1991. This series demonstrates the suitability of the new lidar for routine measurements and has led to first conclusions concerning the principal features of the tropospheric ozone distribution in our area.

II. DESCRIPTION OF THE TECHNICAL DETAILS

An assembly drawing of the lidar system is shown in Fig. 1. The system is distributed over two laboratories. The first room contains the laser, two Raman shifters, and a large beam-expanding telescope. In the second room, the receiving system and the final laser beam steering mirror are installed. Due to this separation, the laser system is not exposed to the frequently cold air from the open windows in the roof.

A. Generation of the DIAL wavelengths

For the generation of the UV wavelengths applied in the described lidar system a narrow-band KrF excimer laser was purchased (Lambda Physik, model EMG 150T MSC). This laser consists of an oscillator, providing a 0.2 cm\(^{-1}\) bandwidth and a good beam quality, and an amplifier which finally yields pulse energies of more than 250 mJ with a repetition rate of up to 80 Hz. Due to this oscillator-amplifier combination, a low beam divergence of less than 0.2 mrad is achieved. The KrF laser is tunable in a wavelength range of about 0.8 nm without broadband contributions. This may be helpful in order to reduce possible errors due to the cross sensitivity for \( \text{SO}_2 \) which is not important under the clean-air conditions.
conditions at Garmisch-Partenkirchen, but has to be taken into consideration for measurements in urban or industrialized areas.

The narrow bandwidth and the low beam divergence are key properties for the efficient spectral filtering in the receiver which is necessary to achieve high quality and high vertical resolution for daytime measurements in the upper troposphere. The narrow-band filtering could not be completed under the first research contract. Nevertheless, an improved concept has been worked out which is described in Sec. II D. As will be shown below, the chosen KrF laser is also invaluable for the efficient generation of the additional DIAL wavelengths by stimulated Raman shifting.

The 248.5 nm radiation is strongly absorbed by ozone. Therefore, the typical useful vertical range for this wavelength frequently does not exceed 4 km. The preferred wavelengths of this system are, therefore, 277.1, 313.2, and 291.9 nm which are the first and second Stokes orders of the KrF laser light for stimulated Raman shifting in hydrogen and the second Stokes order for shifting in deuterium, respectively. 313.2 nm is located outside the Hartley band system and serves as the reference wavelength. With the wavelength pair 277.2 nm/313.2 nm a vertical range ending between 8 and 12 km can be achieved, depending on the actual ozone density in the troposphere and the amount of daylight-induced signal noise around 313.2 nm. With the pair 291.9 nm/313.2 nm, the upper limit is expected to exceed 14 km even under ozone-rich conditions. Because the wavelength separation capabilities are presently still limited, the use of 291.9 nm had to be postponed.

Due to diffraction by a coated spot on the output mirror, hot-spot formation occurs leading to the destruction of the coatings of any nearby dielectric mirror. Thus, the first 45° mirror is placed at a distance of 1.3 m from the laser head where the high-intensity portion of the beam has already expanded to some uncritical diameter.

The 248.5 nm beam (cross section 24×6.5 mm²) is focused into the 1.9-m-long stainless-steel Raman cell by a "best-form" lens with a focal length f = 1.00 m at 248 nm. The quality of this lens was verified by the observation of an "best-form" lens with a focal length f = 1.00 m at 248 nm. The quality of this lens was verified by the observation of an air breakdown in the focus. Shorter f values are, in general, not recommended because of the onset of efficiency-lowering processes as, e.g., electric breakdown, multiphoton excitation, or intensity-induced refractive-index changes.

The focusing lens and the cell windows are made of quartz and broadband dielectrically coated for at least 248–313 nm. Some aging of the optical components is observed, starting after only a few months of operation, which is, in part, ascribed to color-center formation. There is also some indication that the coating of the windows inside the Raman cell suffers from either the presence of the intense UV radiation or the hydrogen gas or both. In the future, all UV-transmitting optics will be made of CaF₂.

The output of the Raman cell is collimated by an f = 5.0 m concave spherical mirror with more than 99% reflectance for the wavelengths under consideration (Laseroptik G.m.b.H.). By using the mirror, chromatic aberrations are avoided. Because of the beam expansion, previously observed differences in the beam divergences of the different

![FIG. 2. Conversion efficiency for stimulated Raman shifting in hydrogen as a function of the H₂ pressure at a pulse energy of about 196 mJ; the curves for the depleted pump radiation (P), the first anti-Stokes order (aS₁) and four Stokes orders S₁–S₄ are shown.](https://example.com/figure2.png)

Raman orders have disappeared (see Secs. II A 1 and II D).

In order to determine the optimum operating conditions for the Raman shifting, detailed measurements of the conversion efficiencies were made. A few preliminary results are also presented as a part of a TESLAS joint paper on Raman shifting of frequency-quadrupled Nd:YAG and KrF laser radiation. The full account of our work is described here. The results are sometimes surprising and depend strongly on the chosen conditions. This justifies the systematic investigations carried out by the authors and also by various other groups.

1. Results for the stimulated Raman shifting in hydrogen

The results presented in Secs. II A 1 and II A 2 were obtained with linearly polarized 248-nm light. Linear polarization was achieved by placing a Glan polarizer (Hilite) between oscillator and amplifier of the KrF laser and by replacing the birefringent MgF₂ windows of the discharge cells of the laser by CaF₂ ones (see also Sec. II A 6). The repetition rate was set to 2.5 Hz.

Figure 2 shows the energy conversion efficiencies for the first four Stokes orders and the first anti-Stokes order as a function of the hydrogen pressure for average pulse energies of about 198 mJ inside the Raman cell. The conversion efficiencies are related to the 248 nm radiation transmitted through the empty Raman cell, the prism, and the two focusing lenses which were used for the spectral separation and shaping of the spots caused by the different Raman orders on a paper screen or on the energy meter. The slight decrease of the pump pulse energy during the measurements is taken into account in the evaluation. The fifth and sixth Stokes order (S₅ and S₆) look rather bright for pressures around 3 bar because of their location in the visible (513.9 and 653.4 nm, respectively), but their pulse energies did not even reach the detection threshold (1.5 mJ) of our energy monitor (GENTEC, PRJ-M, with an ED200 pyroelectric detector). The data were acquired by taking ten-shot averages which yielded sufficiently reproducible results.
The highest pulse energies for the S1 radiation are observed at rather low pressures whereas for S2 maximum values are reached at 2.2 bar as well as around 30 bar. For simultaneous optimum performance at both wavelengths, 277 and 313 nm, the operating pressure should be set near 2.2 bar.

The intensity dependence of the conversion efficiency is not very pronounced. A measurement with a pulse energy of 128 mJ yields almost the same values, the only differences being a slightly lower S2 efficiency at 2.2 bar (27%) and an almost constant anti-Stokes output remaining above the GENTEC detection threshold now for pressures up to 18 bar.

From a gain calculation using the expressions and data in Ref. 36, the S1 output is expected to increase continuously as a function of the pressure and to level off above 10 bar (see Ref. 37). The observation of a maximum at as low as 1.5 bar is explained by the depletion of the S1 radiation by the generation of S2 and higher Raman orders above this pressure. The lower S2 efficiency for 128 mJ should yield some enhanced conversion into S1 below 2 bar. Since there is no appreciable intensity dependence of the S1 efficiency one has to conclude a slightly lower gain at 128 mJ also for the S1 formation in this pressure range.

Two processes may be responsible for the generation of higher Stokes orders. The first is a cascade-type process in which the frequency of a given Stokes order is again first-Stokes shifted. The second is four-wave mixing which may also involve anti-Stokes photons, in addition to the pump and Stokes photons. In the case of second-Stokes generation, the combinations

$$\omega_{S2} = 2\omega_{S1} - \omega_p \quad \text{and} \quad \omega_{S2} = \omega_p + \omega_{S1} - \omega_{aS1}$$

may contribute. According to Bobbs and Warner, the cascade-type process is frequently efficiently suppressed by some divergence of the first-Stokes radiation.\(^{38}\)

Indeed, the four-wave mixing process is found to be rather efficient, at least for low pressures. Figure 3 shows the cross section of the uncollimated output beam of the Raman cell at a distance of 5 m from the focus, as observed for moderate pulse energies. With the onset of second-Stokes formation, a four-wave mixing double cone appears. The occurrence of a double cone instead of a single one is caused by a dark spot at the center of the 248-nm beam of the EMG 150 T laser which separates the beam into two high-intensity regions. The structure is rather diffuse, the cones being entirely filled with light. Above approximately 3.5 bar, which is a pressure within the rising edge of the S3 curve, an even larger double cone shows up. Finally, with further increasing pressure, the entire exit window of the Raman shifter becomes illuminated. The double cone is not present near the maximum pump pulse energy where just a highly diffuse single-cone structure is observed.

