JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 109, D24205, doi:10.1029/2004JD005142, 2004

Air mass modification over Europe: EARLINET aerosol observations from Wales to Belarus

Ulla Wandinger,[1] Ina Mattis,[1] Matthias Tesche,[1] Albert Ansmann,[1] Jens Bösenberg,[2] Anatoly Chaikovski,[3] Volker Freudenthaler,[4] Leonce Komguem,[5] Holger Linné,[2] Volker Matthias,[6] Jacques Pelon,[7] Laurent Sauvage,[7] Piotr Sobolewski,[8] Geraint Vaughan,[5] and Matthias Wiegner[4]

Received 18 June 2004; revised 15 September 2004; accepted 28 September 2004; published 21 December 2004.

[1] For the first time, the vertically resolved aerosol optical properties of western and central/eastern European haze are investigated as a function of air mass transport. Special emphasis is put on clean maritime air masses that cross the European continent from the west and become increasingly polluted on their way into the continent. The study is based on observations at seven lidar stations (Aberystwyth, Paris, Hamburg, Munich, Leipzig, Belsk, and Minsk) of the European Aerosol Research Lidar Network (EARLINET) and on backward trajectory analysis. For the first time, a lidar network monitored continent-scale haze air masses for several years (since 2000). Height profiles of the particle backscatter coefficient and the particle optical depth of the planetary boundary layer (PBL) at 355-nm wavelength are analyzed for the period from May 2000 to November 2002. From the observations at Aberystwyth, Wales, the aerosol reference profile for air entering Europe from pristine environments was determined. A mean 355-nm optical depth of 0.05 and a mean PBL height of 1.5 km was found for clean maritime summer conditions. The particle optical depth and PBL height increased with increasing distance from the North Atlantic. Mean summer PBL heights were 1.9–2.8 km at the continental sites of Leipzig, Belsk, and Minsk. Winter mean PBL heights were mostly between 0.7 and 1.3 km over the seven EARLINET sites. Summer mean 355-nm optical depths increased from 0.17 (Hamburg, northwesterly airflow from the North Sea) and 0.21 (Paris, westerly flow from the Atlantic) over 0.33 (Hamburg, westerly flow) and 0.35 (Leipzig, westerly flow) to 0.59 (Belsk, westerly flow), and decreased again to 0.37 (westerly flow) at Minsk. Winter mean optical depths were, on average, 10–30% lower than the respective summer values. PBL-mean extinction coefficients were of the order of 200 Mm^{-1} at 355 nm at Hamburg and Leipzig, Germany, and close to 600 Mm^{-1} at Belsk, Poland, in winter for westerly flows. Whereas the optical depth for westerly flows was typically <0.35 during the summer halfyear, it increased to values of 0.5–0.7 over most of the central European sites during easterly flows. Compared to aerosol sources in Poland and southeastern Europe, the highly industrialized and populated western European region was found to contribute only moderately to the European aerosol burden. Comparably clean conditions (low particle optical depth) prevailed at the Munich site, indicating a sensitive influence of the orography on haze conditions. Estimates of the mean effective (upward minus downward) aerosol mass flux into the atmosphere along the way from the Atlantic Ocean to central Europe (Leipzig), and from Leipzig to Belsk, that are consistent with the optical depth increase, yield values of 0.11–0.17 $\mu\mathrm{g}/(\mathrm{m}^2\mathrm{s})$ and 0.25 $\mu\mathrm{g}/(\mathrm{m}^2\mathrm{s})$, respectively. These values correspond to mean effective aerosol mass fluxes for 50 km × 50 km grid cells of the order of 10000 Mg/year and 20000 Mg/year, respectively. The estimates are in reasonable agreement with EMEP emission data. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric

---

[1] Institute for Tropospheric Research, Leipzig, Germany.
[2] Max Planck Institute for Meteorology, Hamburg, Germany.
[3] Institute of Physics, Academy of Sciences, Minsk, Belarus.
[4] Meteorological Institute, University of Munich, Munich, Germany.
[5] Physics Department, University of Wales, Aberystwyth, UK.
[6] GKSS Research Center, Geesthacht, Germany.
[7] Institute Pierre Simon Laplace, Palaiseau, France.
[8] Institute of Geophysics, Polish Academy of Sciences, Warsaw, Poland.

0148-0227/04/2004JD005142

Composition and Structure: Pollution—urban and regional (0305); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 3307 Meteorology and Atmospheric Dynamics: Boundary layer processes; *KEYWORDS:* lidar network, tropospheric aerosols, aerosol transport

Citation: Wandinger, U., et al. (2004), Air mass modification over Europe: EARLINET aerosol observations from Wales to Belarus, *J. Geophys. Res.*, *109*, D24205, doi:10.1029/2004JD005142.

1. Introduction

[2] The European Aerosol Research Lidar Network (EARLINET) is the first continent-scale lidar network for monitoring of anthropogenic haze [*Bösenberg et al.*, 2003]. The main objectives of EARLINET are the establishment of a comprehensive and quantitative statistical database of the horizontal and vertical distribution of aerosols on the European scale, and the use of these data for studies related to the impact of aerosols on a variety of environmental and climate-relevant processes. EARLINET provided the unique opportunity to quantify, for the first time, the changes in the aerosol optical properties of European air masses when clean air crossed the densely populated, highly industrialized continent with the prevailing westerly winds from the most western station at Aberystwyth, Wales, to the most eastern site at Minsk, Belarus. Here we present the essential results of this study. The obtained height-resolved, airflow-related, seasonally dependent aerosol data set provides detailed insight into the haze conditions over western and central/eastern Europe (north of the Alps). The lidar-network observations may thus provide reference scenarios for the validation of aerosol modules in atmospheric models that are used to quantify the impact of anthropogenic particles on the Earth's radiation budget (direct aerosol effect), environmental and cloud processes (indirect effect).

[3] Compared to alternative approaches of long-term aerosol monitoring, the advantage of lidar is obvious. Besides a precise vertical profiling of aerosols, lidars permit the detection of the height of the planetary boundary layer (PBL) and a clear separation of optical properties of PBL particles that originate from local and regional emissions of particles and gases and of free tropospheric particles which are often advected over long distances from other continents and are usually very different from the boundary layer particles regarding their optical and microphysical properties [*Wandinger et al.*, 2002; *Mattis et al.*, 2004]. Such a comprehensive characterization of aerosol optical properties is not possible with passive remote sensing. Ground-based (Sun photometry) and spaceborne remote sensing can be affected by clouds and do not allow the determination of the PBL optical properties in cases with dust and smoke layers in the free troposphere or stratospheric aerosols after major volcanic eruptions. The Earth's surface albedo has a strong impact on radiative fluxes measured with spaceborne radiometers. The spectral reflectance characteristics changes with time of the day (solar zenith angle) and season, and can vary strongly on a meter to hundred-meter scale over the continents [*Wendisch et al.*, 2004]. A rather accurate knowledge of the solar energy reflected by the surface at the measurement wavelength is required for a trustworthy retrieval of particle optical depth (at visible wavelengths) over land [*von Hoyningen-Huene et al.*, 2003]. The respective albedo corrections remain a very crucial task of spaceborne aerosol remote sensing over land.

