Research Articles

Application of Ground-Based Lidar for Studies of the Dynamics of Ozone in a Mountainous Basin

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DOI: http://dx.doi.org/10.1065/espr2001.12.106

Abstract. A ground-based Differential Absorption Lidar was employed to study the dynamics of atmospheric O_3 within the planetary boundary layer of a basin in the 'Fichtelgebirge' mountains, NE Bavaria. In particular, the night-time dynamics of O_3 linked to the ground were investigated. The Lidar system measured vertical profiles of O_3 up to 1 km above ground. For detailed analysis of the night-time dynamics of ozone, supplementary data from three ground-based stations (measuring mixing ratios of O_3 and NO_x, as well as meteorological parameters) are essential. The Lidar results could be evaluated with these data from various altitudes above the basin floor. For the station with the largest (vertical) distance to the ground-based Lidar, the agreement was very good at all times. The Lidar method proved to be useful for examining the spatial distribution of O_3. The observed night-time decrease of O_3 at the bottom of the basin was due to deposition and to advection of air masses containing less O_3 from the mountain slopes.

Keywords: Air pollution; atmospheric chemistry; boundary layer; deposition; katabatic wind; lidar; meteorology; mountain wind; nitrogen oxides; ozone; remote sensing; troposphere

Introduction

The analysis of long-term measurements of atmospheric ozone (O_3) indicates an apparent increase within the past hundred years of the annual mean O_3 concentration in the northern hemisphere from 10–20 ppb to 30–35 ppb (Solomon et al. 2000). This trend has also been observed at rural weather stations in Central Europe, such as the Fichtelgebirge mountains (Klemm and Lange 1999). O_3 is an important trace gas in the troposphere, affecting its radiative properties and numerous homogeneous and heterogeneous reactions. It also has toxic impacts on plants, animals, and humans. For these reasons, knowledge about the distribution and fate of O_3 is an important environmental issue. Four factors determine the abundance of O_3 in the lower troposphere: emissions of the precursors NO_x and reactive volatile organic compounds (VOCs), atmospheric production and destruction of O_3, turbulent transport, and deposition onto surfaces. A decrease of tropospheric O_3 levels during the night is very often observed at ground stations. Four principal reasons, which vary with spatial and temporal conditions, for this decrease can be identified: reaction of O_3 with freshly emitted NO, reaction with aerosol particles (maximum 25% reduction by hydrolysis of N_2O_5, after Jacob et al. 2000), advection of air containing less O_3, and surface deposition (Wesely and Hicks 2000, Klemm and Mangold 2001). Detailed knowledge about the horizontal and vertical distribution of O_3 in the atmospheric boundary layer helps to understand the roles of various processes leading to the observed O_3 concentration changes at a given station. Detailed analysis of the spatial and temporal changes of O_3 concentration and the forces driving change, particularly in complex terrain, is needed to develop further understanding of the mesoscale dynamics of O_3 in the lower atmosphere.

One technique used to measure vertical O_3 profiles is the optical remote sensing technique 'Light Detection and Ranging' (Lidar). The Lidar technique has a higher time resolution than balloon and aircraft measurements, and can also make simultaneous measurements at different heights. For these reasons, Lidar measurements should be useful for understanding regional O_3 dynamics. The scope of this study is the examination of the applicability of the Lidar technique for the study of the dynamics of O_3 in the lower parts of the boundary layer within the Weissenstadt basin in the Fichtelgebirge mountains, NE Bavaria. The usefulness of the Lidar results for understanding the processes driving the night-time ozone dynamics is discussed.

1 The Lidar Technique

Lidar emits a short laser-pulse at given wavelength and given duration into the atmosphere (VDI 1999, Weidauer 1998). The laser pulse is absorbed and scattered by atmospheric molecules through Rayleigh-scattering and by particles through Mie-scattering. The backscattered fraction of light within the field of view of the coaxial telescope is collected and transmitted to the detector. To improve the signal-to-noise ratio several laser shots of each direction have to be added up.

