Lidar Observations of the Vertical Aerosol Flux in the Planetary Boundary Layer

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ABSTRACT

The vertical aerosol transport in the planetary boundary layer (PBL) is investigated with lidars. Profiles of the vertical wind velocity are measured with a 2-μm Doppler wind lidar. Aerosol parameters are derived from observations with an aerosol Raman lidar. Both instruments were operated next to each other at the Institute for Tropospheric Research (IfT) in Leipzig, Germany. The eddy correlation technique is applied to calculate turbulent particle mass fluxes on the basis of aerosol backscatter and vertical wind data obtained with a resolution of 75 m and 5 s throughout the PBL. A conversion of particle backscatter to particle mass is performed by applying the IfT inversion scheme to three-wavelength Raman lidar observations. The method, so far, is restricted to stationary and dry atmospheric conditions under which hygroscopic particle growth can be neglected. In a case study, particle mass fluxes of 0.5–2.5 μg m⁻² s⁻¹ were found in the upper part of a convective PBL on 12 September 2006.

1. Introduction

The vertical exchange of sensible heat (temperature), latent heat (moisture), particles, and trace gases between the surface and the lower troposphere has a strong influence on weather and climate and atmospheric composition, as well as on smog and haze conditions at the ground. Vertical exchange depends in a complicated way on surface characteristics and meteorological conditions in the planetary boundary layer (PBL). The mechanisms within the PBL and in the entrainment zone (the transition layer between PBL and the free troposphere) are not well understood and thus are not well parameterized in atmospheric models. Observations of fluxes covering the entire PBL and the entrainment zone are rare.

With respect to aerosols, vertical transport is even more complicated because the ascent of particles is often combined with water uptake because of a relative humidity increase with height in the PBL. The particle mass concentration and optical and microphysical properties thus change with height during lifting processes, even if the boundary layer is well mixed. Because of the hysteresis effect, the hygroscopic growth factors can be different for updrafts and downdrafts. Entrainment of free-tropospheric dry air is another factor that influences the growth and shrinking of particles and thus the particle mass concentration. At the top of the PBL cloud droplets can form on the swollen aerosol particles. The cloud droplet size distribution sensitively depends on the amount of available aerosol particles, their chemical composition, and the aerosol flux into the cloud (Khain et al. 2000). Subsequent cloud dissolution in downdrafts will affect the particle size distribution, the volume and mass concentration, and the related optical effects too. All of these processes are far from being well understood because adequate observations are missing. This circumstance makes the description of aerosols in atmospheric models very difficult. As a consequence, the uncertainty in the estimation of the vertical aerosol exchange and the related direct and indirect aerosol effects remains rather high unless these aerosol-related processes are studied in detail based on measurements.

Turbulent aerosol fluxes, so far, have been investigated with in situ techniques only. Typically, an ultrasonic anemometer is applied together with a fast particle counter, and fluxes of particle number concentration are derived at a single point, preferably on a tower,
in the surface layer below 100-m height (e.g., Buzorius et al. 1998). Recently, this technique has also been applied on an aircraft near the ocean surface (Buzorius et al. 2006). In situ measurements near the ground are representative for relatively small areas and can be used for describing local fluxes from very specific sources, for example, a forest, a field, or an urban site.

In contrast, flux profile observations throughout the PBL are representative of a much larger scale and can be used for the improvement of flux parameterizations in mesoscale and general circulation models. Such observations can only be done with remote sensing instruments that provide the parameters of interest with high accuracy (<5%-10%) and with a spatial resolution of the order of 50 m and a temporal resolution of a few seconds.

Remote measurements of turbulent fluxes in the planetary boundary layer were first shown by Senff et al. (1994). A water vapor differential absorption lidar (DIAL) was combined with a radar radio acoustic sounding system (RASS) to simultaneously obtain the fluctuations of the water vapor density and vertical wind velocity, from which the turbulent water vapor flux is derived by applying the eddy correlation technique. Later, the method was used to study ozone budget terms that provide the parameters of interest with high accuracy (<5%-10%) and with a spatial resolution of the order of 50 m and a temporal resolution of a few seconds.