[4] The paper is organized as follows. In section 2, the EARLINET measurement program and the methodology of the study are outlined. The results are presented in sections 3 and 4. Section 5 contains a short summary.

2. Methodology

[5] Coordinated lidar measurements began in May 2000. Three observations per week (Monday afternoon and evening, Thursday evening) were performed at 20 EARLINET stations [*Bösenberg et al.*, 2003] from May 2000–November 2002. Clean air arriving from maritime and polar regions was detected by the most northerly and westerly stations of Aberystwyth, Wales, and Linköping, Sweden. Traveling across Europe, these air masses are modified through anthropogenic activities, by which precursor gases and particles are emitted into the atmosphere. Depending on travel distance and residence time over the source regions, particle number concentrations, the physical and chemical state, and thus the optical properties of the aerosol change. The comparison of particle backscatter and extinction profiles measured at stations in central and eastern Europe with those at the boundaries of the network permits us to quantify the anthropogenic impact.

[6] We have restricted our investigations to the northern part of the network to avoid a considerable influence of orographic effects on the aerosol modification processes. The study is thus based on the routine, long-term measurements at seven EARLINET stations which are listed in Table 1. Eleven EARLINET stations located either within the Alps or south of the Alps and south of the Pyrenees were excluded from this study. EARLINET Raman lidar observations at northern and southern European stations are presented by *Matthias et al.* [2004b]. The Pyrenees and the Alps represent a natural orographic barrier across Europe. Measurements from two other stations, Linköping, Sweden, and Kühlungborn, Germany, were excluded because the lowest measurement height at these stations was about 1000 m above ground. The stations considered for the study deliver particle backscatter-coefficient profiles at 355 and/or 532 nm with a lowest measurement height of <500 m above ground so that the PBL is well covered by the measurements. A discussion on the retrieval of backscatter coefficients and on quality assurance activities can be found elsewhere [*Böckmann et al.*, 2004; *Matthias et al.*, 2004a; *Pappalardo et al.*, 2004].

[7] The measurements at each of the seven stations were investigated in the following way. Backscatter profiles were taken from the EARLINET database for five summer and winter halfyears of the EARLINET measurement period. These periods are the summer of 2000 (May–September), the winter of 2000–2001 (October 2000–March 2001), the summer of 2001 (April–September), the winter of 2001–2002 (October 2001–March 2002), and the summer of 2002 (April–September).

Table 1. EARLINET Sites (From West to East) and Measurement Parameters for the Investigation of Aerosol Modification Over Europe

	EARLINET Site	Altitude, m asl	Backscatter Wavelength, nm	Evaluation Method	Lowest Meas. Height, m Above Ground
AB	Aberystwyth (52.4°N, 4.1°W)	15	355	Raman	50–400
PL	Paris (48.6°N, 2.2°E)	156	532, 1064	Fernald	300–600
HH	Hamburg (53.6°N, 10.0°E)	25	355, 532, 1064	Fernald/Raman	200–300
MU	Munich (48.2°N, 11.6°E)	549	355, 532, 1064	Fernald	0–200
LE	Leipzig (51.4°N, 12.4°E)	90	355, 532, 1064	Raman	100–300
BE	Belsk (51.5°N, 20.5°E)	188	532, 694	Fernald	200
MI	Minsk (53.9°N, 27.4°E)	200	532, 694	Fernald	100

[8] Only evening measurements, i.e., measurements taken shortly after sunset were considered. In the evening, the optical properties of the fully developed daytime PBL (above the developing nocturnal PBL, i.e., above 250-m height) can still be observed. Each individual profile was inspected. Profiles which could bias the statistical approach were omitted. Especially, profiles which did not cover the entire aerosol layer because of the presence of clouds below 3-km height were excluded. The remaining profiles were extrapolated down to the ground by assuming a constant backscattering between the lowest measurement height and the surface. The uncertainty in the column-integrated backscatter introduced by this assumption (constant backscatter) was estimated to be of the order of 10% [*Bösenberg et al.*, 2003]. We restricted the study to the height range up to 3 km. Airflows and aerosol properties above 3 km (typically in the free troposphere) are usually very different from those in the PBL.

[9] The measurements were classified by means of analytical 96-h backward trajectories (1900 UTC arrival time). These trajectories were calculated from hourly wind fields of the global numerical weather prediction model of the German Weather Service [*Kottmeier and Fay*, 1998]. Daily backward trajectories for arrival heights corresponding to pressure levels of 975, 850, 700, 500, 300, and 200 hPa and arrival times of 1300 and 1900 UTC were available for all EARLINET sites for the entire time period from May 2000 to November 2002. Here only the lowest two pressure levels (975 and 850 hPa) were considered for the classification. These trajectories best describe the advection pattern in the PBL. Typical airflows representing, e.g., the transport of clean maritime air masses into the European continent, the air mass transport across western and central Europe, or the westerly transport of polluted air masses from eastern Europe were defined. The geographical conditions for each individual station were taken into account, e.g., by distinguishing flows from maritime and continental regions.

[10] In the next step, we averaged all backscatter profiles (of a season) which belong to a specific trajectory cluster. In addition to the air mass related, seasonal mean profiles we calculated respective summer and winter mean profiles from all summer (three summers) and winter profiles (two winters).