Apparently, due to the higher-order intensity dependence of the higher-order Stokes beam formation, there is a trend to favor the generation of the highest possible Stokes order. At further elevated pressures, the coherence length and the phase-matching conditions for the four-wave mixing processes become worse and the conversion efficiency for the lower orders starts to increase again. Above 18 bar, even the S1 intensity starts to recover. It should be noted that the anti-Stokes radiation remains rather weak in a wide pressure range which reflects the lower gain for its formation, but possibly also indicates some conversion into Stokes radiation by four-wave mixing (see Fig. 2). The aS1 efficiency drops below the GENTEC detection threshold as the S2 efficiency grows. The anti-Stokes results may, however, be somewhat influenced by not considering the lower system transmittance at these shorter wavelengths.

In the presence of four-wave mixing a higher beam divergence is observed. This effect was confirmed and measured quantitatively by Diebel et al. who obtained a divergence angle of about 1.8 mrad (full width) for the second Stokes order.\(^{39}\) This divergence can be substantially reduced at elevated pressures which we tentatively ascribe to the reduction of the four-wave mixing and a faster collision-induced \(\nu = 1 \rightarrow \nu = 0\) relaxation. In order to make possible an essentially divergence-free operation of our system at low pressures, the 5:1 beam expander was installed (see Fig. 1 and Sec. II D).

Figure 4 shows the pressure dependence of the conversion efficiency for about 187 mJ of incident broadband 248-nm radiation. The broadband light is generated by operating the KrF amplifier as a laser which is achieved by blocking the oscillator beam. It could be verified that the beam divergence of the broadband beam did not change with respect to the normal operating mode. This is ascribed to the optical configuration of the beams expander in the amplifier section acting like an unstable resonator. Consequently, the focusing conditions are expected to be the same as for the narrow-band beam.

A comparison of Figs. 2 and 4 shows that the S1 and S2 energy conversion efficiencies achieved for low pressures with the narrow-band version of the laser are not reached when the oscillator beam is blocked by a piece of paper. For
FIG. 4. Conversion efficiency for stimulated Raman shifting in hydrogen as a function of the H\textsubscript{2} pressure at a pulse energy of about 187 mJ for broadband operation of the KrF laser (oscillator blocked); the curves for the depleted pump radiation (P), the first anti-Stokes order (aS1) and four Stokes orders (S1–S4) are shown.

FIG. 5. Conversion efficiency for stimulated Raman shifting in deuterium as a function of the D\textsubscript{2} pressure at a pulse energy of about 94 mJ; the curves for the depleted pump radiation (P), the first anti-Stokes order (aS1) and four Stokes orders (S1–S4) are shown.

FIG. 6. Conversion efficiency for stimulated Raman shifting in deuterium as a function of the D\textsubscript{2} pressure at a pulse energy of about 201 mJ; the curves for the depleted pump radiation (P), the first anti-Stokes order (aS1) and four Stokes orders (S1–S4) are shown.

The dependence of the S1 gain on the laser bandwidth was discussed by Trutna et al. Their theoretical considerations and experimental results for a variable-bandwidth Nd:YAG laser suggest the absence of bandwidth effects on the Raman gain in the absence of a dispersion of the Raman medium. The lack of a bandwidth dependence of the gain was further hardened by the theoretical treatment of Raymer and Mostowski. Our own experience with narrow and broadband versions of a Nd:YAG laser confirms this.

If the laser bandwidth does not influence the gain one must consider the polarization of the laser as a possible source of the observed differences. The broadband KrF laser may be assumed to be more or less naturally polarized, in contrast to the linear polarization in the oscillator-amplifier case. Flusberg and Holmes state that the Raman gain is lower for unpolarized than for polarized light. The importance of the state of polarization for the stimulated Raman shifting is further discussed in Sec. II A 6.

It is interesting to note that the pulse-to-pulse fluctuations of the Stokes energies are clearly smaller in the case of the broadband operation. In general, the fluctuations are higher for low pressures than for higher ones where they no longer exceed those of the pump radiation. For narrow-band operation, the ten-shot standard deviations are mostly restricted to values below 4 mJ, 1 mJ under optimum pressure conditions. The influence of the repetition rate on the S1 output was tested up to about 30 Hz which can still be resolved by the energy meter. A 9% decrease of the S1 pulse energy was found which is mostly due to a decline in the pump pulse energy.

2. Results for the stimulated Raman shifting in deuterium

Despite a gain coefficient which is lower by almost one order of magnitude compared with that for Raman shifting in hydrogen similar conversion efficiencies were observed in the case with deuterium as the Raman medium. Figs. 5–7 show the pressure dependence of the efficiency for average pulse energies inside the cell of approximately 94 and 201 mJ narrow band, and 179 mJ broadband, respectively. The threshold pressures are slightly higher than for hydrogen (S1: almost 0.7 bar instead of 0.5 bar) and the rising slopes for the various Stokes orders are less steep, but the maximum values, which are reached at higher pressures, are not much lower. The efficiencies at low pressures exhibit a more pronounced intensity dependence than those for hydrogen.

The shot-to-shot fluctuations of the Stokes radiation are slightly higher than those for hydrogen. Since this effect might be caused by thermal overload in the focus the repetition rate of the laser was increased to 30 Hz. Indeed, a drop of the S1 intensity by almost 30% was observed. If thermal overload in the focus was the reason for this intensity drop, gas circulation would be a good solution for the operation at high repetition rates. Our recent results for Stokes shifting Nd:YAG laser radiation demonstrate that gas contamination...
by, e.g., hitting the walls of the Raman cell with the laser beam during the adjustment phase may be a major source of troubles.

3. Mixtures of hydrogen and buffer gas

Luches et al. demonstrated that the use of buffer gases such as Ar and He may efficiently reduce the contributions of the higher Stokes orders and enhance the conversion into S1, although an upper limit of about 43% for S1 could never be surpassed. This behavior is explained by reduced four-wave mixing at elevated buffer gas pressures due to an increasing phase mismatch.

In our case, for 10 bar H2 in He, the S1 conversion efficiency climbed from 11% for pure H2 to not more than 20% at a total pressure of 30 bar. For S2, an increase was also seen. The efficiency rose from 25% to 34% in the same pressure range. We ascribe this increase to the choice of the H2 partial pressure near the maximum for the S3 formation in pure hydrogen. Obviously, some of the S3 energy must be converted into the enhancement of S2 radiation as the helium pressure is raised.

A higher S1 efficiency increase for the admixture of buffer gas is achieved if a frequency-quadrupled Nd:YAG laser (266 nm) is used instead of a KrF laser. By adding about 25 bar of buffer gas to 10 bar of hydrogen, peak S1 efficiencies of more than 70% have been demonstrated. We tentatively ascribe these higher values to the shorter pulse length and a possibly better beam quality of the 266 nm light.

It is interesting to note that, for low H2 partial pressures, the S1 and S2 efficiencies may even decrease with increasing buffer gas pressure. This was recently found by Krause for 1.5 bar H2 in Ar using a similar KrF-laser model. At this low hydrogen pressure, however, there is no much conversion of S1 or S2 light into the higher orders. Thus, diminishing the efficiency for these orders by enhanced buffer-gas pressure cannot yield better results for S1 and S2. Nevertheless, these discrepancies demonstrate that the H2 partial pressure has to be properly chosen if optimum results for the mixing with buffer gas are desired.

For the lidar operation, we preferred to use pure hydrogen near 2.2 bar where the best simultaneous S1 and S2 conversion efficiencies can be achieved. With fresh optics, up to 70 mJ were observed in both orders. Nevertheless, one must compensate for the substantially stronger absorption of the 277 nm radiation by ozone by favoring the S1 generation. The S1/S2 ratio is chosen according to the seasonal ozone variation which causes enhanced S1 absorption in summer. The typical operating pressures vary between 1.5 and 1.9 bar.