Kiemle et al. (2007). With the Doppler wind lidar technique one of the most limiting factors in the observation of vertical exchange phenomena, that is, the lack of highly accurate measurements of the profile of the vertical wind component, could be overcome. Only with lidar can profiles of the vertical wind speed be measured with an accuracy of <10 cm s\(^{-1}\) and the required high resolution up to the top of the entrainment zone, that is, up to 3–4-km height in summer.

Lidar also allows the observation of aerosol scattering properties throughout the PBL with adequate spatial and temporal resolution. However, for the investigation of aerosol fluxes, next to optical aerosol properties, which control the radiative impact of the aerosol particles and possible feedbacks on the wind field, microphysical parameters such as particle number and mass concentrations are of primary interest. The application of the multiwavelength Raman lidar technique seems promising in this context. This technique permits an accurate determination of particle extinction and backscatter coefficients at several wavelengths. Particle surface area, volume, and mass concentrations can be derived from the multiwavelength backscatter and extinction data with an inversion scheme (e.g., Müller et al. 2001). Even if microphysical particle parameters cannot be determined with the high resolution required for flux investigations, we believe that combined information from highly resolved vertical wind and multiwavelength backscatter data together with the mean aerosol properties obtained with the inversion scheme will lead to a significant increase in the understanding of the turbulent aerosol transport in the PBL.

In 2003, we made a first successful feasibility study and performed combined measurements with the three-wavelength Raman lidar of the Institute for Tropospheric Research (IfT) and the Doppler wind lidar of the Max Planck Institute for Meteorology (Wandinger et al. 2004). Since the new Doppler wind lidar of IfT was completed in 2005, this instrument has been used in conjunction either with the IfT six-wavelength lidar (Althausen et al. 2000) in field campaigns or with the IfT three-wavelength Raman lidar (Mattis et al. 2002) for long-term observations of the PBL development under different meteorological conditions in flat terrain at the IfT site in Leipzig, Germany. In this paper, we present instrumentation, methodology, and first results of these studies. In section 2 the new 2-µm Doppler wind lidar of IfT is described in more detail. The methodology of determining vertical aerosol fluxes from the combined vertical wind and aerosol observations is discussed in section 3. Results based on a case study are presented in section 4. We begin with the simplest case of a dry, cloud-free convective boundary layer observed on 12 September 2006. The relative humidity was below 50% at the top of the PBL, so that hygroscopic growth did not play any role. Concluding remarks and an outlook are given in section 5.

2. Instrumentation

For the measurements presented in this paper IfT’s coherent Doppler lidar was combined with the institute’s three-wavelength Raman lidar. The Raman lidar applies a Nd:YAG laser and emits radiation at 355, 532, and 1064 nm. In addition to the three elastic backscatter signals (at 532 nm with polarization discrimination), vibration–rotation Raman signals of nitrogen at 387 and 607 nm and of water vapor at 408 nm, and two rotational Raman signals at 532 nm are observed. From these nine return signals, profiles of particle backscatter coefficients at 355, 532, and 1064 nm; particle extinction coefficients at 355 and 532 nm; the particle depolarization ratio; the water vapor mixing ratio; and the temperature are determined. Details of the Raman lidar setup and the data evaluation schemes have been pre-
sented elsewhere (e.g., Mattis et al. 2002; Ansmann and Müller 2005). In recent years, several measurement capabilities have been added to the system. Especially useful for the studies shown here are a 0.1-m near-range telescope, which can be used alternately to the 1-m far-range telescope for observations between 0.1- and 1.5-km height, and an analog detection channel at 532 nm, which runs parallel to the nine photon-counting detection channels and provides aerosol backscatter data with very high spatial and temporal resolution of 7.5 m and 5 s, respectively.

The optical setup of the coherent Doppler wind lidar is shown in Fig. 1 (Žeromskis et al. 2003); it is realized on two breadboards within one mechanical frame. On the lower level the laser system is assembled. It consists of a continuous-wave (CW) laser (the master oscillator) and a pulsed laser (the power oscillator). On the upper level the transceiving and mixing optics are arranged.