Differential Absorption Lidar (DIAL) uses two similar wavelengths to calculate the concentration of a given gas (such as O_3) and to minimize possible differences of scattering and extinction behaviour by atmospheric constituents other than the one being measured. The combination of the Lidar equations for the two wavelengths yields the concentration of the detected gas as a function of the distance to the Lidar instrument.

The used DIAL system ('Lidar 510M', manufactured by Elight Laser Systems GmbH) uses the reference-wavelength 286.3 nm and the measure-wavelength 282.4 nm at the long-wave range of the Hartley absorption band of O_3 (O_3 absorption cross-sections 21.49×10^{-18} and 31.41×10^{-18} cm^2).

2 Experimental Set-up

Over several days in May 2000, O_3 vertical profiles were measured from a location 625 m a.s.l. in the Fichtelgebirge, in the valley of the Eger River, near the municipality of Weissenstadt (**Fig. 1**). Three additional ground-based stations providing continuous measurements of O_3, NO_x and meteorological parameters were used. One station was situated next to the Lidar system. A second was on a 52 m high tower platform at the summit of the Schneeberg, the highest peak in the Fichtelgebirge (1051 m a.s.l.). The third station was the ecosystem research site Waldstein, 765 m a.s.l. (c.f. Klemm and Lange 1999).

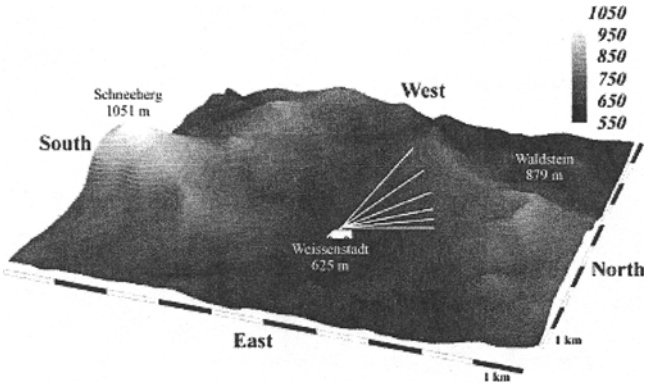

Fig. 1: Three-dimensional model of the topography of the Weissenstadt basin in the Fichtelgebirge mountains (height in m)

The Lidar measurements were performed in 2-dimensional scans over a meadow with seven different north-eastern shot directions, each with 1100 shots added up and characterised by its zenith angle. One scan lasted approximately 10 minutes and measured one vertical O_3 profile. Within each scan, the zenith angles of –91°, –89.5°, –88°, –85°, –80°, –65°, and –45°, respectively, were chosen in order to cover the near-surface air masses with the best possible representativeness. The calculation of mean vertical O_3 profiles and the corresponding inaccuracies were performed following standard routines (VDI 1999).

O_3 concentrations measured with the Lidar had to be converted to mixing ratios in order to compare the Lidar data to mixing ratios measured at the ground stations. This was done using the ideal gas law after estimating the respective air temperatures at different heights through interpolation of temperatures measured at the Waldstein and Schneeberg stations. About 87% of the interpolations were in the range between an isothermal and dry adiabatic lapse rate. The potential error in the O_3 mixing ratio resulting from the interpolation routine of the air temperature was below 1% and thus negligible as compared to the accuracy (around 5 ppb) of the Lidar measurements. The same was true for the error resulting from assuming an atmospheric vertical pressure gradient of $\partial p/\partial h$ = –12 Pa/m. The correction of the different backscatter coefficients at the two wavelengths is calculated following standard routines as suggested by the manufacturer (Elight Laser Systems GmbH 2000; Weidauer 1998).