The surprisingly good Raman-shifting performance at low hydrogen pressures is advantageous for safety reasons too, in particular when considering a future use of fragile CaF2 cell windows, where a thickness of about 15 mm has to be chosen at pressures around 40 bar. In addition, stimulated backscattering is negligible in the range below 20 bar.

4. Results for two consecutive Raman cells

Experiments with two consecutive Raman cells were carried out in order to lower the threshold for the S1 and S2 generation in the second cell by the injection of the Stokes radiation emerging from the first one. However, no significant improvement over the single-cell maximum conversion efficiencies could be achieved. This might, in part, be ascribed to the losses due to the additional windows and lenses used in the extended setup. No need is seen to use more than a single Raman cell for the shifting in any of the gases.

Grant et al. emphasize on using two consecutive Raman cells with different gas fills, the first containing deuterium, the second hydrogen. The aim of this choice is the generation of 302 nm in addition to 277 and 292 nm which would result in some higher flexibility for the operating-range selection. The achieved efficiencies for 220 mJ of incident pump radiation are not very high, possibly due to the use of an unpolarized KrF laser.

5. Mixtures of hydrogen and deuterium

Some preliminary measurements with mixtures of hydrogen and deuterium were made with the hope of achieving a high simultaneous energy conversion into 277, 292, and 313 nm. A surprisingly high number of spectral components was observed and these investigations were abandoned. As pointed out below, a quantitative spectral separation of so many emission lines at the receiver is a difficult task. A good spectral separation is, nevertheless, crucial for the correct evaluation of ozone densities.

6. Spectral purity of the generated light

The spectral purity of the Raman-shifted 248-nm radiation is essential for a high-quality evaluation of ozone densities. The maximum population of the few populated rotational levels of H2 is found in J = 1, which is particularly pronounced due to the enhanced degeneracy caused by the ortho nuclear angular momentum. This gives rise to a concentration of the Raman conversion efficiency in the Q1 transition. However, significant intensities for rotational satellites
of the predominant $Q_1$ transition were recently reported by Schaberl and Bösenberg and by Krause and Weitkamp.\textsuperscript{46,48,49} The rotational spacing for $H_2$ and $D_2$ is large enough that the occurrence of additional Raman lines can give rise to a modified absorption cross section of ozone at a given Raman order and, thus, to erroneous densities. Since the spectral composition sensitively depends on the intensity, the pressure and the polarization the stimulated rotational Raman effect is highly undesirable.

The laser models used in Refs. 46, 48, and 49 are essentially the same as that applied in our laboratory. Nevertheless, we were not able to observe appreciable amounts of rotational Raman effect under the typical operating conditions of our measurements in 1991. As an example, Fig. 8 shows the vicinity of the second Stokes order for 1.8 bar $H_2$ as obtained by dispersing the output of the Raman cell with a 0.25-m monochromator (Jarell Ash). Any non-$Q_1$ contribution has to be smaller than the noise level of 0.2%.

This discrepancy between the performances of the three systems has been ascribed to a different polarization of the three lasers. We learned from the manufacturer that the discharge-cell windows of the KrF laser are made of birefringent MgF\textsubscript{2}. The thickness of these windows determines the degree of ellipticity. In our case, we found at least 80% linear polarization.

It is well known that circularly polarized light leads to a 50% enhancement of two-photon line strengths.\textsuperscript{50,51} This also holds for the $O$ and $S$ branches of a $^1\Sigma \rightarrow ^1\Sigma$ transition which is the case for Raman shifting. In contrast to this, the $O$-branch line strengths decrease strongly as a function of $J$ for circularly polarized light. This explains the occurrence of $S_1$ rotational transitions (Stokes and anti-Stokes) in addition to the $Q_1$ transitions in the case of some of the oscillator-amplifier KrF lasers. The influence of the degree of polarization on stimulated rotational Raman shifting was discussed previously.\textsuperscript{52-55}

After repolishing the laser windows the polarization became, by chance, almost circular. A large manifold of rotational Raman lines were observed at pressures around 1 bar, up to 7 satellites (3 anti-Stokes, 4 Stokes) for the pump wavelength and less for the other wavelengths. These lines were not all intense but clearly discernible from their fluorescence on a sheet of paper (which has a sub-$\mu$A sensitivity). The threshold hydrogen pressure was found to be as low as 0.4 bar which reflects the 50% increase in the line strengths mentioned above. The rotational satellites disappeared above a pressure of about 4 bar.

With the laser oscillator blocked no rotational Raman shifting was observed at all. This, once more, indicates natural polarization for the broadband operation.

Recently, as already mentioned in Sec. II A 1, the MgF\textsubscript{2} windows of the KrF laser were replaced by CaF\textsubscript{2} ones and a Glan polarizer placed at the output of the laser oscillator. The final results on Raman shifting presented in Secs. II A 1 and II A 2 were obtained with this modification and replace those from earlier measurements. With linearly polarized 248-nm light, the rotational Raman effect in hydrogen could only be observed (on a piece of fluorescent paper) up to 1.7 bar. The efficiencies for the rotational satellites are very low and could not be determined.

B. Receiving optics

The considerable dynamic range of the backscattered light is greatly reduced by the use of two spatially separated Newtonian receiving telescopes. The smaller of these telescopes (0.13 m diameter, Vehrenberg K.G.), located at a distance of 0.2 m from the emitted UV beam, is used for the less sensitive measurements in the near-range (up to 2 km for, e.g., 277 nm). The large telescope (with a 0.50-m diameter parabolic primary mirror from Lichtenknecker Optics) is displaced from the UV beam by 1.6 m and does not receive light backscattered from distances $r<0.6$ km. The two telescopes collect all light for $r>0.08$ km and $r>1.4$ km, respectively. Due to the use of these two separate telescopes, no major overload-induced detector nonlinearities\textsuperscript{56,57} could be observed within the operating range of this lidar system.

In the present configuration, two wavelength pairs (248 nm/277 nm and 277 nm/313 nm) can be selected by simply shifting the required beam splitters on a translation stage and by exchanging the interference filters. The bandwidth of the interference filters (Schott) is specified as 7–12 nm, limited by the need for at least some transmittance at the specified UV wavelengths (about 0.15 for these filters). Because of this large bandwidth a full separation of 292 nm from 277 and 313 nm is not possible. The use of 292 nm/313 nm as a third wavelength pair has, thus, to be postponed. Due to the limited vertical range for 248 nm the pair 248 nm/277 nm was only used in the beginning and will be omitted in the future. All results presented in this paper are for 277 nm/313 nm.

In the future, the detection bandwidth will be improved by almost two orders of magnitude by replacing these optics by a 1.1-m grating spectrograph. Its design is described in Sec. II D. Due to the improved bandwidth, an optimum performance of the lidar system during the day and the additional use of 292 nm will be possible.

The layout of the receiver optics is rather compact and is the result of redesigning a substantially larger earlier version. Prior to the application of the beam-expanding telescope.
shown in Fig. 1, beam divergences up to 2.5 mrad (full angle) had been observed for the different Stokes orders. Since, in the receiver, the divergence is amplified by the focal-length ratio of the telescope (2000/50 for the early version of the large telescope) with subsequent loss of light, an enlarged field of view had to be achieved.

However, with the more compact optics, surprisingly, an initially unobserved signal contribution appeared, which was found to be present only in the shorter-wavelength channel of both wavelength pairs. From this and other observations we tentatively suggest that a weak fluorescence of the beam splitter in front of the two detectors is the origin of this effect. With 277 nm in the 277 nm/313 nm pair, this fluorescence increases the lidar signal at distances beyond \( r = 6 \) km. A mathematical correction can be applied to remove this contribution and all examples shown in this paper are treated in this way. The shape of this correction was determined in a separate experiment by firing the UV beam across the telescopes thus avoiding lidar backscattering from larger distances. Only its amplitude must be adjusted to improve the ozone results in the upper troposphere. Despite the reproducible success of this method an experimental solution is desirable which would increase the reliability in the range around \( r = 10 \) km. This solution is expected to be provided by the large grating spectrograph mentioned above, in which no optical components will be placed next to the detector.