The master oscillator is a CW single-frequency near-hemispherical laser. The active medium is a LuAG crystal of 3-mm length doped with 3% Tm. One end of the crystal has a high-reflection coating for 2022 nm and serves as a resonator mirror; the other end is antireflection coated. The crystal is pumped from one end with 785-nm radiation of a fiber-coupled CW laser diode. The diode delivers a maximum power of 2.5 W. Single-mode operation is achieved with two etalons—an uncoated one of 90-μm thickness and a coated one of 250-μm thickness and a reflectivity of 30%. By tilting the etalons, laser operation at either 2021.8 or 2022.55 nm is possible. These wavelengths correspond to minima of the atmospheric absorption spectrum in the spectral range of laser operation. The Brewster plate maintains linearly polarized laser emission. With an outcoupling mirror of 99% reflectivity and a curvature radius of 10 cm, a laser power of 25 mW in single-mode operation is achieved. The master oscillator is thermally isolated, and the temperature within the housing is stabilized to better than 0.1 K. A single-longitudinal mode drift of about 300 MHz h⁻¹ was observed during long-term measurements with a scanning Fabry–Perot interferometer.

A lens behind the master oscillator is used to collimate the laser beam to a divergence of about 0.7 mrad.
The output radiation of the master oscillator is divided into two parts—one for injection seeding of the power oscillator and the other for heterodyne detection. Back reflection from the power oscillator to the master oscillator is prevented by the use of two optical isolators.

For the power oscillator we chose a design similar to the high-resolution Doppler lidar (HRDL) system (Grund et al. 2001; Wulfmeyer et al. 2000). The active medium is again a LuAG crystal doped with 3% Tm; the crystal length is 12 mm. The laser is L shaped to allow longitudinal pumping of the crystal from both sides with two CW fiber-coupled laser diodes. Each diode delivers a maximum output power of 30 W. Two quarter-wave plates near the crystal are inserted to prevent spatial hole burning. A Q-switched pulse operation is obtained with an 80-MHz acousto-optical modulator, which is also used for injection seeding and frequency-offset generation. Usually, the laser runs with a pulse repetition rate of 750 Hz at which a pulse energy of 1.5 mJ is achieved. Longitudinal cavity stabilization to ±1 MHz is based on the Pound–Drewer–Hall method (Wulfmeyer et al. 2000).

In the transceiving optics a Glan–Taylor polarizer and a quarter-wave plate are inserted in order to use the off-axis Mersenne telescope for expanding and transmitting the laser beam as well as for receiving of atmospheric backscattered radiation. The received signal is directed to an InGaAs detector and optically heterodyned with the reference radiation from the master oscillator (Engelmann 2003).

The data-processing line starts with a low-noise transimpedance amplifier behind the photodetector. The amplified signal and a pulse monitor signal are connected to a high-frequency switch to toggle both signals. In this way, the monitor signal is switched into the signal line during pulsing time. Data acquisition is performed with an 8-bit digitizer PC card, the sampling rate of which is set to 250 mega-samples per second. Real-time processing software on the double 2.8-GHz processor computer splits the signal into approximately 200 half-overlapping height bins of 1 μs each. After applying a Blackman–Harris (−74 dB) window, the signals are transformed to the frequency domain by a 256-point fast Fourier transform (FFT). We end up with a spatial resolution of 75 m from almost independent range gates because of the laser pulse length of 450 ns and the windowing of the signals. The resulting power spectra are shifted to correct for the monitor offset frequency and are finally averaged for a fixed time interval. It was found that 3-s temporal averaging is sufficient to obtain well-analyzable spectra. For later processing purposes the spectra are saved into a NetCDF file format. Afterward, spectral peak finding, and hence wind speed determination, is performed by a center-of-mass approach (H. Linné 2005, personal communication; Rhone 2004).

The main parameters of the Doppler wind lidar are summarized in Table 1. The system is set up in a container. A fast hemispherical scanner is mounted on the roof and used for three-dimensional wind measurements. Conical scans are usually performed to determine the horizontal wind vector.