3 Results

The results of our night-time Lidar measurement of the 15. and 16. May and the ground station results from 13. to 16. May are presented. During these days, a high pressure sys-

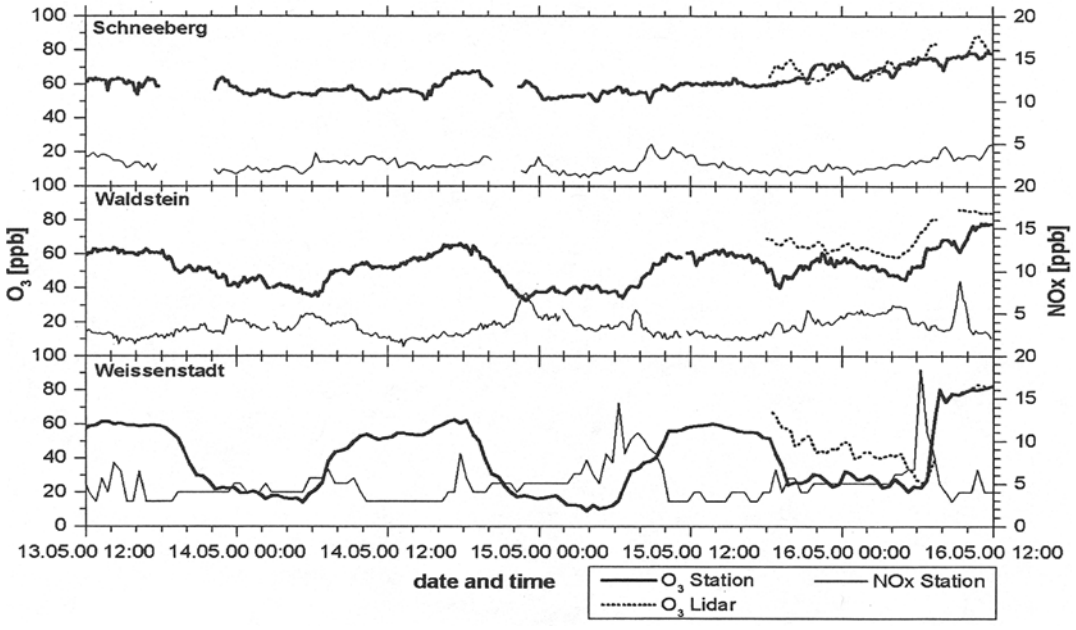

Fig. 2: O_3 and NO_x mixing ratios at the three ground stations, as well as the O_3 Lidar values at corresponding heights from May 13 to May 16, 2000

tem with almost cloudless conditions and highest air temperatures around 25°C dominated over Central Europe.

The O_3 mixing ratio measured at the Schneeberg station and the Lidar values at corresponding height show good agreement (Fig. 2). For 99 half-hourly data sets of synchronous measurements, the reduced major axis correlation (Miller et al. 1962) yields a linear correlation with slope 0.93 ± 0.10 ppb (average ± 1 σ) and intercept 14.7 ± 11.6 ppb. The square of the correlation coefficient is 0.86, which is not significantly different from the ideal value 1.0. Therefore, the data from the ground-based station and the Lidar are in agreement within the scatter of the data sets. The Lidar seems to measure slightly higher O_3 mixing ratios than the Schneeberg station, possibly because of the larger influence of dry deposition within air masses that reached the Schneeberg station, as compared to the free air masses sampled with the Lidar remote sensing technique.

For the Waldstein and Weissenstadt sites, the comparison of Lidar and ground-based O_3 data shows larger differences (Fig. 2). These are probably due to the larger influence of the surface at the stations as previously explained. Simultaneously to the temperature decrease by radiative cooling, and the increase of relative humidity during the early nights (not shown), the O_3 mixing ratios at Weissenstadt station decreased rapidly, with a further reduction during the nights. This decrease cannot be explained by reaction of O_3 with freshly emitted NO_x, because only during the early morning hours slightly higher NO_x mixing ratios, resulting from street traffic, were measured. Nor were higher aerosol concentrations observed during the beginning of the night. Therefore, the O_3 decrease at Weissenstadt can only be explained by dry deposition or advection of air masses carrying less O_3. The Waldstein station also shows nightly decreases of O_3, however, these decreases are less sharp than those at Weissenstadt, so that the pattern at Waldstein is in between those of the other two stations (Schneeberg and Weissenstadt).

