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1991 AND 1993 TROPOSPHERIC OZONE MEASUREMENTS AT IFU

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INTRODUCTION

In 1990, a wide-range tropospheric ozone lidar was completed at IFU. It is based on a KrF excimer laser, Raman shifted in hydrogen to yield the operating wavelengths 277 nm and 313 nm. A single measurement with typically 15000 laser shots can be carried out in less than 5 min. The vertical range is presently limited by the ozone density, which greatly influences the light absorption (signal loss) at 277 nm, and the noise from residual daylight in the 313 nm detection channel to about 12 km in winter and 8 km in summer. The near range profiles can, in principle, be evaluated starting at less than 0.15 km above the ground. Due to small oscillations superimposed on the signal it has so far been difficult to obtain good results for distances below 0.3 km.

The operating range corresponds to more than eight decades of backscatter signal. It is therefore divided into subranges by using two receiving telescopes, detector range gating and single-photon counting for 277 nm. Although this has resulted in a highly acceptable system performance there have been severe consequences for the data evaluation program which has become rather long in the meantime.

From comparisons with the data of the IFU ozone monitors on mountain summits as well as with distributions from ECC-sonde ascents we estimate the density error of our system to about 7.5×10^{16} m^{-3} (i.e., 3.1 ppbv near the ground). This error is quite constant as a function of the height because the vertical resolution is dynamically chosen based on an estimate of the varying signal-to-noise ratio. The most significant contributions to the overall uncertainty are presently given by the not precisely known wavelength dependence of the light backscattering and extinction by the aerosols as well as by evaluation-induced errors. In the near range also the quality of the backscatter signal matters. Details on this lidar system are given in a recently submitted paper[1].

RESULTS

The lidar has successfully been applied for ozone measurements since 1991. The 1991 series consisted of about 580 backscatter profiles. Only less one third of these profiles could be evaluated because of the considerable evaluation time required at that stage of the experiment. Nevertheless, this clearly demonstrates the advantages of lidar in comparison with conventional vertical-sounding methods for which measurements at short time intervals are more or less impossible. In 1992, no measurements were performed due to a lack of funding. Although no grant was available also in 1993 a limited amount of profiles was recorded. Preliminary examples from the 1991 series have been given on previous occasions[2,3]. A more detailed account of the results obtained up to now will be published soon[4].

Our measurements are carried out as a part of the TOR ("Tropospheric Ozone Research") subproject of the European EUROTRAC programme. Scientific goals for the lidar work are to gather information for the evaluation of the ozone budget in the different ranges of the troposphere and of the input from the boundary layer or the stratosphere into the free troposphere.

Fig. 1 shows, in a three-dimensional representation, the smoothed results of the 1991 annual series. A clear seasonal variation of the densities by approximately a factor of two is seen in the entire troposphere which is ascribed to the photochemical ozone production mechanisms. Apart from this variation, the basic structure of the density distribution does not change much during the year. The density in the free troposphere ($r > 3$ km) is typically about 50 % of the maximum value inside the boundary layer. If one omits the lowest few hundred metres above the ground, where the ozone concentration is frequently lowered by deposition or chemical destruction, the boundary-layer ozone almost consistently exhibits a two-step structure which is visible in Fig. 1. In the lower of the two steps high densities occur with average maximum values around $1.6\times10^{18}\,\mathrm{m}^{-3}$ (67 ppbv) in summer which exceed the densities reported for the end of the last century[5] by almost a factor of seven. The anthropogenic origin of these high concentrations is therefore obvious.