### C. Detection electronics

Two types of photomultiplier tubes (PMTs) were used, EMI 9813 B for the two detection channels of the small telescope and EMI 9893 B for those of the large one. The former type has been used at IFU for many years, the latter was purchased for the sensitive single-photon counting because of its reduced dark current. The signals from the 9893 B multiplier contained less contributions from electromagnetic interference, although the cables to the amplifier are significantly longer. The 9813 B PMTs are used with the small telescope and may pick up more noise from the laser which peaks at short times.

To obtain the present performance, the signal cables had to be doubly shielded. In addition, the trigger pulse which defines \( r = 0 \) is derived in the usual way from a photodiode exposed to a 248-nm reflection. Thus, a ground loop including the noisy laser power supply is avoided.

In order to prevent excessive anode currents next to the peak signals the gain of the four PMTs is switched (range gating). All four PMT sockets were modified and equipped with identical range gating circuits. The diagram for such a circuit is shown in Fig. 9. D1–D10 denote the first ten dynodes of the PMT (D11–D14 are missing in Fig. 9) which are normally interconnected by 150 kΩ resistors for the voltage divider. This voltage-divider chain is modified between D9 and D7. Dynode D7 is used for the range gating. Whereas the voltages for the other dynodes are maintained, that of D7 can be adjusted by a potentiometer \( R_x \) during a time interval complementary to that of the applied adjustable-length trigger pulse. With the trigger pulse off, the gain is lowered, which is the case during most of the time. With the trigger pulse on, the PMT reaches its normal, high gain, which is set in the high-altitude region of the lidar signal. The status selection is made by the transistors T1 and T2 which are controlled by complementary values of the trigger signal.

The TTL trigger pulse is transferred to the circuit by an optocoupler to shift its reference voltage from zero to the applied high-voltage level. This, though being an elegant solution, restricts the choice of the gating electrode to the lower-voltage section of the PMT. In principle, the gating of dynodes near the photocathode would be the optimum choice, but this is presently impossible due to the specified damage threshold of the optocoupler.

The trigger pulse is switched off at fixed 105 μs after the laser pulse. At 313 nm, this turns out to be too early since, after 105 μs, the lidar signal has not yet settled to the constant background level which has to be determined very accurately at the end of the trace. A somewhat longer trigger pulse should still be acceptable considering the very slow voltage decrease at D7.

The excellent performance of the range-gating circuits is demonstrated in Fig. 10. For this measurement, both detection channels of the large telescope were equipped with 313-nm interference filters and exposed to daylight. The horizontal axis (time after the laser pulse) is marked in altitude units. Just above \( r = 3.0 \) km the range-gate trigger pulse is applied (marked by T). The rectangular shape of the 313-nm daylight-induced signal is almost perfect. After about 0.10 km (corresponding to 0.67 μs) a highly constant voltage level is reached which differs from the final level by not more than 0.2%. The PMT gain returns to the low value above \( r = 15.7 \) km. The performance for the 9813 B PMT was just slightly less impressive, with the overshoot remaining below 1.5%.

At several meetings, we presented an example of a vertical ozone density distribution which indicated a deviation of the channel 4 PMT response from the behavior shown in Fig. 10. In a recent detailed investigation, a slower rise shortly before the maximum signal level is reached, could, indeed, be verified for a certain, confined setting of \( R_x \) in channel 4. The very few backscatter profiles featuring a characteristic "ozone hole" above the switching point are now
corrected using the measured modified temporal shape of the PMT4 gain.

The analog signals are collected and digitized by four 8-bit, 30-MHz transient digitizers with amplifiers and 24 bit signal averagers (all DSP). An amplifier gain of 10 is selected. For the 277-nm channel a 50-MHz photon-counting multichannel scaler (CMTE, 1 μs/channel) is connected to the PMT parallel to the analog output. After unsuccessful attempts to match the impedance of these two outputs with a resistor bridge, the input resistance of the photon counting channel was substantially increased which, finally, reduced the noise level in this channel to an acceptable level, at which the following discriminator could be operated over a wider range of PMT voltages. The input resistor was placed directly next to the PMT output in order to avoid multiple pulse reflections.

The digital resolution of the transient digitizers is rather limited in view of the enormous dynamic range of the lidar signal even after the introduction of two receiving telescopes. The PMT range gates are, thus, indispensable to overcome this problem. The gain is adjusted to yield an optimum coverage of the digitizer bits in both subranges. This may be very critical in the presence of high ozone densities which lead to a low signal level already before the PMT gain is switched. Averaging a single-bit step may then result in a sinusoidal modulation of the ozone density distribution in this region. In the future, the voltage offset of the input amplifier will be varied during the measurement in order to avoid any such digitization-induced effect over a wide range of gain settings.

The data of the four transient digitizers and the photon counter are transferred to a personal computer (PC) and stored on optical and mini disks. The entire data acquisition is controlled by a PC program.

Figure 11 shows logarithmic $r^2$-corrected backscatter profiles for the large telescope, accumulated for 15 000 laser shots. All profiles are compensated for Rayleigh backscattering and extinction. This is particularly useful at 313 nm where an almost horizontal line is seen in ranges of good visibility. Thus, one can easily verify the quality of the profiles, especially in the vicinity of the range-gate switching point ($r = 2.8$ km). Major deviations from the horizontal behavior are seen in the planetary boundary layer ($r < 2.3$ km) and in the cirrus-cloud region between 9 and 12 km where enhanced backscattering by aerosols is observed. The very small residual slope in the aerosol-free regions is consistent with the actual ozone density. The enhanced noise above $r = 5$ km is caused by the daylight background, the level of which is about the same as the peak value of the backscattered laser light. At night, the onset of noise occurs above 10 km.

The peak signal at 277 nm is roughly the same as that at 313 nm. This is a favorable situation because both detectors are exposed to about the same amount of light in (at short times) almost congruent pulses. The remarkable differences seen in Fig. 11 are caused by the multiplication by $r^2$ and are much smaller in the original backscatter profiles. The 313 nm trace nicely demonstrates the absence of signal-induced detection nonlinearities \cite{56,57} even to beyond 12 km. At 277 nm, the profiles for analog and photon counting measurements are displayed after the subtraction of the presumed beam-splitter fluorescence (see Sec. II B) in order to make visible the true noise of these traces. The steeper slope of these two 277 nm profiles is caused by the stronger light absorption by ozone. The much lower noise of the photon-counting trace is obvious. Not all the noise reduction is due to the long 300 m height interval of the multichannel scaler.

FIG. 10. Demonstration of the excellent range-gate performance by recording the 313 nm daylight background in detection channels 3 and 4 (large telescope); the gain is switched from a reduced value to the normal one at a time corresponding to a distance $r = 2.9$ km (marked by T). The full rise time corresponds to a vertical interval of less than 100 m. The peak output voltage exceeds the settled value by less than 0.25% of the full signal step only.

FIG. 11. Logarithm of the range-corrected 277 and 313 nm backscatter profiles for the large telescope as recorded on August 6, 1991, at about 8:30 a.m., both profiles are corrected for the effects of Rayleigh scattering. In this representation, the 313 nm curve demonstrates the excellent quality of the recorded data.
which indicates the presence of additional noise in the analog channel. With the beginning of the thick cirrus cloud above \( r = 10 \) km, the extinction of the 277 nm light leads to enhanced noise also in the photon-counting channel. Below 5.5 km, pulse pileup starts to cause a lower count rate.

**D. Grating spectrograph**

Although reasonable results could be obtained with the present experimental setup, the high residual solar background level in the 313 nm lidar signal during the summer months, as seen in Fig. 11, and the resulting elevated noise level for \( r > 5 \) km are highly inconvenient. An occasional success in deriving vertical ozone density distributions beyond 10 km is possible by lowering substantially the vertical resolution in the upper half of the lidar profile. This is dictated anyway by the photon-counting system used at the moment, with its 300 m height interval. Nevertheless, with the advent of faster photon-counting systems with single-channel dwell times as small as 0.1 \( \mu s \), as well as with the demand for higher accuracy up to 12 km, a greatly improved daylight suppression by applying some superior spectral filtering becomes unavoidable. In order to maintain an operating range up to 12 km throughout the year a 292 nm wavelength channel has to be added. The 292 nm signal must not contain any 277 or 313 nm contributions. This is not possible with interference filters such as those used now.