The wind directions at the three ground-based stations were in agreement with each other during the days with wind speeds above approximately 1.5 m s^{-1} (Fig. 3). Only during night-time conditions with low wind speed, the Weissenstadt station shows a westerly wind direction, independent of the wind direction of the other stations. This different wind regime is apparently caused by slope and mountain winds. At night, cool katabatic winds along the mountain slopes flow into the valley from the west. If no stronger mesoscale winds dominate the direction at the Weissenstadt station at night, the direction and speed of this mountain wind is recorded.

The vertical O_3 profiles during the night of 15./16. May show a slight increase with altitude within the lowest few hundred metres above ground (Fig. 4). Single standard deviations of the data within the profiles (Fig. 4) are approximately ± 5 ppb (not shown). The O_3 mixing ratios, as measured with the Lidar close to the ground, decrease from 62 ppb to 47 ppb throughout the night. (Note that the low O_3 mixing ratio, as measured by the ground station at the same site (Fig. 2), are not detected by the Lidar). At altitudes above approximately 250 m above ground, the O_3 profiles show an increase in the early morning (after 04:46 hrs). A corresponding increase is also recorded at the Waldstein station (Fig. 2). Apparently, air masses with higher concentrations of O_3 were advected at this time and altitude. This phenomenon is not strong enough to be deductible from the weather maps, but it is supported by the observation that the wind direction turned from east to west at that time at Waldstein and Schneeberg stations (Fig. 3). The new air masses, carrying about 80 ppb O_3, descend by about 100 m vertically within one hour (last 3 profiles in Fig. 4), which is confirmed by the observation that the turn of wind direction occurs earlier at the Schneeberg than at the Waldstein site.

4 Conclusions

At the Schneeberg, Waldstein, and Weissenstadt stations, the altitude a.s.l., as well as the influence of surface-atmosphere exchange processes, on the physical and chemical characteristics of air masses arriving at these stations increase, in this order. At the Schneeberg site, diurnal changes of concentrations of atmospheric trace gases are hardly observed. In contrast, the Waldstein site exhibits stronger diurnal changes, resulting from a high deposition of O_3, particularly during the nights (Klemm and Mangold 2001). The Weissenstadt site is characterised by its location in the bottom of the basin. Air masses transported by katabatic winds are subject to depletion of O_3 through dry deposition onto the surfaces along their trajectories. This results in the sudden and rapid decrease of O_3 mixing ratios associated with the mountain wind, directed downhill and towards the Weissenstadt basin. Compared to the moderate decrease of O_3 mixing ratios measured close to the ground by the Lidar-system, this leads to the conclusion that the Lidar could not detect the low O_3 mixing ratios in the layer influenced by the mountain winds,

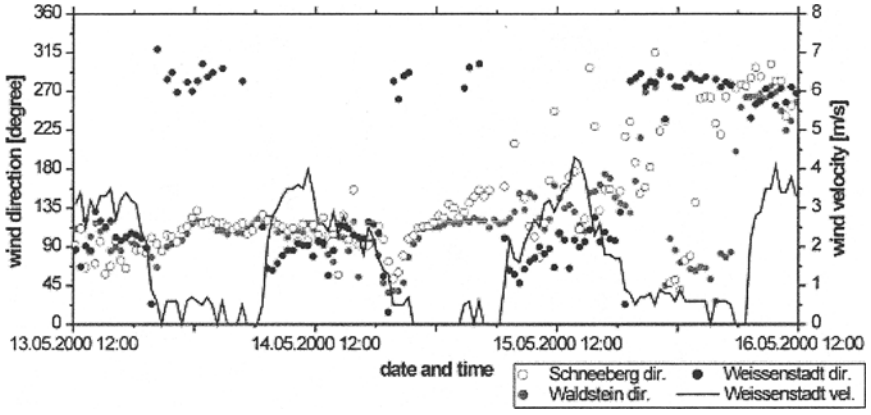

Fig. 3: Wind directions at the ground stations and wind velocity at the Weissenstadt station from May 13 to May 16, 2000