### Table 1. Specifications of the Doppler wind lidar.

<table>
<thead>
<tr>
<th>Master oscillator</th>
<th>Power oscillator</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laser crystal</td>
<td>Tm:LuAG</td>
</tr>
<tr>
<td>Design</td>
<td>Near hemispherical</td>
</tr>
<tr>
<td>Wavelength</td>
<td>2022.5 nm</td>
</tr>
<tr>
<td>Power</td>
<td>25 mW</td>
</tr>
<tr>
<td>Laser crystal</td>
<td>Tm:LuAG</td>
</tr>
<tr>
<td>Design</td>
<td>L shaped</td>
</tr>
<tr>
<td>Pulse energy</td>
<td>1.5 mJ</td>
</tr>
<tr>
<td>Pulse repetition rate</td>
<td>750 Hz</td>
</tr>
<tr>
<td>Frequency offset</td>
<td>80 MHz</td>
</tr>
<tr>
<td>Frequency stability</td>
<td>±1 MHz</td>
</tr>
<tr>
<td>Pulse duration</td>
<td>450 ns</td>
</tr>
<tr>
<td>Chirp</td>
<td>0.95 MHz μs⁻¹</td>
</tr>
<tr>
<td>Type</td>
<td>Off-axis Mersenne</td>
</tr>
<tr>
<td>Free aperture</td>
<td>140 mm</td>
</tr>
<tr>
<td>Photodetector</td>
<td>InGaAs, 75-μm diameter</td>
</tr>
<tr>
<td>Data acquisition</td>
<td>Preamplifier</td>
</tr>
<tr>
<td></td>
<td>DC, 200 MHz, 20 kV A⁻¹</td>
</tr>
<tr>
<td></td>
<td>Digitizer</td>
</tr>
<tr>
<td></td>
<td>PCI, 8 bit, 250 MS s⁻¹</td>
</tr>
<tr>
<td></td>
<td>Computer</td>
</tr>
<tr>
<td></td>
<td>Two 2.8-GHz processors</td>
</tr>
<tr>
<td></td>
<td>Data processing</td>
</tr>
<tr>
<td></td>
<td>15 km at 1 kHz, 75-m resolution</td>
</tr>
</tbody>
</table>
limited by the Doppler wind lidar. At lower heights the outgoing pulse disturbs the signal detection. Above the PBL, the aerosol content usually is too low to produce a sufficiently strong backscatter signal for coherent detection.

For the calculation of turbulence parameters, we investigate the highly resolved time series of vertical wind speed and aerosol scattering data at specific height levels. For ground-based profile observations, Taylor’s frozen turbulence hypothesis is applied; that is, it is assumed that the temporally resolved datasets well represent the spatial ensemble average. Then, the turbulent flux \( F_b \) of an atmospheric parameter \( b \) is given by the covariance between \( b \) and the vertical wind speed \( w \):

\[
F_b = \overline{b w'},
\]

where the prime indicates deviations from the mean value and the overbar represents the temporal average (Stull 1997). By definition, updrafts have a positive sign, and positive values of \( F_b \) imply upward fluxes. In principle, the application of Taylor’s hypothesis requires an infinite length of the time series. Because, in reality, only measurements over limited time intervals are possible, the sampling error must always be taken into account in the interpretation of flux observations.

Usually, the parameter \( b \) represents a scalar species density or mixing ratio. In case of aerosols, the turbulent transport in terms of particle number, volume, or mass is of interest. However, from lidar observations we can only determine particle backscattering with sufficient temporal resolution for turbulence investigations. Therefore, we use the time series of the 532-nm analog signal from the Raman lidar and calculate the 532-nm particle backscatter coefficient \( \beta \) with a resolution of 5 s with the Klett method (Fernald 1984; Ansmann and Müller 2005). From the Raman lidar observations taken with a resolution of 30–60 min, preferably under low background light conditions after sunset, we get the information on the actual lidar ratio. Thus, trustworthy information on this input value for the Klett method is available, and the backscatter coefficient is determined with an accuracy of 5%–8%. The linear trend is removed from the time series of backscatter coefficient and vertical wind, and the covariance is calculated. Thus, the primary output of the flux calculation after Eq. (1) is the covariance \( \overline{b w'} \).

In a second step, we use the multiwavelength Raman lidar observation (extinction coefficients at 355 and 532 nm; backscatter coefficients at 355, 532, 1064 nm) and retrieve microphysical particle parameters with our inversion scheme (Müller et al. 2001; Ansmann and Müller 2005). In this way, among other quantities, we obtain profiles of the particle volume concentration with low temporal resolution from which we can estimate the mean particle mass concentration \( m \) by assuming a typical particle density.