[11] From the mean backscatter-coefficient profiles, column-integrated backscatter values for the 0–3000-m height range (above ground) were calculated. These values were then converted to optical depths by applying typical lidar ratios (extinction-to-backscatter ratios) for the different airflow clusters. Lidar ratios were measured at 10 EARLINET stations [*Bösenberg et al.*, 2003]. From the seven sites used in this study, Aberystwyth and Hamburg deliver lidar ratios at 355 nm, whereas in Leipzig measurements at 355 and 532 nm are performed. Values of 50–60 sr are typical for polluted continental sites. The PBL mean values obtained at Leipzig are 58 ± 12 sr and 53 ± 11 sr for 355 and 532 nm, respectively [*Mattis et al.*, 2004]. At Hamburg an average value of 55 sr at 355 nm has been measured [*Bösenberg et al.*, 2003]. Nonabsorbing, maritime aerosols show lower lidar ratios. In very clean maritime air lidar ratios around 25 ± 5 sr were observed [*Ansmann et al.*, 2001; *Franke et al.*, 2001], also found from individual measurements at Aberystwyth. From all EARLINET measurements at Aberystwyth a mean value of 46 sr has been derived. Lidar ratios of 25 to 60 sr, depending on the investigated airflow, were therefore taken to convert column-integrated backscatter coefficients into optical depths. The relative uncertainty in the individual optical-depth values caused by the lidar-ratio estimates is assumed to be 20%–25% according to the standard deviations of the lidar ratios measured over Leipzig [*Mattis et al.*, 2004]. In the case of the air mass related mean or seasonal mean optical-depth values discussed below the relative uncertainty reduces to $\leq$10%.

[12] For stations which provide measurements at 532 nm only, the backscatter-coefficient profiles were converted to 355 nm by the use of Ångström exponents for backscattering. The Ångström exponent characterizes the spectral slope of an optical parameter and is defined in our case as $k = -\ln[\beta(\lambda_1)/\beta(\lambda_2)]/\ln(\lambda_1/\lambda_2)$, with the backscatter coefficient β and the wavelength λ. Figure 1 shows the mean Ångström-exponent profile (355–532-nm wavelength range) obtained from all EARLINET measurements at Leipzig as well as the mean profiles for summer and winter. Typical values of 1–2 with a mean value of 1.6 ± 0.3 were found. Significant differences between different airflows have not been observed. Ångström exponents of >1.5 usually indicate anthropogenic particles (pronounced accumulation mode with particle diameters <600 nm [*Remer and Kaufman*, 1998; *Remer et al.*, 1999; *Ansmann et al.*, 2002; *Mattis et al.*, 2004]), whereas smaller Ångström exponents (<1) are typical for large particles (sea salt, desert dust, aged smoke [*Wandinger et al.*, 2002; *Franke et al.*, 2003; *Müller et al.*, 2003]). The decrease of the Ångström exponent from values around 2 in the lowest 1000 m to values around 1.4 above indicates that the measurements at the Leipzig site are strongly influenced by local sources (new particle formation, emission of particles [*Mattis et al.*, 2004]). Aged and cloud-processed aerosol particles from remote areas increasingly contribute to light

scattering and extinction with increasing height. Whereas the height of the PBL, i.e., the height up to which locally produced aerosols can be mixed, is 1500–3000 m during the summer halfyear, the PBL is typically lower than 1200 m at Leipzig during the winter halfyear [*Mattis et al.*, 2004]. For the wavelength conversion of backscatter profiles measured at 532 nm at the stations in Paris, Belsk, and Minsk the mean Ångström exponent of 1.6 was used, because these sites are located, similar to Leipzig, in industrialized, urban areas with minor influence of large maritime particles on the optical effects. The uncertainty in the wavelength conversion factor is assumed to be 25%, and corresponds to an uncertainty of ±0.6 in the Ångström exponent (see Figure 1).

[13] The changes in the aerosol optical properties along main air mass advection routes were investigated on the basis of the obtained set of backscatter profiles, column-integrated backscatter coefficients, optical depths, and mean PBL top heights. The overall uncertainty (standard deviation) in the lidar-derived air mass mean and seasonal mean optical depths, discussed in the next section, is 15% (Aberystwyth, Hamburg, Leipzig, Munich) to 25% (Paris, Belsk, Minsk) taking uncertainties in the assumed constant backscatter in the near field (10% uncertainty), in the column mean lidar-ratio estimates (10% uncertainty), and in the column mean wavelength conversion factor (15%, Paris, Belsk, Minsk) into account. These air mass related and seasonal mean values rely typically on 5–15 and 10–25 observations, respectively.

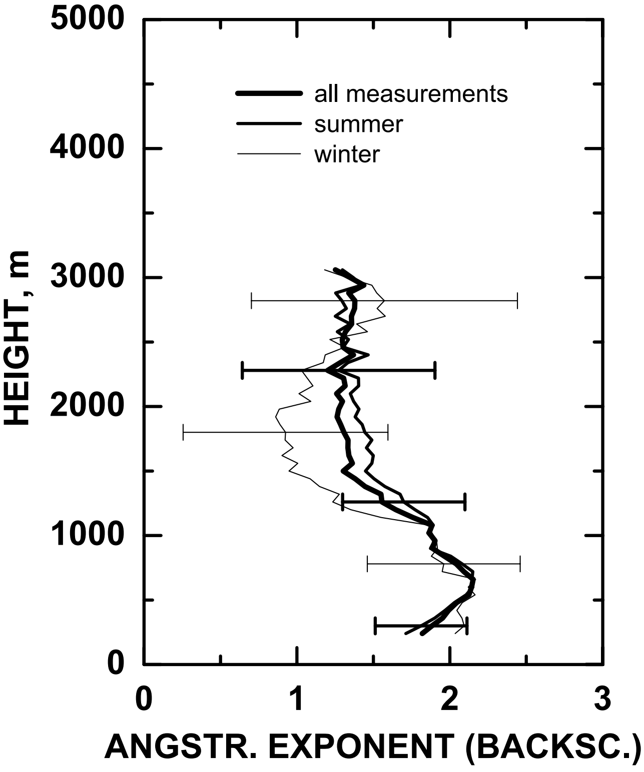

Figure 1. Ångström exponent for backscattering at 355/532 nm derived from all EARLINET measurements at Leipzig. Error bars indicate the standard deviation of single profiles from the summer and winter means.

3. Results

3.1. Reference Profiles for Clean Conditions

[14] Aberystwyth is the most northwesterly station of the network and is located directly at the west coast of Wales. The arriving air masses very often did not have any land contact for several days before detection. The measurements at this site can therefore be used to define reference profiles which represent the properties of air masses entering Europe from clean environments in the North Atlantic.

[15] In the first step, seasonal averages from all measurements at Aberystwyth were investigated. As an example, Figures 2a and 2d show the measurements of the particle backscatter coefficient at 355 nm performed in the summer halfyear of 2001 and in the winter halfyear of 2001/2002 together with the mean profiles for these seasons. All backscatter coefficients in Figure 2 and in the following four figures (Figures 3–6) are shown as a function of height above sea level (asl).

[16] The backscatter peaks in Figure 2a below 2500 m indicate the top of the PBL of the individual measurements. Such pronounced PBL tops are mostly absent during the winter halfyear. The PBL characteristics of individual cases are widely smoothed out when the profiles are averaged (see Figures 2b and 2e).