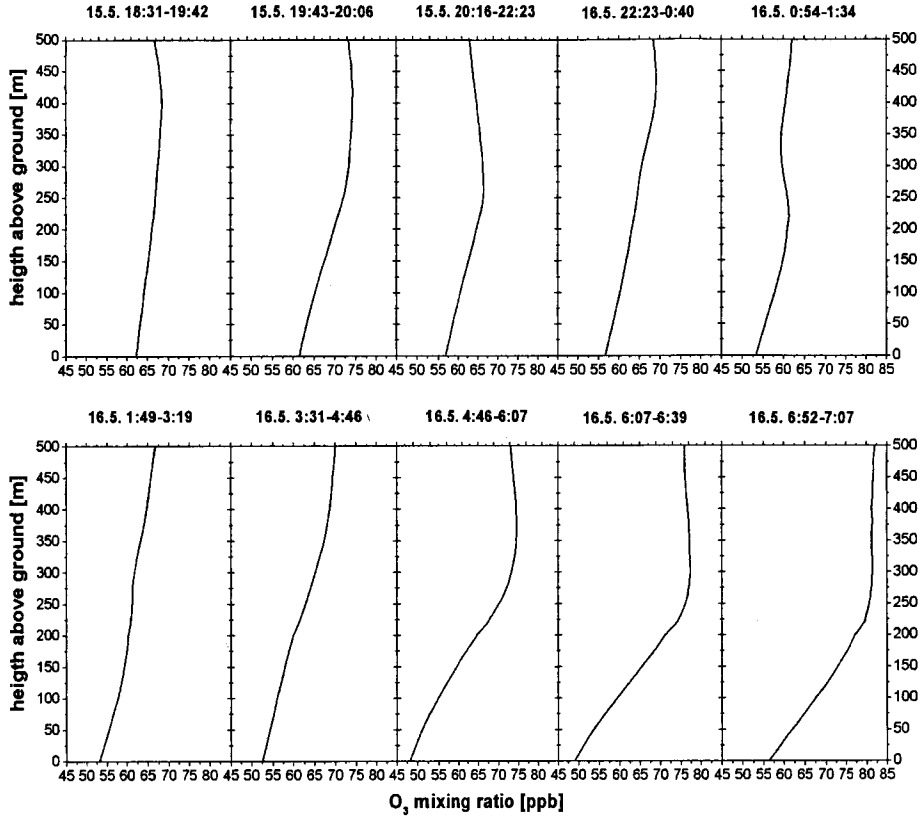

Fig. 4: Vertical profiles of O_3 mixing ratios during the night of May 15 to May 16, 2000. Profiles are shown for altitudes up to 500 m only for clarity of presentation

because the Lidar starts measuring at altitudes above ground of approximately 5 m, and the calculation of averaged mixing ratios starts at about 20 m. This limits the applicability of the Lidar techniques in near-surface air masses, particularly in complex terrain.

At higher altitudes above ground, the Lidar technique proved to be very useful and lead us to understand the processes relevant for the ozone budget within the boundary layer. The advection of air masses containing higher mixing ratios of O_3 in the morning hours of 16 May 2000, could be clearly detected and quantified (Fig. 4). The quantification of turbulent transport within the lower few hundreds of metres of the boundary layer is critical to develop our understanding of the dynamics of O_3 in complex terrain.

At this point, our understanding is still semi-quantitative in nature. Further experiments employing the quantification of O_3 deposition onto the forests and agricultural patches with micrometeorological techniques, are needed to quantitatively describe the dynamics of O_3 in such mountain valleys. In our opinion, the quantification of the relative roles of deposition and advection in the remote continental atmospheric boundary layer is crucial for the appropriate parameterisation of mesoscale atmospheric chemistry-transport models.

Acknowledgements. This study was supported by the German Federal Ministry for Research and Education (BMBF) through grant No. BEO 51-0339476C. We are indebted to the Bavarian Landesamt für Umweltschutz (LfU) for supporting our experiment with their mobile air chemistry laboratory, and to J. Gerchau and W. Eugster for help during the field phase and data evaluation. Thanks are due to Marlin Schmidt for language-editing of the manuscript.

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Received: July 25th, 2001
Accepted: December 11th, 2001
OnlineFirst: December 17th, 2001