The narrow bandwidth of the KrF laser permits, in principle, an almost quantitative elimination of the solar background. By using a stack of Fabry–Perot etalons with a transmission bandwidth of about 13 pm, Steinbrecht et al., in their stratospheric-ozone lidar, were able to lower the solar light flux to the detector to a level which allowed even single-photon counting. Obviously, a substantial amount of signal was sacrificed to pay for this capability.

For 313 nm in our system, there is no need for single-photon counting because of the demonstrated good quality of the analog signal at night even beyond 10 km. Thus, a detection bandwidth reduction by less than two orders of magnitude with respect to the interference filters is sufficient.

Fabry–Perot etalons are not applicable in a lidar system for tropospheric measurements. Due to a lateral beam walk of the light backscattered from the first few kilometers, the acceptance angle of the receiver must be quite large (several mrad), which also excludes an additional background reduction by efficient spatial filtering in the telescope focus. Fabry–Perot etalons suitable for such measurements substantially limit the exit divergence of the receiving telescope. For realistic parameters of such an etalon, a minimum plate distance of 50 pm and a maximum reflectance of 0.9, the exit divergence of the telescope must not exceed 3.6 mrad for 99.9% of backscattered 313 nm light to pass through the central spot of the fringe system. In view of the divergence magnification in the telescope this is very hard to achieve, even without taking into account the diameter limitations for such an etalon.

In the UV, the light dispersion by prisms is not sufficient to achieve the proposed increase in resolution. Thus, the only viable solution is a grating spectrograph. A grating spectrograph has successfully been applied to tropospheric ozone measurements by Ancellet et al. A 3-nm bandwidth is reported which is sufficient for the operating wavelengths of that lidar system below 300 nm. Grating spectrographs of different design were recently constructed for several other lidar systems (see, e.g., Refs. 27, 28, and 58).

As pointed out above, a receiver bandwidth of almost 0.1 nm is desirable for our KrF-laser-based lidar system. For this purpose, a concave holographic grating with 1995 mm radius and 2400 grooves per mm was purchased for each telescope (Carl Zeiss, Oberkochen). The grating efficiency at 300 nm was measured by the manufacturer to be more than 70% even for unpolarized light which guarantees transmissions for the almost linearly polarized backscattered light far beyond the capability of any combination of narrow-band UV filters. This is particularly valuable for the strongly absorbed 277 nm radiation. It is also important for the future simultaneous use of 292 nm light which will require the 248 nm beam to be split in order to pump two parallel Raman shifters and which will, thus, result in a transmission of approximately half the pulse energies presently available for the different Raman orders.

For a grating spectrograph, to a good approximation, the acceptance angle problems mentioned above for the Fabry–Perot etalon do not exist. The near-range beam walk of the backscattered light is restricted to the plane between the laser and telescope axes. If one orients the grooves of the grating along this plane the beam walk has no significant influence on the spectral separation. Even “one-dimensional” spatial filtering is possible by placing a slit in front of the PMT which is oriented parallel to the grooves. This helps to improve further the daylight reduction.

The spectrograph design is depicted in Fig. 12. Figure 12(b) shows the principal rays for the four main wave-
Thus, the wavelength separation should be close to the mini-
diameter of the grating. A 90°-deflecting parabolic mirror
of the telescope. This focal length is substantially larger than
with a 90°-focal length of 300 mm will be used as the ocular
grooves, the achievable spectral resolution will be deter-
bined by the beam divergence of the Raman-shifted laser
beam which could no longer be resolved after the installation
discrimination limit of about 0.1 mrad inside the laboratory.
Thus, the wavelength separation should be close to the mini-
value of 0.2 nm, which was determined by ray tracing and
and corresponds to the imaging errors of both the grating and
the 90° parabolic mirror.

The astigmatism is kept small by rotating the grating
within an angular range not too far from the Wadsworth
angle. The Wadsworth condition (exit angle \( \beta = 0 \)) for the
chosen incidence angle \( \alpha = 35° \) is almost met for 248 nm.
\( \alpha = 35° \) is a good compromise. It is selected to avoid a
potential Wood anomaly\(^{59,60} \) for 313.2 nm at \( \alpha = 30.22° \) and is
still low enough to guarantee a reasonable grating efficiency.
The grating efficiency will be further optimized by placing a
Fresnel rhomb into the KrF laser between oscillator and am-
plifier to provide the appropriate orientation of the polariza-
tion. For the purchased grating, only the zeroth and first or-
der order are present. The first order is suppressed by confining \( \alpha \)
to values greater than 24°. We recently realized that a
Wadsworth-type configuration was also chosen by McDer-
mild et al.\(^{26} \)

Figure 12(a) shows how the spectrograph will be
mounted at the side of the large telescope. Because of its
considerable size the number of adjustable optical com-
ponents are kept as small as possible. All components will be
contained in a black housing together with additional light
baffles shielding scattered photons.

The construction of the two spectrographs could not be
completed within the budget of this contract, which ended in

III. DATA EVALUATION

Perhaps the most difficult task of the lidar development
has been to prepare the computer program for the data evalu-
ation. This program has become rather elaborate and large
because of the numerous options involved and because of the
concatenation of the multiple lidar data segments from the
different detectors and range-gate intervals. Its size exceeds
640 kbyte storage limit for the applied programming lan-
guage, TURBO PASCAL 5.5 (personal computer: COMPAQ,
model Deskpro 386/20). Segmented loading and also tempo-
ral storage on disk is applied to overcome these problems.

The program consists of three blocks. Two of them serve
for inspection of the lidar profiles and the various operations
applied to them in different representation modes. The third
provides the final ozone density evaluation and will be out-
lined in the following.

A. Calculation of the ozone density

The expression for the ozone density calculation is ob-
tained from the derivative of the logarithmic lidar equation
\[
\frac{d}{dr} \ln \left[ \frac{r^2 S(r, \lambda) / \beta(r, \lambda)}{P(r, A)} \right] = -2 \alpha(r, \lambda),
\]
with the lidar signal \( S \), the total backscatter coefficient \( \beta \), and
the total extinction coefficient \( \alpha \). No \( r \) values within the rising
edges of the range gates are accepted (see, e.g., Fig. 11,
\( r = 2.8 \) km). Equation (1) is formed for the two DIAL wave-
lengts \( \lambda_1 \) and \( \lambda_2 \). Subtraction yields
\[
\frac{d}{dr} \ln \left[ \frac{S(r, \lambda_1) / \beta(r, \lambda_1)}{S(r, \lambda_2) / \beta(r, \lambda_2)} \right] = -2(\alpha(r, \lambda_1) - \alpha(r, \lambda_2)).
\]
Equation (2) is more or less equivalent to the traditionally
used Eq. (4.41) in Ref. 61 (DIAL equation). In contrast to the
latter it is exact. The derivative formation is solved numeri-
cally by a least-squares fit to
\[
\ln \left[ \frac{S(r', \lambda_1) / \beta(r', \lambda_1)}{S(r', \lambda_2) / \beta(r', \lambda_2)} \right] = 2 \int_0^r \left[ \alpha_s(r'', \lambda_1) - \alpha_s(r'', \lambda_2) \right] dr'',
\]
(with the “scattering” extinction coefficient \( \alpha_s = \alpha_R + \alpha_P \),
where \( R \) denotes “Rayleigh” and \( P \) “particles”; the calcula-
tion of \( \beta \) and \( \alpha \) is described in Sec. III B), as determined
from the experimental data for distances \( r' \) located in a given
interval which is symmetrically arranged around \( r \) wherever
possible. The derivative at distance \( r \) is calculated from the
coefficients of the fit polynomial. The least-squares fit is
based on a third-order polynomial which was chosen in view
of the excellent smoothing properties of cubic splines. As a
matter of fact, there was some indication that even-order
polynomials yield slightly inferior results.

The least-squares solution chosen is quite attractive as it
allows a direct error calculation. The standard deviations of
the ozone densities can be deduced directly from the data
noise by using the covariance matrix of the fit.

The size of the fit interval is closely related to the verti-
cal resolution interval \( \Delta r \) which one finds in the standard
DIAL equation. For example, in a linear polynomial the fit
interval is equal to \( \Delta r \) whereas for a cubic polynomial we
expect \( \Delta r \) to be, on average, about one third of the fit inter-
val, due to the possible existence of two extremes.