[17] The mean values in the boundary layer over Aberystwyth are of the order of 2–4 $\mathrm{Mm}^{-1}\ \mathrm{sr}^{-1}$ (1 $\mathrm{Mm}^{-1}\ \mathrm{sr}^{-1} = 10^{-6}\ \mathrm{m}^{-1}\ \mathrm{sr}^{-1}$, see Figures 2a and 2d). By multiplying the column-integrated backscatter coefficients of about $4 \times 10^{-3}\ \mathrm{sr}^{-1}$ (summer 2001) and $5 \times 10^{-3}\ \mathrm{sr}^{-1}$ (winter 2001/2002) with a lidar ratio of 30 sr, which is typical for clean to slightly polluted maritime conditions [*Ansmann et al.*, 2001; *Franke et al.*, 2003], we obtain particle optical depths of 0.12 and 0.15, respectively. Similar values were found for the other seasons.

[18] In the next step, the measurements were grouped with respect to air mass origin. In order to find reference profiles for conditions of lowest anthropogenic influence, measurements from all five seasons were considered for which the air was advected from clean maritime environments to the north and west of Great Britain and Ireland. From these measurements we selected those with maximum backscatter coefficients <2 $\mathrm{Mm}^{-1}\ \mathrm{sr}^{-1}$ for the reference profile calculations. The resulting mean profiles for summer and winter conditions are shown in Figures 2b and 2e (solid curves), respectively.

[19] The solid trajectories in Figures 2c and 2f give the corresponding backward trajectories. The dashed curves in Figures 2b and 2e represent the averages from all remaining profiles (northern and western directions, maximum backscatter coefficients >2 $\mathrm{Mm}^{-1}\ \mathrm{sr}^{-1}$). The corresponding backward trajectories are indicated by the dashed lines in Figures 2c and 2f.

[20] Two main flows, one from the north across or along Scotland and one from the west across or around Ireland, turn out to cause the lowest observed aerosol loads at Aberystwyth. The profiles shown in Figures 2b and 2e can be assumed to represent the reference profiles for very clean (solid lines) and slightly polluted air (dashed lines) that arrives at the European continent rim from the pristine North Atlantic. No major differences are found between

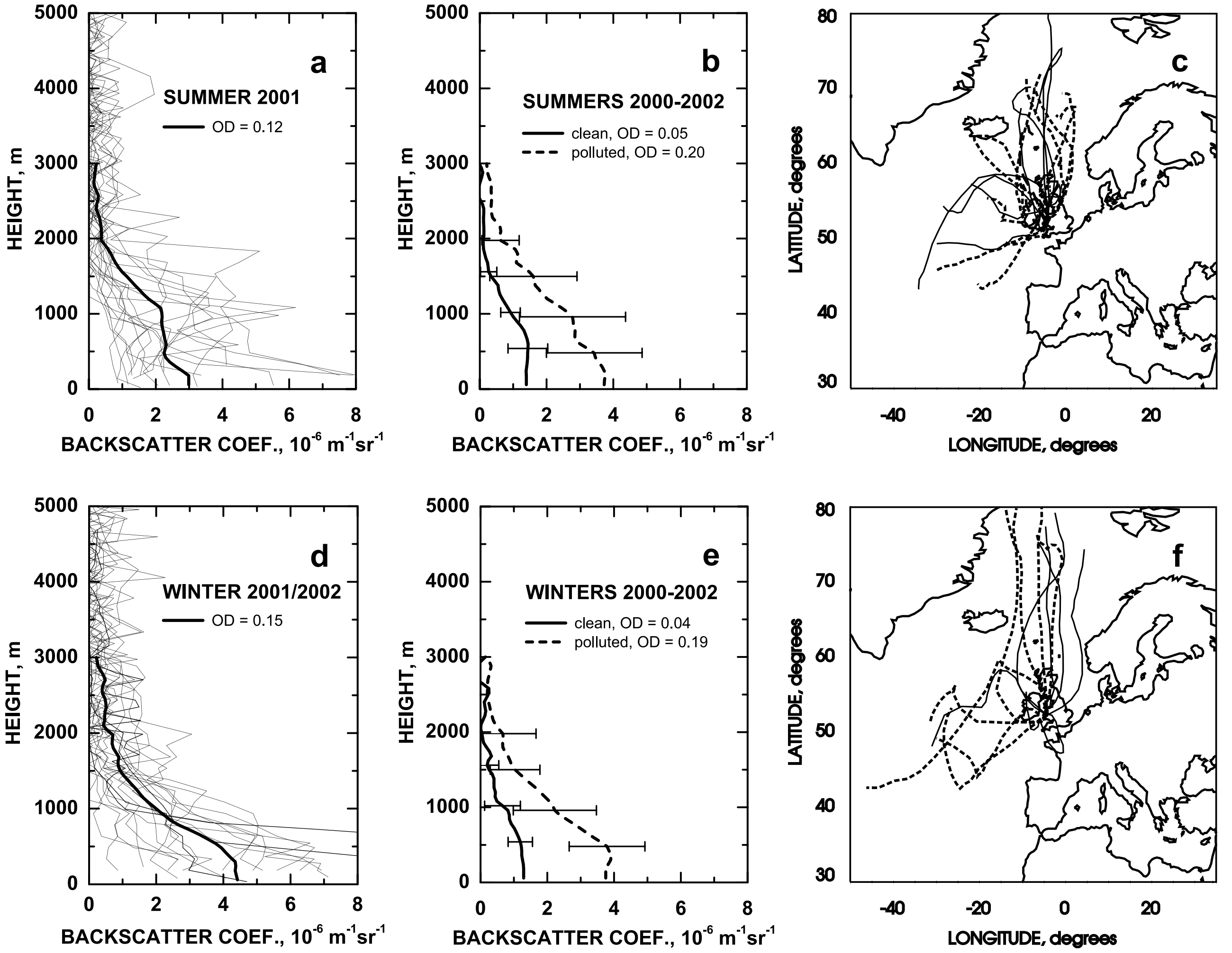

Figure 2. Backscatter-coefficient profiles at 355 nm observed at Aberystwyth (a) during the summer halfyear of 2001 and (d) the winter halfyear of 2001/2002, together with the mean profiles for these seasons. (b) Mean summer and (e) mean winter backscatter-coefficient profiles and corresponding backward trajectories at 975 hPa, (c) and (f), for clean (solid) and slightly polluted (dashed) conditions and northerly and westerly flows. Error bars indicate the standard deviation of single profiles from the mean. The 355-nm optical depths (OD) are given as numbers.

summer and winter seasons. A mean optical depth of about 0.05 and 0.04 is obtained for very clean conditions during the summer and winter halfyears, respectively, assuming a typical lidar ratio for maritime aerosols of 30 sr. These values are in agreement with lidar observations during the Aerosol Characterization Experiment 2 (ACE 2) for westerly airflows (rather clean maritime conditions) at the southern Portuguese coast [*Ansmann et al.*, 2001]. The EARLINET observations are also in agreement with other measurements in clean maritime environments [*Kaufman et al.*, 2001; *Franke et al.*, 2003].