In the third step, we assume that the obtained relationship between \( \beta \) and \( m \) holds for the entire measurement region (middle and upper PBL), so that any change of \( \beta \) with height and any fluctuations \( \beta' \) are caused by respective changes in the mass concentration \( m \) and its fluctuations \( m' \). We postulate

\[
\frac{\beta'}{\beta} = \frac{m'}{m}
\]

throughout the convective boundary layer and estimate the turbulent aerosol mass flux at a given height to

\[
\overline{m w'} = (\overline{m/\beta})\beta\overline{w'}.
\]

The basic assumption of our approach here is that the conversion factor \( \overline{m/\beta} \) is temporally and vertically constant. This, in turn, is true when the aerosol size distribution and the particle chemical composition do not vary with time and height in the convective PBL.

By plotting the Ångstrom exponent \( \ln[\beta(\lambda_1)/\beta(\lambda_2)]/\ln(\lambda_1/\lambda_2) \), with the observation wavelengths \( \lambda_1, \lambda_2 = 355, 532 \) nm or \( \lambda_1, \lambda_2 = 532, 1064 \) nm, as a function of height and time, we can test our basic assumption. If the Ångstrom exponent is invariant, it indicates temporally and vertically constant aerosol properties (size distribution and chemical composition) and thus a temporally and spatially constant \( \overline{m/\beta} \) ratio (O’Neill et al. 2001a,b).


a. Measurements

The data for our investigations were taken at IfT on 12 September 2006. The institute is located in the eastern suburbs of Leipzig. The city is surrounded by flat terrain. The Raman lidar and the Doppler wind lidar were used to measure coherent datasets of aerosol backscatter and corresponding vertical wind speed during daytime. The measurement with the Raman lidar was extended until nighttime to obtain accurate data for a characterization of microphysical aerosol properties. Figure 2 presents the PBL development during daytime between 0930 and 1730 UTC (1030 and 1830 LT, central European time). The range-corrected signal of the 532-nm analog channel in the Raman lidar and the vertical wind speed show very good correlations. Thermal updraft regions (yellow and red) can be clearly distinguished from downward mixing processes (dark green).
Figure 3 shows profiles of temperature, potential temperature, relative humidity, and water vapor mixing ratio obtained with radiosondes launched at the IfT site around 0700 and 1400 UTC. The stable layering in the morning evolved to well-mixed conditions up to 1100 m at 1400–1600 UTC. The relative humidity was less than 40% throughout the entire PBL in the afternoon. Therefore, hygroscopic particle growth can be neglected in further investigations. The meteorological conditions were remarkably stationary from 11 to 13 September because of the influence of a strong high pressure system. Minimum temperatures of 8°C (±0.2°C) in the morning and maximum temperatures of 26°C (±0.2°C) in the afternoon were observed on each of the 3 days. Southeasterly winds of 5–6 m s⁻¹ dominated in the PBL. Similar developments of the PBL, as shown in Fig. 2, were found on all 3 days.

From the evening measurements taken on 12 September 2006, we derived profiles of particle backscatter coefficients at three wavelengths and particle extinction coefficients at two wavelengths with the Raman method. The data for the time period from 1900 to 2000 UTC are shown in Fig. 4. The resulting lidar ratios of 73 ± 5 sr for 355 and 532 nm indicate relatively small, absorbing particles, which are typical for anthropogenic pollution from eastern Europe. The data at 670-m height were used as input for the inversion scheme to determine microphysical particle properties. It is assumed that the aerosol properties in this height level were representative for the ensemble of particles obtained in the PBL during the whole day. In addition, systematic retrieval errors resulting, for example, from the incomplete overlap of laser beam and receiver field of view, can be neglected at this height level.

The results of the lidar data inversion are shown in Fig. 5. The derived particle volume distribution agrees very well with data obtained from regular in situ measurements with a differential mobility particle sizer (DMPS) at IfT. The effective, that is, the area-weighted mean, particle radius retrieved from the lidar data of
160 ± 37 nm compares well to the value of 135 nm from the in situ data on the ground. The somewhat lower value obtained with the DMPS can be explained by the cutoff particle diameter of 800 nm. In addition, the lidar data inversion is insensitive to particles smaller than about 100 nm. A particle volume concentration of 19 ± 8 μm³/cm³ is found from the lidar data.