The interval size for the analog data is dynamically set
as approximately determined by the local signal-to-noise ra-
tio at distance \( r \). Minimum interval sizes are typically about
0.1 km (corresponding to \( \Delta r = 0.033 \) km). In order to avoid
excessive computation times, the maximum interval size is
set to 0.75 km which is reached just before \( r = 5 \) km. Slightly
above that altitude, photon-counting data are taken for the
evaluation with a constant interval size of 2.4 km which
comprises 9 points 0.30 km apart. The small number of
available photon-counting data per height interval is not ad-
vantagous for the slope and noise determination. As will be
further discussed below, this is particularly severe near the
upper end of the full evaluation range. Multichannel photon
continued across the switching points in an improved version of the program. This requires an additional parameter for the beginning and the end of the full range.


affect the calculation of the derivative. Thus, problems are encountered at the switching points and at the beginning of the corresponding fit interval. It was found that the derivatives at the ends of the interval may be larger than the signal ratio rather than to its logarithm above the highest switching point. Just a slight change in the derivative calculation algorithm was necessary. Polynomial fitting to the data in nonlogarithmic representation is not possible below the uppermost switching point because the calculation of derivatives across a switching point is only applicable for the logarithmic version.

For storage in a data base, the density at the central $r$ of each height interval may be picked after the end of the evaluation in order to avoid excessively large data files.

**B. Backscatter and extinction corrections**

Equation (2) does not directly yield the ozone density. The total backscatter and extinction coefficients

$$\beta(r, \lambda) = \beta_p(r, \lambda) + \beta_R(r, \lambda)$$

(3a)

and

$$\alpha(r, \lambda) = \alpha_p(r, \lambda) + \alpha_R(r, \lambda) + \alpha_R(r, \lambda)$$

(3b)

together with the contributions from Rayleigh ($R$) and particle ($P$) scattering. These contributions must be approximated using suitable models of the atmospheric density distribution or of the backscatter-to-extinction ratio for the light scattering by the aerosols. Because of the availability of an "on" and an "off" wavelength the calculation of the ozone density distribution and the light scattering coefficients can be treated separately.

This separation is not entirely perfect. Therefore, we have chosen an iterative approach. In the first step of this iteration, Eq. (2) is solved for $\beta_p = \alpha_p = 0$, which gives a crude initial guess for the ozone density. This preliminary ozone density distribution is then inserted into Eq. (1) which is then solved for $\lambda = 313$ nm to yield $\beta$ and $\alpha_p + \alpha_R$ (see Sec. III B 2). The corresponding coefficients for 277 nm are derived from these values by using a simple model for the wavelength dependence. Afterwards, an improved ozone distribution is computed now including all the contributions specified in Eqs. (3). These steps can be repeated until the ozone densities from two consecutive iterations do not differ by more than the desired tolerance level. Usually, such a repetition is not necessary because, in the vast majority of all cases, the second ozone density calculation reproduces the
The agreement with the cross sections by Yoshino et al.,\textsuperscript{64,65} this requires an enhancement of the values by 1.9%. The agreement at the mercury wavelength 253.7279 nm with the results by Mauersberger et al.\textsuperscript{66} A slight wavelength shift with respect to the data by Molina and Molina\textsuperscript{63} is reported. The agreement with the cross sections by Yoshino et al., Malicet et al. and Mauersberger et al. care-\textsuperscript{67,68} Some uncertainty arises from the depolarization factor of the Rayleigh backscatter and extinction coefficients (see, e.g., Refs. 67 and 68). For the present work, we used the value for unpolarized light as given in Ref. 68 which means a multiplication of the uncorrected Rayleigh cross section by 1.0608. Attempts to determine physically meaningful expressions for linearly polarized light from the theory given in Ref. 67 have been unsuccessful. The signal extinctions calculated for two mutually orthogonal linear polarization components of the incident radiation differed by roughly a factor of three, which should not be the case for randomly oriented molecules. Penney confirms that the classical theories have assumed a fixed orientation of the molecular axis,\textsuperscript{69} which does not influence the calculation for natural polarization, but is not acceptable in general. Nevertheless, we found that the mean value of the two calculated extinction coefficients is identical with the value for unpolarized light. This may explain why no significant discrepancies could be observed in the course of the evaluation of our backscatter profiles. For finer details, the discussion in the review article by Young should be taken into consideration.\textsuperscript{70}

2. Aerosol contribution

In the presence of aerosols the backscatter and extinction corrections may become somewhat less reliable. This is due to the uncertainties concerning the light-scattering properties of aerosols and the applicability of the inversion methods. In our program, the total backscatter coefficients [Eq. 3(a)] and the extinction coefficients for atmospheric scattering, are derived from the 313 nm profiles by using Klett's inversion method.\textsuperscript{71} Klett discusses a numerically stable integration scheme for Eq. (1). The Rayleigh scattering coefficients are supposed to be known with sufficient accuracy. Because the particle backscatter and extinction coefficients, $\alpha_p$ and $\beta_p$, both have to be determined from a single equation, a simple relation

$$\beta_p - B_p(\alpha_p)\beta_p,$$  \hspace{1cm} (4)

is assumed which finally reduces the number of unknown quantities to one. The ozone contribution to the total extinction coefficient [Eq. 3(b)] is not very critical at the "off" wavelength 313 nm, but is, nevertheless, provided by a preliminary calculation of the ozone density distribution. The corresponding values for 277 nm are obtained from those at 313 nm by assuming a $\lambda^{-4}$ wavelength dependence. This may be a gross simplification, particularly in the UV,\textsuperscript{72}
but so far has yielded acceptable results. Empirical values for the exponent \( \varepsilon \) were determined by two simultaneous measurements with two of the IFU lidar systems at different wavelengths to range within 0.7 and 0.9 in Winter and within 1.3 and 1.6 in Summer. These results demonstrate the wide range of exponents one has to take into consideration. The lower values in Winter might indicate the presence of ice crystals for which \( \varepsilon = 0.73 \).

A simple, analytical solution of the Klett inversion is obtained if \( k = 1 \) in Eq. (4). However, as demonstrated by Klett, this does not impose too much of a restriction since the best inversion results were obtained for \( k = 1 \) and an empirically determined \( B_\rho(\alpha_\rho) \) function

\[
B_\rho(\alpha_\rho) = 0.0074 + 0.055 \exp\left(-\frac{4}{3.1}\right) \text{ (presumably sr}^{-1}\right), \tag{5}
\]

which implies a variation of \( B_\rho \) between 0.0074 and 0.0624 sr\(^{-1} \) for the aerosols examined by Klett.

Equation (5) cannot be directly applied because no units are specified. In addition, other types of aerosol than that examined in Ref. 71 might lead to different expressions. The greatest problem is that \( B_\rho(\alpha_\rho) \) is an input quantity for the inversion, but \( \alpha_\rho \) is not known at the beginning of the calculation. Therefore, an iterative approach is necessary to solve Eqs. (1) and (5) [or some other expression for \( B_\rho(\alpha_\rho) \)] simultaneously. An iterative solution with variable \( B_\rho(r) \) was recently discussed by Kovalev. Since the aerosol distribution depends on the height, the application of such a procedure may become important as soon as the technical performance of a lidar system allows ozone measurements with errors next to \( \pm 1 \) ppbv.

In the present version of our program, we preferred to use constant \( B_\rho \) (Fernald solution\(^75\)). This approximation was found to yield acceptable results at least outside clouds. We tested values between 0.017 and 0.040 sr\(^{-1} \). It turned out that the choice of \( B_\rho \) is not very critical and typically results in ozone density variations of the order of just a few times \( 10^{10} \text{ m}^{-3} \) in that \( B_\rho \) range. We finally selected \( B_\rho = 0.030 \) sr\(^{-1} \) which, subjectively judged, gave the best agreement with ozone density distributions from ECC sondes. This value is in almost perfect agreement with the mean value \( B_\rho = 0.031 \) sr\(^{-1} \) evaluated by Reagan et al. from measurements in Arizona between 1979 and 1982.\(^76\)

As prescribed by Klett, the inversion is started at the upper end of the evaluation range. There, a reference value \( \beta_m \) for \( \beta \) must be chosen (integration constant). The choice of \( \beta_m \) is very critical for the calculation of \( \beta \) in the case of low optical thickness which prevails in the free troposphere. Fortunately, for the short wavelengths used, Rayleigh scattering is mostly the dominating process outside the cirrus-/\( R(\rho) \) is possible in all but a limited number of cases.