[21] Optical depths of 0.20 (summer) and 0.19 (winter) are found at Aberystwyth for slightly polluted conditions, applying a lidar ratio of 40 sr to the mean backscatter coefficients. The increase in aerosol load can be explained by pollution uptake during the travel over Ireland, Scotland, and northern England but also over maritime sites which are influenced by the dense ship traffic in the English Channel and southwest of it. In addition, an increase of the concentration of sea-salt particles in the case of strong winds over the ocean has to be taken into account as a natural source.

3.2. Air Mass Modification Over the Continent

[22] Northwesterly and westerly flows across western and central Europe were investigated to study the modification of the aerosol properties in this region. Measurements at Hamburg, Paris, Leipzig, and Belsk were used. Figure 3 shows mean profiles for summer (Figure 3a) and winter conditions (Figure 3b) in the case of northwesterly flows at Hamburg and Leipzig. These profiles are compared with the reference profiles from Aberystwyth. Panel 3c indicates the characteristic flows which were used in the investigation. From Figures 3a and 3b it becomes clear that both the particle optical depths as well as the mean PBL heights increase while the air travels into the continent. As is especially the case for Belsk and Minsk (discussed below), a pronounced backscatter peak appears at Leipzig below

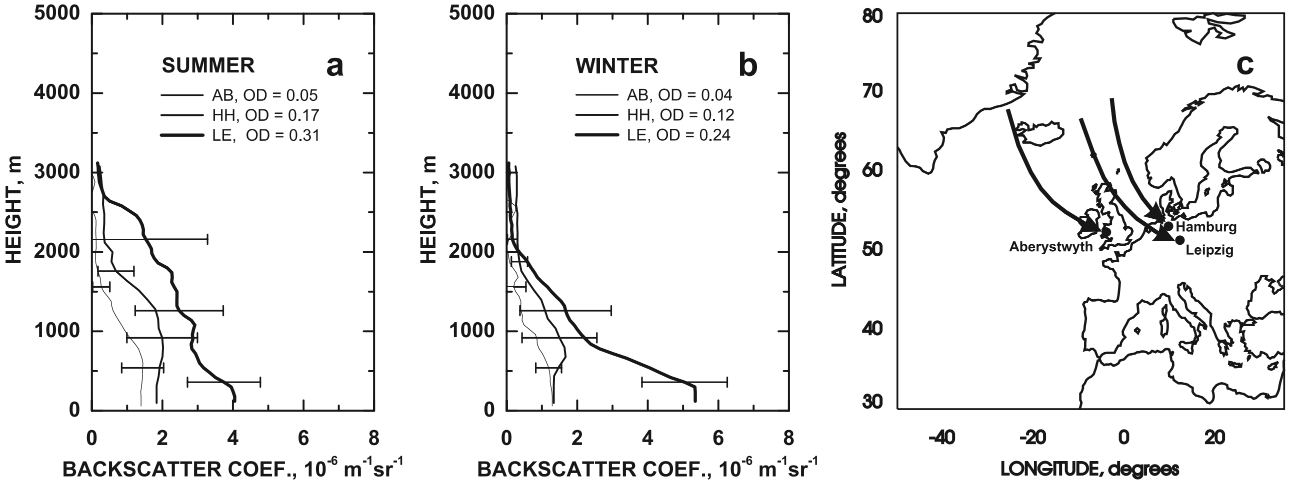

Figure 3. Mean backscatter-coefficient profiles at 355 nm observed in northwesterly flows at Aberystwyth (AB, clean conditions only), Hamburg (HH), and Leipzig (LE) during (a) summer and (b) winter. Error bars indicate the standard deviation of single profiles from the mean. The 355-nm optical depths (OD) are given as numbers. (c) Typical flow pattern.

1000-m height in winter, reflecting the frequent occurrence of very shallow boundary layers during the cold seasons. Aerosol pollution is trapped in these shallow PBLs with heights significantly below 1000 m.

[23] Hamburg is located about 100 km away from the southeastern coast of the North Sea. Northerly and northwesterly flows arriving from the North Sea are expected to represent slightly polluted maritime conditions similar to those at Aberystwyth. Relatively clean air was observed in most cases indeed. The column values are similar to the mean values for northerly and westerly flows at Aberystwyth, considering both the clean and the slightly polluted cases (see Figure 2, solid and dashed lines). In comparison to the reference profiles, the uptake of aerosol particles is obvious. Optical depths of 0.17 and 0.12 in summer and winter, respectively, are found assuming a lidar ratio of 50 sr. Industrialized regions adjacent to the North Sea, the ship traffic in the North Sea, and local sources are responsible for the increased aerosol load.

[24] Air masses which have crossed northern Germany and arrive at Leipzig from northwesterly directions show a further increase of the aerosol load and the PBL height (see Figure 3, thick solid lines). With a lidar ratio of 50 sr, optical depths of 0.31 and 0.24 are found in summer and winter, respectively.

[25] Westerly flows originating in the North Atlantic and traveling along the northwestern rim of the European continent were used to quantify the impact of pollution over industrialized regions in central/western Europe on