Profiles of the backscatter-related Ångström exponents for four time intervals are presented in Fig. 6. The long-wavelength exponent shows an almost constant value of 0.9 for all heights and all time intervals. The short-wavelength Ångström exponent is noisier, but remains almost constant at values around 1.4 as well. This behavior of the Ångström exponents, which are rather sensitive to any small changes in the aerosol characteristics, again indicates very stationary aerosol properties and the absence of hygroscopic growth during the entire day.

Fig. 3. Temperature ($T$), potential temperature ($\Theta$), relative humidity (RH), and water vapor mixing ratio (MR) obtained with collocated radiosondes launched at 0700 and 1400 UTC.

Fig. 4. Temperature ($T$), potential temperature ($\Theta$), relative humidity (RH), and water vapor mixing ratio (MR) obtained with collocated radiosondes launched at 0700 and 1400 UTC.

Fig. 5. Aerosol volume distributions derived from lidar data inversion and from in situ measurements with DMPS.

Fig. 6. Backscatter-related Ångström exponents for the time periods 1300–1430, 1430–1600, 1600–1730, and 1900–2000 UTC for two wavelength intervals.
Figure 7 shows the time series of the aerosol optical depth (AOD) in the PBL at 532 and 1064 nm. The AOD was calculated from the height-integrated backscatter coefficient profiles with an extinction-to-backscatter ratio of 75 sr (cf. Fig. 4). The backscatter coefficients were assumed to be constant in the region of incomplete overlap between the laser beam and receiver field of view below 300 m where no trustworthy data can be obtained. This assumption may lead to a bias in the AOD in the morning and evening hours when the aerosol is not well mixed, even though only a slight variation around the mean PBL AOD values of 0.11 and 0.06 at 532 and 1064 nm, respectively, was found over the whole day.

In summary, it can be stated that information obtained from radiosoundings, lidar measurements, and in situ observations clearly indicates stationary aerosol conditions and low relative humidity throughout the PBL during the entire measurement period on 12 September 2006. There is no indication for a change of air mass, aerosol composition, and size distribution, and no indication for particle growth because of water uptake. Therefore, temporal and spatial variations of the particle backscatter coefficient can explicitly be attributed to turbulent mixing and vertical transport. The eddy correlation method is applicable without restrictions, and particle mass fluxes can be calculated after Eqs. (1)–(3). A conversion factor \( \frac{m}{H} \) (532 nm) of \( 21.1 \pm 9 \mu g \: m^{-3} \: Mm \: sr \) is derived for this specific day. It results from the volume concentration of \( 19 \pm 8 \mu m^3/cm^3 \) obtained for \( \beta(532 \: nm) = 1.44 \: Mm^{-1} \: sr^{-1} \) (cf. Fig. 4) and a particle density of 1.6 g cm\(^{-3}\), which is typical for continental, ammonium sulfate–like particles with a volume water content of 20% (Van Dingenen et al. 2004).

Figure 8 presents the variance spectra of the time series of the backscatter coefficient and vertical wind speed for three height levels in the time interval from 1430 to 1600 UTC. The spectra were smoothed by interpolating the data to a logarithmic equidistant scale for frequencies \( f > 1.1 \times 10^{-5} \: Hz \). The total variance of the backscatter coefficient increases with height, whereas the variance of the vertical wind speed decreases. The spectra obviously cover the energy-containing part of the spectrum and at least some part of the inertial subrange (\( f^{-5/3} \) rolloff). Noise on the time series, which gives a constant contribution to the spectra at high frequencies, is found for the backscatter coefficient only. Assuming a mean horizontal wind velocity of 5 m s\(^{-1}\), we can estimate that eddy sizes of more than 50 m are covered by the measurement.