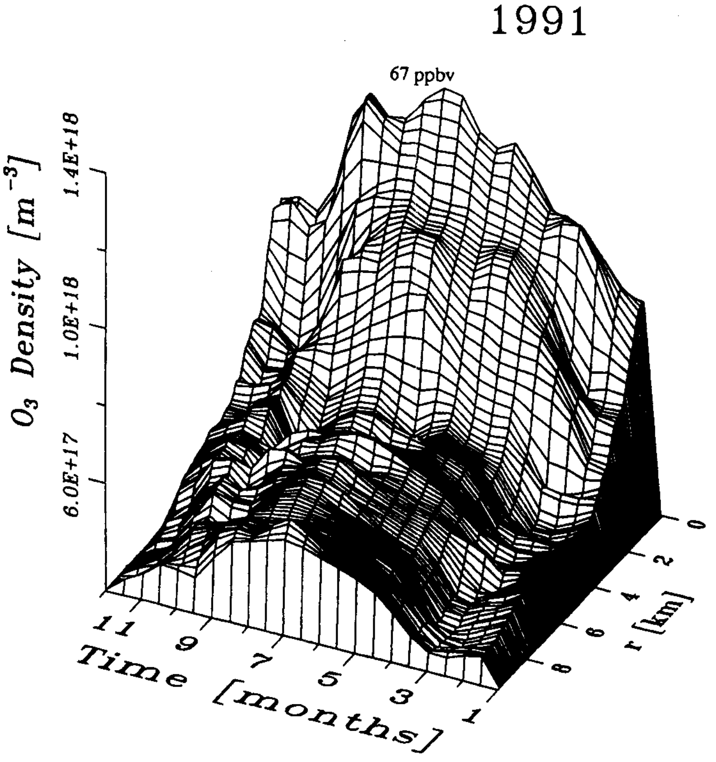

Fig. 1: Three-dimensional representation of the 1991 annual series of ozone soundings with the IFU lidar; r = 0 corresponds to 0.74 km a.s.l.. Please, note the offset of the density scale.

In summer, the two-step structure may be explained by the expansion of the well-mixed layer[6] during the sunshine hours which implies some upward transport of ozone to altitudes even beyond 3 km. In winter, a similar process cannot be invoked since convection is restricted to heigths below 1 km above the ground. The observation of enhanced ozone densities in such an "extended boundary layer" (up to 2.5 km) also from November till February in a two-step distribution is puzzling and calls for clarification.

The vertical ozone transport is almost negligible in the absence of convection. On several occasions, we observed changes in the thickness of aerosol layers, but no corresponding change in the boundary-layer ozone distribution. In contrast, the influence of horizontal transport is pronounced and can nicely be traced in the case of air-mass changes.

Most of the observed enhanced ozone densities are caused by human activities. The strongest natural source for tropospheric ozone is expected to be subsidence of ozone-rich stratospheric air. Such events take place particularly frequently in spring[7]. In the evaluated 1991 data we did not find any indication of a subsidence process.

A first example of a stratospheric air intrusion was registered on July 24, 1993, following a frontal passage which implied low ozone densities in the lower troposphere. Three ozone distributions from this day are depicted in Fig. 2 which show the progress of the subsidence process. The evaluation of meteorological data confirms all our observations[8].

FUTURE ACTIVITIES

In 1994, an extension of the lidar system is planned which had to be postponed for more than three years because of the funding problems mentioned above. A new wavelength will be added (292 nm) which is expected to yield more accurate results in the upper troposphere and to extend the operating range to at least 15 km. The necessary improved wavelength-separation capability will be provided by grating spectrographs which will be added to the two receiving telescopes. We expect a detection bandwidth reduction by almost a factor of fifty. Thus, also a greatly improved daylight suppression in the 313 nm channel will be achieved.

A part of the system modification will be to implement automatic system control. This will result in full night-time availability of the instrument and eventually enable us to run almost continuous two- or three-week series of ozone measurements which are necessary for an improved understanding of the contributions to the tropospheric ozone budget.

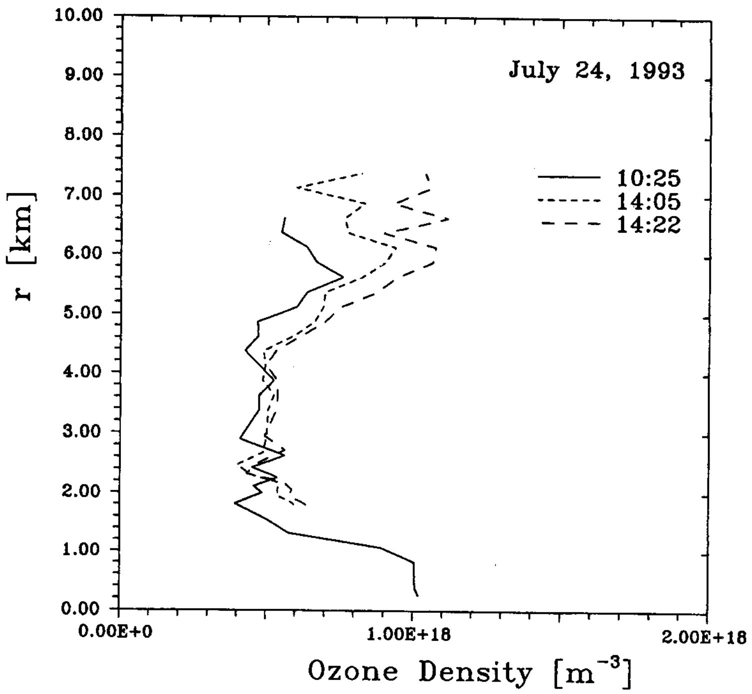

Fig. 2: Three ozone density profiles obtained on July 24, 1993, in the presence of stratospheric-air subsidence; unfortunately, the useful range was restricted to less than 6.5 km on that day because of unexpected problems in the photon-counting channel.

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