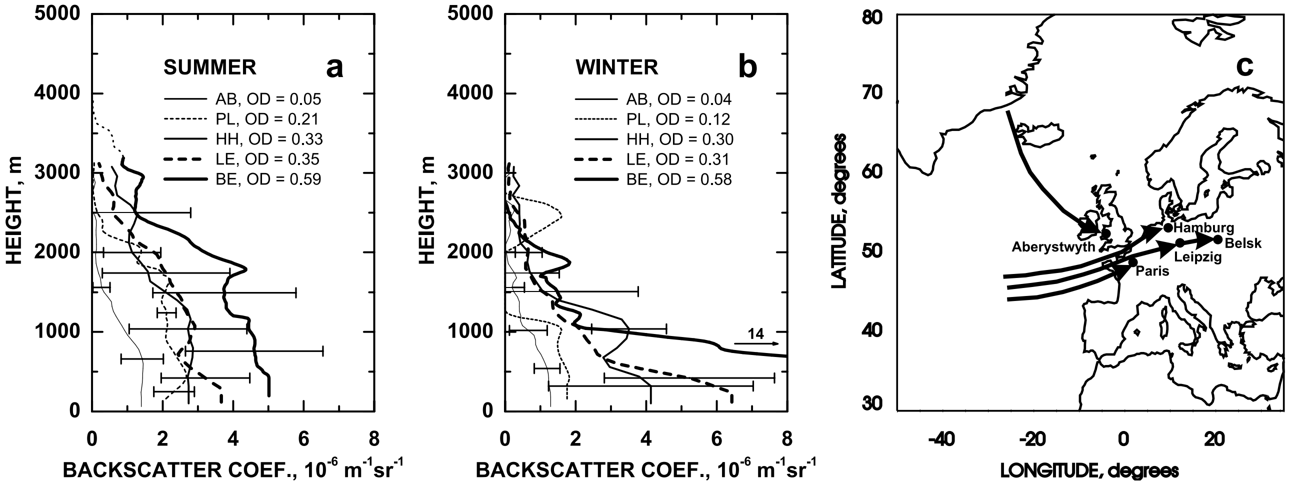

Figure 4. Mean backscatter-coefficient profiles at 355 nm observed in westerly flows at Aberystwyth (clean conditions only), Paris (Palaiseau, PL), Hamburg, Leipzig, and Belsk (BE) during (a) summer and (b) winter. Error bars indicate the standard deviation of single profiles from the mean. The 355-nm optical depths (OD) are given as numbers. (c) Typical flow pattern.

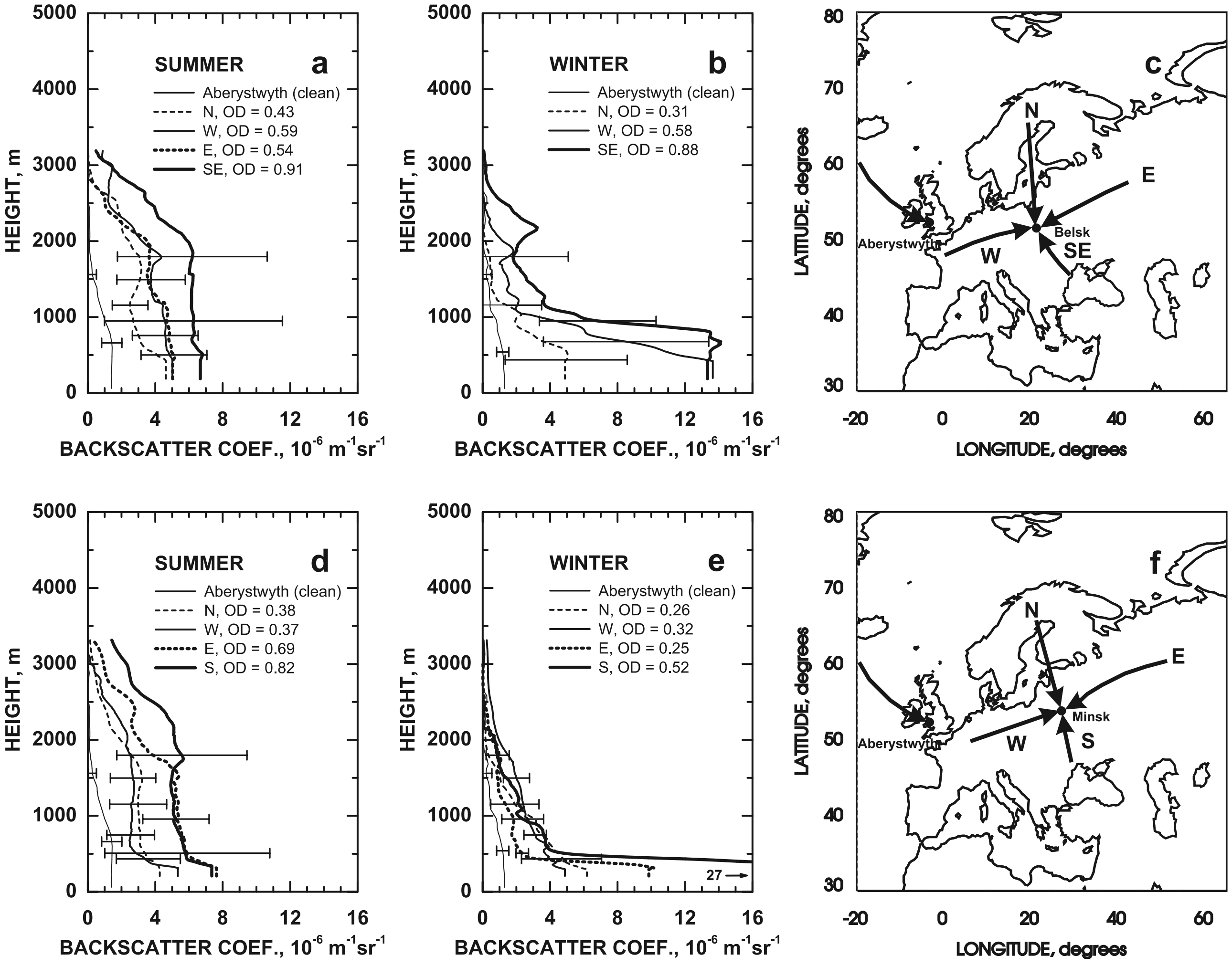

Figure 5. Mean backscatter-coefficient profiles at 355 nm observed in westerly (W), northerly (N), easterly (E), and southeasterly (SE) flows at Belsk during (a) summer and (b) winter and in westerly, northerly, easterly, and southerly (S) flows at Minsk during (d) summer and (e) winter. Error bars indicate the standard deviation of single profiles from the mean. The 355-nm optical depths (OD) are given as numbers. (c) and (f) Typical flow pattern.