By calculating the cospectrum of vertical wind and
particle backscatter fluctuations, we can find out which frequencies mainly contribute to the turbulent flux (Stull 1997). Figure 9 shows the cospectrum for the measurement height of 700 m and the time interval from 1130 to 1600 UTC. The presentation of $f_C$ versus $\log(f)$ preserves the area and gives realistic weight to high frequencies. The cospectrum indicates contributions to the vertical flux in the frequency range from $2 \times 10^{-2}$ to $3 \times 10^{-4}$ Hz. With a horizontal wind velocity of $5 \text{ m s}^{-1}$ this range corresponds to horizontal sizes of $250 \text{ m} - 15 \text{ km}$. The maximum occurs at $f = 3.15 \times 10^{-3}$ Hz, corresponding to an eddy size of 1.6 km. This value agrees very well with the dominant frequency of $(2\pi IS)^{-1} = 3.12 \times 10^{-3}$ Hz predicted from the horizontal flux integral scale (IS) of 51 s (Lenschow et al. 1994). The flux integral scale was estimated from the vertical wind and backscatter integral scales with Eq. (6), given by Giez et al. (1999). The vertical wind and backscatter integral scales were 51 and 55 s, respectively, at 700-m height and increased to values of 60 and 90 s, respectively, in the height region of 900–1000 m. The fact that no significant contributions are found in the cospectrum at frequencies higher than $2 \times 10^{-2}$ and lower than $3 \times 10^{-4}$ Hz supports the strategy of taking measurements with a resolution of $5 - 10 \text{ s}$ and a minimum length of the time series of 60–90 min for the flux computation. The latter values are also in agreement with the findings of Lenschow et al. (1994) who showed that systematic errors resulting from “flux losses” are <5% if the length of the measurement period is $\approx 100 \text{ IS}$, that is, 80–100 min in our case. Therefore, we chose time series of 90-min length for the investigations below.

Our findings on the relevant time scales and eddy sizes are in good agreement with the results from ground-based remote sensing measurements of water vapor (Senff et al. 1994; Giez et al. 1999) and ozone fluxes (Senff et al. 1996). Horizontal integral scales of the vertical wind speed between 50 and 200 s were reported for different measurement cases by Giez et al. (1999). Dominant eddy sizes for the upward transport of water vapor of 1–2.5 km (Senff et al. 1994; Giez et al. 1999) and for the downward mixing of ozone of approximately 1.5–3 km (time scale of 10 min; horizontal wind speed is not given in the paper) were obtained.

Figure 10 shows profiles of the aerosol mass flux for three selected time periods (cf. Fig. 2). The conversion factor of $21.1 \mu g \text{ m}^{-3} \text{ Mm} \text{ sr}^{-1}$ has been applied to convert the measured values of $\overline{\beta w}$ (upper axis) to the aerosol mass flux $\overline{m w}$. The linear flux profile observed from 1430 to 1600 UTC is consistent with a source-/sink-free PBL and either no or constant advection of aerosol with height. The flux profile is then completely defined by the surface flux and the entrainment flux. During the PBL development (for the time period of 1300–1430 UTC), the strongest turbulent flux is observed in the entrainment region where the ascending PBL mixes with the cleaner air from the residual layer.
Above that region, the flux decreases to zero. In the time period from 1600 to 1730 UTC, the turbulent aerosol flux profile is close to zero throughout the PBL because the atmospheric convection slowed down in the late afternoon.

The obtained values of the aerosol mass flux of 0.5–2 μg m⁻² s⁻¹ in the active upper PBL are reasonable. We can prove that with a simplified budget estimate, which may give us an idea on the magnitude of the entrainment flux. From Fig. 2 it can be seen that the backscatter coefficient between 800 and 1000 m increased from values close to 0 in the morning to values around 1.8 Mm⁻¹ s⁻¹ in the afternoon. With our $\overline{m/\beta}$ conversion factor this corresponds to an increase of the mass concentration of 38 μg m⁻³. We assume that this change is only caused by convective mixing of particle-rich air from the lower PBL with clean air from the free troposphere, where the aerosol flux is zero. Then, a mass flux in the entrainment zone of 2 μg m⁻² s⁻¹ would fill an atmospheric column of 200-m depth with the respective amount of particles within 1 h. This corresponds very well with the observed PBL growth rate of about 150–200 m h⁻¹ before 1300 UTC. Later (between 1430 and 1600 UTC) the mass flux in the upper PBL was of the order of 0.5–1 μg m⁻² s⁻¹, which again is consistent with the observed increase of the PBL height of 50–75 m h⁻¹ after 1400 UTC (cf. Fig. 2). As mentioned, such a budget determination is a very simplified approach and is not valid in general; it neglects the sources and sinks of particles in the PBL as well as horizontal and vertical advection. However, because of the lack of validation methods, it is used as a consistency check here.