The integration is carried out in 5 m steps which guarantees high accuracy. It is interrupted at each switching point (change of data channels or PMT gain, see Sec. III A). If data points are missing around such switching points the \( \beta \) values are incremented by the corresponding differences of the Rayleigh backscatter coefficients. Great care is taken to allow a correct execution for all possible mutual positions of the six switching points, which causes considerable branching of the algorithm.

Figure 14 shows examples of vertical distributions of the backscatter coefficient \( \beta \) obtained by Klett inversions of the 313 nm profiles from the measurement underlying Fig. 12 using \( B_\rho = 0.020, 0.030, \) and 0.040 sr\(^{-1} \). The Rayleigh backscatter coefficient \( \beta_R \) is included for comparison.

![Fig. 14. Backscatter coefficients \( \beta \) obtained by Klett inversions of the 313 nm profiles from the measurement underlying Fig. 12 using \( B_\rho = 0.020, 0.030, \) and 0.040 sr\(^{-1} \). The Rayleigh backscatter coefficient \( \beta_R \) is included for comparison.](image-url)
variation of the ozone density. In the example of Fig. 14, $B_P$ values between 0.020 and 0.040 sr$^{-1}$ were chosen. However, in this $B_P$ range the maximum departure from the ozone density distribution calculated for $B_P=0.030$ sr$^{-1}$ was just $\pm 2.5 \times 10^{16}$ m$^3$ (i.e., $\pm 2\%$ of the density) at $r=1.8$ km. This is rather surprising because the aerosols lead to $B$ values almost twice as large as $B_R$ in the boundary layer, which vary markedly as a function of $B_P$. The small influence of $B_P$ can be understood as follows. The aerosol-induced contribution in Eq. (2) would be

$$-\frac{d}{dr} \ln \frac{\beta(r,\lambda_1)}{\beta(r,\lambda_2)} + 2[\alpha_P(r,\lambda_1) - \alpha_P(r,\lambda_2)]. \quad (6)$$

Due to the moderate wavelength dependence of the aerosol scattering coefficients, the extinction part rarely exceeds the corresponding contribution from Rayleigh scattering. As in the Rayleigh case, it consistently lowers the ozone densities (by $\pm 1.1 \times 10^{17}$ m$^{-3}$ in this example) whereas the backscatter part can assume either sign. The logarithm of the $B$ ratio should not vary much with $B_P$. As can be seen from Fig. 14, $\beta(r, 313$ nm) grows with increasing $B_P$. Since $\alpha_P = \beta_P/B_P$, also both $\alpha_P$ values as well as their difference in (6) should be almost constant. This confirms the approximate overall $B_P$ independence.

As one can see from Fig. 15, the aerosol correction itself is not at all negligible. It may be positive or negative, depending on the magnitude of the gradient of the backscatter coefficient as shown in the following.

The backscatter contribution

$$-\frac{d}{dr} \ln \frac{\beta(r,\lambda_1)}{\beta(r,\lambda_2)} = 0, \quad (7)$$

if either $\beta_R$ or $\beta_P$ vanish. If, as it is the case in a realistic atmosphere, both do not vanish, Eq. (7) is still valid if

where ' means derivative formation. Equation (8) is obtained by assuming a $\lambda^{-\epsilon}$ (or even more general) wavelength dependence for the aerosol backscatter coefficients and a $\lambda^{-4}$ wavelength dependence for the Rayleigh backscatter coefficients. By definition, the same equation also holds for $\lambda_2$. If the derivative ratio on the left-hand side in Eq. (8) is greater than the ratio on the right-hand side, a compensation (or even overcompensation, see below) of the extinction term in (6) may be possible. If the opposite is the case, the backscatter term enhances the extinction term. A comparison of Figs. 14 and 15 nicely confirms this prediction. With the exception of the range below 1 km, where the extinction term shifts the curve down by up to $1.1 \times 10^{17}$ m$^{-3}$, the differences between both curves in Fig. 15 essentially reflect the variation of the backscatter correction.

It is obvious that $\beta_P'$ is one of the key quantities for backscatter as well as, of course, for the total aerosol correction of the ozone density distribution. In the examples of Fig. 14, the gradient of the backscatter coefficient is moderate with the exception of $r \approx 2.0$ km. With the exception of $r \approx 2.0$ km, the aerosol extinction and backscatter corrections within the boundary layer are, consequently, also quite moderate ($\pm 1 \times 10^{17}$ m$^{-3}$ or 5 ppbv). The chosen $\lambda^{-1.5}$ wavelength dependence (Summer) also contributes to this fact. For comparison, an application of $\lambda^{-0.7}$ (Winter) would result in ozone densities higher by up to $7 \times 10^{16}$ m$^{-3}$ (3.4 ppbv) at $r=1.1$ km (see Fig. 16) which is not supported by the results from a simultaneous ECC-sonde ascent (see Sec. III C).

At around $r=2.0$ km, the strongest gradient of $\beta$ is calculated. As indicated above, this causes an overcompensation of the extinction contribution to the aerosol correction.
sequently, the ozone density is appreciably enhanced by up to \(2.8 \times 10^{17} \text{ m}^{-3}\).

The importance of the gradient of the backscatter coefficient for the correction of the ozone distribution has been discussed in the literature, as deduced from simulations (see, e.g., Refs. 24 and 77). Extensive systematic simulations, using artificial backscatter profiles and profiles from our measurements, were also carried out by Kempfer as a part of his thesis,\(^\text{77}\) but are not reported here. These calculations reveal finer details of the \(B_P\) and \(\epsilon\) dependence of the aerosol correction algorithm as well as its limitations.

The observed variations of the ozone density due to different assumptions for the aerosol correction are not significant at the present accuracy level. As soon as the accuracy approaches 1 ppbv, more work will be necessary to characterize fully the aerosol light scattering properties under our local conditions and as a function of season. In particular, their wavelength dependence has to be analyzed in more detail which, in our opinion, constitutes the most severe potential error source. A range-dependent choice of \(B_P\) should be considered. With the planned grating spectrograph, Raman lidar measurements may also be possible. This would, to some extent, allow an independent determination of the aerosol backscatter and extinction coefficients.\(^\text{72,77}\)

**C. Validation of the ozone distributions and discussion of the error contributions**

The profiles of Fig. 11 were recorded simultaneously with an ECC ozone-sonde ascent. This ascent took place under favorable conditions so that, for the first few kilometers, the balloon rose almost vertically, followed by a lateral drift of not more than 5 km up to \(r=10\) km. Figure 17(a) compares the final lidar ozone distribution (\(B_P=0.030\) sr\(^{-1}\), \(\epsilon=1.5\)) with that obtained from the sonde and also with the half-hour averages of the local monitoring stations of the IFU near the institute (G, 730 m a.s.l.), on the summit of the Wank mountain (W, 1780 m a.s.l.) and on the summit of the Zugspitze (Z, 2960 m a.s.l.). The ECC data had to be multiplied by 0.72 for an adjustment to the ground value. The agreement of the corrected values with the distribution from the lidar measurement is highly satisfactory.

On August 6 and 7, 1991, the IFU aircraft group crossed the Alps several times in north-south direction in a series of successive ascents and descents.\(^\text{79}\) These flights were carried out as a part of the MEMOSA campaign. The longitudinal and vertical distributions of ozone, other trace gases, as well as of the most important meteorological parameters, were measured along the flight path in order to determine the major origin of air pollution in the Alps. During the first half of the 8:30 lidar measurement on August 6 a descent of the aeroplane over Garmisch-Partenkirchen took place. This gives us a good opportunity to compare our results. The lower section of Fig. 17(a) is expanded in Fig. 17(b) and shown together with the MEMOSA aircraft data.\(^\text{78}\) The agreement is once more very convincing, apart from some minor discrepancy around \(r=2.3\) km which, as to the lidar data, is most likely due to a not perfectly averaged single-bit step. Such steps sometimes become observable in the low-signal region just below the range-gate switching height (see Sec. II C).

A total of four ECC-sonde ascents were carried out in 1991. Only two of them could be evaluated because of data-transmission problems. Throughout the year, the data of the IFU monitoring stations were also taken for comparison. If the air masses do not arrive from across the Alps (Fohn winds) the ozone densities of the different methods usually coincide within the specified errors.