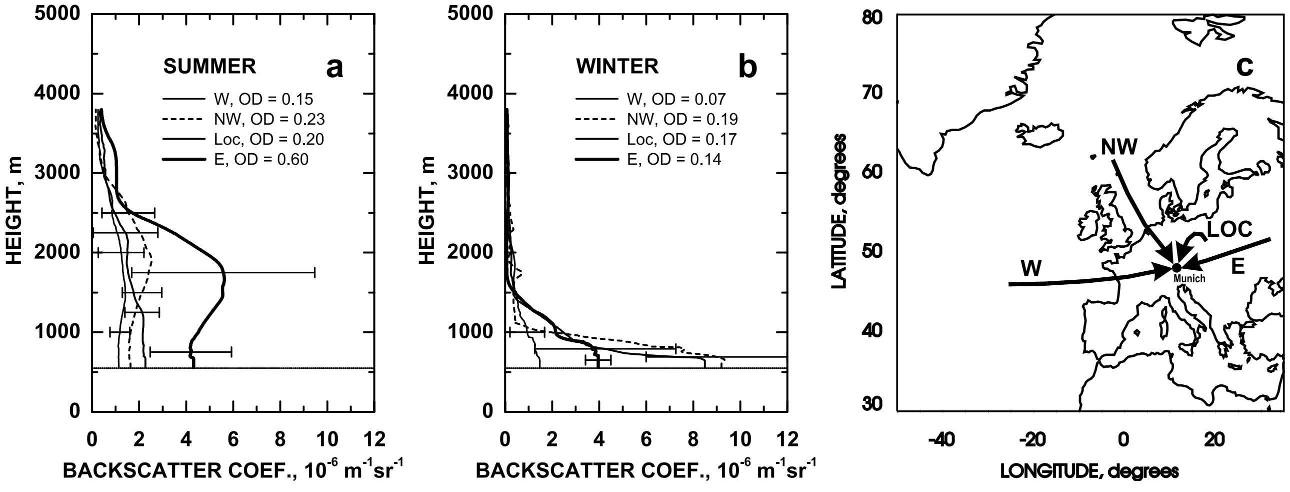

Figure 6. Mean backscatter-coefficient profiles at 355 nm observed in westerly, northwestery, easterly, and local (Loc) flows at Munich during (a) summer and (b) winter. Error bars indicate the standard deviation of single profiles from the mean. The 355-nm optical depths (OD) are given as numbers. (c) Typical flow pattern.

the optical properties further east. In Figure 4, mean backscatter profiles for westerly flows obtained at Paris, Hamburg, Leipzig, and Belsk are shown in comparison to the reference profiles. Air masses reaching Paris from the west could not yet uptake much pollution. The lidar station is located at Palaiseau which is to the southwest of the metropolitan and therefore not in the pollution plume of the city when the air is advected from the west.

[26] Air masses that reach Hamburg and Leipzig from the west have already passed highly industrialized regions in northwestern Europe, namely Belgium, The Netherlands, southern England, and western Germany. This area has the highest population density in Europe. The optical depth for westerly flows at Hamburg is estimated to 0.33 and 0.30 in summer and winter, respectively, with a lidar ratio of 60 sr for polluted aerosols. Similar values of 0.35 and 0.31, respectively, are found at Leipzig. The Leipzig lidar data have been compared with the Aerosol Robotic Network (AERONET) Sun phototmeter observations of the particle optical depth at Leipzig. Good agreement was found [*Mattis et al.*, 2004].

[27] The aerosol concentration significantly increases when the air masses cross Poland. The particle optical depth at Belsk (40 km southwest of Warsaw) is almost twice as high as at Leipzig with mean values of 0.59 in the summer halfyear and 0.58 in the winter halfyear for westerly flows. Observations at Leipzig, performed under opposite advection conditions, i.e., taken in air masses that traveled to the west corroborates that Poland is a significant source of anthropogenic particles. The optical depth also increases by almost a factor of 2 under these airflow conditions. For easterly winds the optical depths are 0.62 (summer) and 0.40 (winter) at Leipzig (see also section 4).

[28] Figure 5 shows seasonal mean profiles for different flows observed at Belsk and Minsk (Belarus). Flows from all directions were investigated. Only measurements for which the air arrived from south of the Carpathian Mountains were excluded. In Belsk a very high aerosol load distributed over 3 km in height is found in summer for all flow plattern. Northerly flows show the lowest values with optical depths of 0.43 in summer and 0.31 in winter. As mentioned, westerly flows cause optical depths of 0.59 and 0.58 in summer and winter, respectively. Unfortunately, observations taken in winter during easterly airflows are not available. For the summer, an optical depth of 0.54 is found in the case of easterly winds. Highest mean values around 0.9 are observed when the air masses arrive from southeasterly directions, i.e., from the Ukraine and the Black Sea region.

[29] Similar aerosol conditions were observed at Minsk. However, compared to Belsk, the optical depths were on average lower by 20%. AERONET Sun photometer observations at Belsk and Minsk (taken from the AERONET database) are in agreement with this finding. Mean values of the particle optical depth of 0.6–0.65 (Belsk, March to December, 2002) and 0.45 (Minsk, January to December, 2003) were estimated from the Sun photometer measurements for the wavelength of 355 nm. Also at Minsk, the highest aerosol load is found when the air arrives from the Ukraine and the Black Sea region.

[30] The strong backscattering below 500-m (Minsk) and 1000-m height (Belsk) during the winter half year, especially for eastern and southeastern directions (see Figures 5b and 5e), is caused by local and regional aerosol sources, but indicates also the influence of the sometimes rather low wintertime PBL heights on the aerosol concentration near the ground.

[31] Munich in southern Germany, 520 m above sea level, is about 1000 km away from the Atlantic Ocean and the Baltic Sea and almost completely surrounded by mountains. Hundred kilometers south of Munich the Alps build a significant barrier (for air pollution) with mountains higher than 3000 m, whereas mountains in the east and northeast are 1500 m high and between 100 and 300 km away. Several hundred kilometers west of Munich, mountains (ridges of 800–1400-m height) stretch from the black forest area to the northeast. Hence, even if we restrict the investigation to westerly and northwesterly flows, which are not influenced by the Alps, the orography has an impact on the air mass transport.

[32] Four advection pattern have been investigated: (1) the westerly flow across central France and southern Germany, (2) the northwesterly flow from the North Sea region across Belgium, The Netherlands, and western Germany, (3) the easterly flow from continental sites in central and eastern Europe, and (4) local flows mainly across southern Germany. The findings are shown in Figure 6.

[33] Very shallow aerosol distributions were found during the winter halfyear. Aerosols were almost absent above 1500-m height (1000 m above ground). The shallow winter aerosol distributions in Figure 6b suggest that boundary layer developments are very weak in winter and that advection of aerosols, e.g., from the polluted eastern Europe, cannot have a significant impact on the aerosol load over Munich. The opposite situation is the case in summer when the aerosol is typically mixed up to 2000–3000 m above ground.