**b. Error analysis**

In Table 2 the relative errors of the aerosol mass flux for three height levels for the time period of 1430–1600 UTC are summarized. The dominating error of the determined aerosol mass flux is the sampling (random) error $\sigma_{F,s}$ (Lenschow et al. 1994). The experimental data lack statistical significance because a finite number of samples over a finite time period is taken instead of an ensemble average. The large sampling error is a general problem of all surface-based measurements of flux profiles, because the time series cannot be extended arbitrarily in a diurnally varying PBL. The sampling error is calculated after Eq. (5) from Giez et al. (1999). It is proportional to the horizontal flux integral scale divided by the sampling time. As mentioned above, the flux integral scale is estimated from the vertical wind and backscatter integral scales with Eq. (6), given by Giez et al. (1999). The systematic deviation from the ensemble mean, which results from the finite length of the time series, is found to be of the order of 2% [after Eq. (27) in Lenschow et al. (1994)] and can therefore be neglected.

The flux error due to instrumental noise $\sigma_{F,i}$ is calculated after Eq. (4) from Giez et al. (1999). In our case, this error is small compared to the sampling error. The instrumental noise of the backscatter and vertical wind measurements is estimated from the noise level of the power spectral density functions in Fig. 8 to $\sigma_{F,i} = 0.06$ Mm⁻¹ s⁻¹ and $\sigma_{w,i} < 0.04$ m s⁻¹, respectively.

Errors due to possible incoherence of the datasets in horizontal and vertical direction $\sigma_{F,h}$ and $\sigma_{F,v}$ respectively, were estimated by shifting the datasets by one bin (5 s and 75 m) in either direction, followed by a recalculation of the flux profile and a comparison to the original one. The maximum discrepancy found in this way was 30%. However, we assume that the respective errors are much smaller because the measurements were synchronized in time and the coherence of the height profiles has been checked on several occasions, for example, in the presence of sharp gradients in the signals at cloud bases. The error of the conversion factor from backscatter coefficient to aerosol volume and mass concentrations $\sigma_{F,v}$ follows from the uncertainties of the inversion procedure and was found to be 43% in this specific case (Ansmann and Müller 2005).

## 5. Summary and outlook

We proposed a concept for the determination of vertical aerosol fluxes in the PBL by combined measurements with a Doppler wind and a three-wavelength aerosol Raman lidar. Aerosol fluxes were determined from highly resolved vertical wind and aerosol backscatter data. The information on spectral extinction and backscatter coefficients was used to convert the optical aerosol data to particle microphysical properties and to estimate particle mass fluxes. In a case study, we investigated the development of a dry late-summer PBL in which the humidity growth of particles could be neglected. Aerosol mass fluxes of the order of 0.5–2 μg m⁻² s⁻¹ were found in the upper part of the active PBL in the early afternoon. These values are in accordance

<table>
<thead>
<tr>
<th>Height (m)</th>
<th>$\sigma_{F,s}$</th>
<th>$\sigma_{F,i}$</th>
<th>$\sigma_{F,h}$</th>
<th>$\sigma_{F,v}$</th>
<th>$\sigma_{F,e}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>487</td>
<td>88%</td>
<td>10%</td>
<td>&lt;11%</td>
<td>&lt;21%</td>
<td>43%</td>
</tr>
<tr>
<td>787</td>
<td>58%</td>
<td>4%</td>
<td>&lt;2%</td>
<td>&lt;23%</td>
<td>43%</td>
</tr>
<tr>
<td>1012</td>
<td>51%</td>
<td>2%</td>
<td>&lt;6%</td>
<td>&lt;30%</td>
<td>43%</td>
</tr>
</tbody>
</table>
with the measured increase of the PBL height and the observed values of the particle backscatter coefficient and the corresponding aerosol mass concentration within the upper PBL. Significant contributions to the turbulent transport of particles in the middle and upper PBL were found for time scales from 50 to 3000 s or from 250 m to 15 km (with a horizontal wind speed of 5 m s\(^{-1}\)). A dominant eddy size of about 1.6 km was observed, which is in agreement with the measured integral scales of vertical wind speed and particle backscatter coefficient of 50–55 s in the middle PBL.

Our future work will include the investigation of a variety of measurement cases under dry conditions. We will also examine situations with relative humidities >70% (hygroscopic growth of particles), and finally we will study aerosol flux characteristics in the cloudy PBL.

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**REFERENCES**