Despite this success, which exceeds our initial expectations, a realistic error analysis shows that one needs to make a number of improvements in order to reach the accuracy levels proposed in Sec. III B 1. The main concern is the quality of the backscatter signal which we expect to enhance further. However, it is obvious that the ozone-evaluation al-
The algorithm also contributes to the overall error. The main variation is caused by the choice of the beginning and size of the actual evaluation intervals. The errors originate in this case, e.g., different positions of the intervals with respect to local ozone maxima or minima and exhibit roughly the same behavior for all three evaluation methods mentioned. Included in this are discontinuities as one goes from one interval to the next. Evaluation-induced step sizes in the ozone distributions of more than $5 \times 10^{16} \text{ m}^{-3}$ have been observed although only values from the inner third of the fit interval are taken. A few examples can be seen in Figs. 17, e.g., at $r = 0.6$ km where a step of about $3.5 \times 10^{16} \text{ m}^{-3}$ occurs, or at $r = 1.55$ km where the switching point between a small and large telescope was chosen. Of course, the error may be expected to be smaller at the center of a fit interval. The ozone value at the center is the one used for database storage.

In the future one should consider the use of spline fits instead of overlapping polynomials in order to avoid such steps. In addition, a more refined strategy for the positioning of the fit intervals is advisable. Together with the technical improvements this should eventually yield an optimum approximation of the ozone distributions.

By comparison with the ECC-sonde data and by including guesses for the noise and evaluation-induced errors we arrive at an error limit of, on average, $7.5 \times 10^{16} \text{ m}^{-3}$ (i.e., $\pm 3.1 \text{ ppbv near the ground}$) for our system under the present conditions. The density error as a function of $r$ is almost constant because of the dynamic choice of the evaluation-interval size according to the local signal level (see Sec. III A). Within the uppermost two kilometers of the useful evaluation range, the error may grow to about $1.5 \times 10^{17} \text{ m}^{-3}$ because of the uncertainties concerning the $277 \text{ nm corrections}$ described in Sec. II B.

**IV. APPLICATION: THE 1991 SERIES OF OZONE MEASUREMENTS**

In 1990, following its completion, the lidar system was operated exclusively for testing purposes. In 1991, a first annual series of ozone measurements with about 580 vertical profiles was completed which demonstrates the capabilities of the lidar method for routine measurements. Because of ongoing improvements of the data evaluation procedure, and because of the considerable time required at that stage of the project to calculate a single vertical density distribution only less than one third of these profiles has been evaluated. In this section, we confine ourselves to the description of a few general features of our observations. A more complete discussion, which will also include more about the scientific goals, will be published elsewhere.

The measurements took place exclusively under fair-weather conditions (during periods of high pressure as well as before and after frontal passages) and at time intervals of typically 1–2 h, which is sufficient for a determination of the diurnal variation of the ozone density. The backscatter signal was averaged for 15,000 laser shots emitted with a repetition rate of about 20 Hz. Because the lidar operation is not yet fully automatic just a limited number of night-time measurements have been performed so far.

Figure 18 summarizes the results for 1991 in a three-dimensional plot. All the evaluated profiles are included in this figure and smoothed by the graphics program. In order to make the basic structure of the ozone density distributions clearer three representative ozone profiles obtained in Winter and Summer 1991 are given in Fig. 19.

Throughout the year, there are enhanced ozone densities in a range up to $r = 2-4$ km above the ground. We define this range, which frequently exceeds the planetary boundary layer, as the "extended boundary layer" (EBL) since the high ozone concentrations must originate in some slow upward transport of $\text{O}_3$ and/or its precursor molecules. In the free troposphere above the EBL, the observed ozone densities are about half the peak values in the EBL throughout the year. In the entire troposphere, substantially higher ozone densities are found, especially in the EBL.
densities are observed more in summer than in winter due to the photochemical production mechanisms. In the EBL, up to \(1.6 \times 10^{18} \text{ m}^{-3}\) (corresponding to about 67 ppbv at the typical position of the maximum, \(r = 0.8 \text{ km}\)) accumulate in Summer, whereas between November and January, \(1.0 \times 10^{18} \text{ m}^{-3}\) are rarely reached. Nevertheless, the occurrence of enhanced ozone densities in the EBL even during the period of the lowest sun elevation is quite remarkable.

The ozone distribution in the EBL exhibits a characteristic two-step pattern, with the highest densities being observed near the top of the lower of these sublayers (\(r = 0.8 - 1.0 \text{ km}\)). This pattern is seen not only during periods of stable high pressure, it is also quickly restored after a frontal passage (within a single day). This two-layer structure is frequently (but not exclusively) confirmed by the distribution of the aerosol backscatter coefficients (see, e.g., Fig. 14). An important finding is that the vertical variability of the principal structure of the ozone density distribution is mostly low, even in cases when the aerosol measurements indicate considerable changes inside the EBL. This confirms that there is little convection above the well-mixed layer.\(^8\)

It is obvious that the results of the monitoring station in the valley are not representative of the ozone concentrations in the lower troposphere. The considerable diurnal changes recorded here far exceed those observed at higher altitudes. They are mostly caused by the local meteorological conditions near the ground. The strong ozone decrease in the evening is synchronized with the onset of the downslope winds.\(^8\) Since these winds originate in an ozone-rich zone one has to invoke efficient ozone destruction in the vegetation as a possible explanation for the low \(O_3\) values in the valley during the night. Kelly et al. concluded that dry deposition of ozone is the dominating nighttime ozone-depleting mechanism in rural areas with low NO concentrations, even without the assistance of a wind system like that in our alpine valley.\(^8\) They suggested the presence of a low-lying nocturnal inversion layer to be responsible for the enhanced deposition in the cases studied in Ref. 82. The concentration increase near the ground before noon is mostly attributed to downward mixing of ozone from aloft, started by the onset of convection.\(^8\)

In contrast to this, the much smaller ozone concentration changes above \(r = 0.5 \text{ km}\) demonstrate that the results of the IFU monitoring stations on the Wank (1780 m a.s.l.) and, in particular, on the Zugspitze (2960 m a.s.l.) should be much less affected by interaction with the ground. At these stations, long-term trends of ozone and some of its precursors at the respective altitudes are determined. The concentrations (half-hour averages) measured there are mostly in excellent agreement with those from the lidar measurements at the corresponding heights. This demonstrates the reliability of both methods as well as the good horizontal mixing of ozone under the prevailing wind conditions, i.e., if the air does not arrive from across the Alps. The lidar, of course, is able to provide information for all altitudes. In particular, the positions of the maximum ozone densities can be localized, as well as the regions where strong concentration gradients occur. In one case, a slow decrease of the ozone concentration registered at the Zugspitze station could be related to a downward motion of the upper gradient zone. This shows how both methods can be effectively combined for an optimum selection of the ozone data.

V. PLANS FOR THE FUTURE

The results presented in the previous sections demonstrate the suitability of the IFU ozone lidar system for ozone measurements in almost the entire troposphere, with a reasonable vertical resolution. The accuracy for the chosen range-dependent evaluation interval is estimated to be about \(\pm 7.5 \times 10^{16} \text{ m}^{-3}\) (i.e., \(\pm 3.1 \text{ ppbv near the ground}\)). For the final 2 km, the error is higher due to a growing sensitivity of the derived densities to the correction of the signal offset and to the signal-induced contributions (see Sec. II C).

Improvements in the specifications so far achieved are possible. By further reducing the receiver bandwidth the daytime performance will be optimized and the vertical range will be extended by adding the 202 nm wavelength channel. A substantially higher receiver transmittance may contribute to a better signal-to-noise ratio for 277 nm. The data acquisition will be fully automated in order to guarantee a full 24 h availability of the system. Thus, continuous one- to three-week series of ozone measurements will be possible which would allow us to trace all the interesting observations back to their beginnings.

The 1991 and future measurements were and will be carried out within the framework of the TOR ("tropospheric ozone research") Subproject of EUROTRAC. We hope that we may contribute to a better understanding of the tropospheric ozone budget and some of the processes controlling it. Topics of particular interest are the effective ozone production in the EBL as well as the ozone exchange between the EBL or the stratosphere and the free troposphere. The availability of precursor concentrations from monitoring stations at different altitudes up to about 3000 m a.s.l. offers the almost unique chance to quantify to some extent the chemical processes involved. This is particularly interesting in the case of the puzzling distributions observed in winter.

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