[34] In comparison to the other German stations, the observations at Munich in general show lower optical-depth values and a much stronger seasonal variation. The Munich lidar data were also compared with Sun photometer data. In these sporadic comparisons good agreement was obtained. A very low aerosol load is observed in the case of westerly winds. The optical depths of 0.15 in summer and 0.07 in winter (lidar ratio 50 sr) are comparable to those observed in maritime environments at the continental rim. Obviously, air masses do not cross strong source regions on their way across central France and southern Germany. Local flows lead to moderate optical depths of the order of 0.2, which indicate that there are no strong aerosol sources in Bavaria. Somewhat higher values are found for northwesterly flows what can be interpreted as the influence from the source regions in central/western Europe. For easterly flows, high optical depths of 0.6 (lidar ratio 60 sr) are observed in summer, whereas in winter the optical depth is only 0.14. A possible explanation is that the transport from source regions in eastern Europe, especially from Bohemia, across the mountain sites to the east and northeast of Munich can only take place if convection leads to an significant upward mixing of polluted air, which obviously does not occur in winter. Consequently, the free troposphere over Munich is very clean. Low temperature

Table 2. Mean Aerosol Properties Obtained at Seven EARLINET Sites for Different Airflows, Sorted From Low to High Summertime Particle Optical Depth[a]

EARLINET Site, Flow	Integr. Backsc. Coef. (355 nm), 10^{-3} sr^{-1} Su/Wi	Lidar Ratio (Estimate), sr	Optical Depth (355 nm) Su/Wi	PBL Height (Mean), km Above Ground Su/Wi
Aberystwyth, N/W clean	1.8/1.5	30	0.05/0.04	1.5/0.9
Munich, W	2.9/1.3	50	0.15/0.07	2.2/1.3
Hamburg, NW	3.3/2.4	50	0.17/0.12	1.6/1.1
Aberystwyth, N/W poll.	5.1/4.8	40	0.20/0.19	1.7/1.3
Munich, local	4.1/3.4	50	0.20/0.17	2.0/0.7
Paris, W	4.2/2.3	50	0.21/0.12	1.9/1.2
Munich, NW	4.5/3.7	50	0.23/0.19	2.2/0.4
Leipzig, NW	6.2/4.8	50	0.31/0.24	2.1/1.4
Hamburg, W	5.5/5.1	60	0.33/0.30	1.7/1.2
Leipzig, W	5.9/5.2	60	0.35/0.31	1.9/1.3
Minsk, N	6.4/4.4	60	0.38/0.26	2.6/1.2
Minsk, W	6.2/5.3	60	0.37/0.32	2.2/1.9
Belsk, N	7.1/5.1	60	0.43/0.30	2.4/1.0
Belsk, E	9.0/–	60	0.54/–	2.1/–
Leipzig, local	9.8/5.0	60	0.59/0.30	2.3/1.0
Belsk, W	9.8/9.7	60	0.59/0.58	2.2/1.0
Munich, E	9.9/2.4	60	0.60/0.14	2.0/0.8
Leipzig, E	10.3/6.7	60	0.62/0.40	2.2/0.9
Minsk, E	11.5/4.2	60	0.69/0.25	2.4/1.1
Minsk, S	13.7/8.7	60	0.82/0.52	2.8/0.4
Belsk, SE	15.2/14.6	60	0.91/0.88	2.6/1.3

[a] Relative standard deviations of the column backscatter, the optical depth, and the PBL height are 30–50%. Su and Wi denote summer and winter halfyear, respectively.

inversions over Munich, on the other hand, suppress the vertical exchange so that any locally produced aerosol is trapped in the lowermost layers (see Figure 6b). This hypothesis is supported by the fact that the typical PBL height for all flows is only 400–800 m in Munich in the winter halfyear. The Munich measurements indicate that horizontal transport of polluted air is suppressed by orographic barriers especially in wintertime.

4. Integration of the Observations and Discussion

[35] Table 2 and Figures 7 and 8 summarize the findings discussed above. Figure 7 illustrates the increase of the aerosol burden with increasing penetration path of air masses into the continent. As an example, one may compare the westerly flows arriving at Aberystwyth (blue arrow), Paris (yellow arrow), Leipzig (orange arrow), and Belsk (red arrow). The strongest source of aerosol pollution seems to be central eastern and southeastern Europe. Compared to these areas, the aerosol sources in western and central Europe contribute only moderately to the European haze plume. Table 2 corroborates this finding. The selected flows for the seven sites are sorted by increasing summertime optical depth from 0.05 to 0.91. During westerly flows most summertime optical depths (except for Belsk) were <0.35 at 355 nm. During easterly flows, optical depths accumulate between 0.5 and 0.7. The winter mean optical depths were mostly 10%–30% lower than the summer values. Reasons might be the contribution of natural particles (biogenic particles, agricultural activities, erosion), the higher photochemical conversion rate of precursor gases into particles in summer, and meteorological conditions (larger humidity effects on particle growth in summer).

[36] The mean PBL heights in Table 2 are determined from the individual PBL heights which are given, in most cases, by the steep slope in the backscatter-coefficient profile [*Bösenberg et al.*, 2003]. In cases with complex layering (10%–20% out of all cases) the PBL height was estimated by eye from the backscatter profiles. Temperature profiles of nearby radiosondes were taken into account to identify the free troposphere. Mean summer PBL heights are 1.9–2.8 km at the continental sites of Leipzig, Belsk, and Minsk and 1.5–1.9 km at Aberystwyth and for westerly and northwesterly flows at Paris and Hamburg. During the winter halfyear boundary layer heights were mostly found from 0.7–1.3 km.

[37] Figure 8 finally gives an impression how the particle optical depth, the boundary layer height, and the boundary layer mean extinction coefficient increase with distance from the Atlantic Ocean. A pronounced seasonal dependence of the boundary layer height is found at all stations for the selected air mass transport clusters. The estimated boundary layer mean extinction coefficient at 355 nm is, in most cases (with the exception of Belsk), slightly higher in winter than in summer mainly caused by the low boundary layer heights in winter. High PBL mean winter extinction coefficients of the order of 500–600 Mm^{-1} as observed at Belsk were measured at Leipzig 10 years ago, shortly after the German unification [*Mattis et al.*, 2004]. The shown PBL mean extinction coefficients are simply calculated from the mean values of the optical depth and the PBL height presented in Figures 8a and 8b. These values are close to respective mean values that were calculated from the individual observations of the ratio of optical depth to PBL height. On average, the PBL aerosol contributes about 80%–90% to the optical depth (up to 3 km height) so that the PBL extinction coefficients in Figure 8c are overestimated by roughly 10%–20% [*Bösenberg et al.*, 2003; *Mattis et al.*, 2004]. Mean PBL extinction coef-

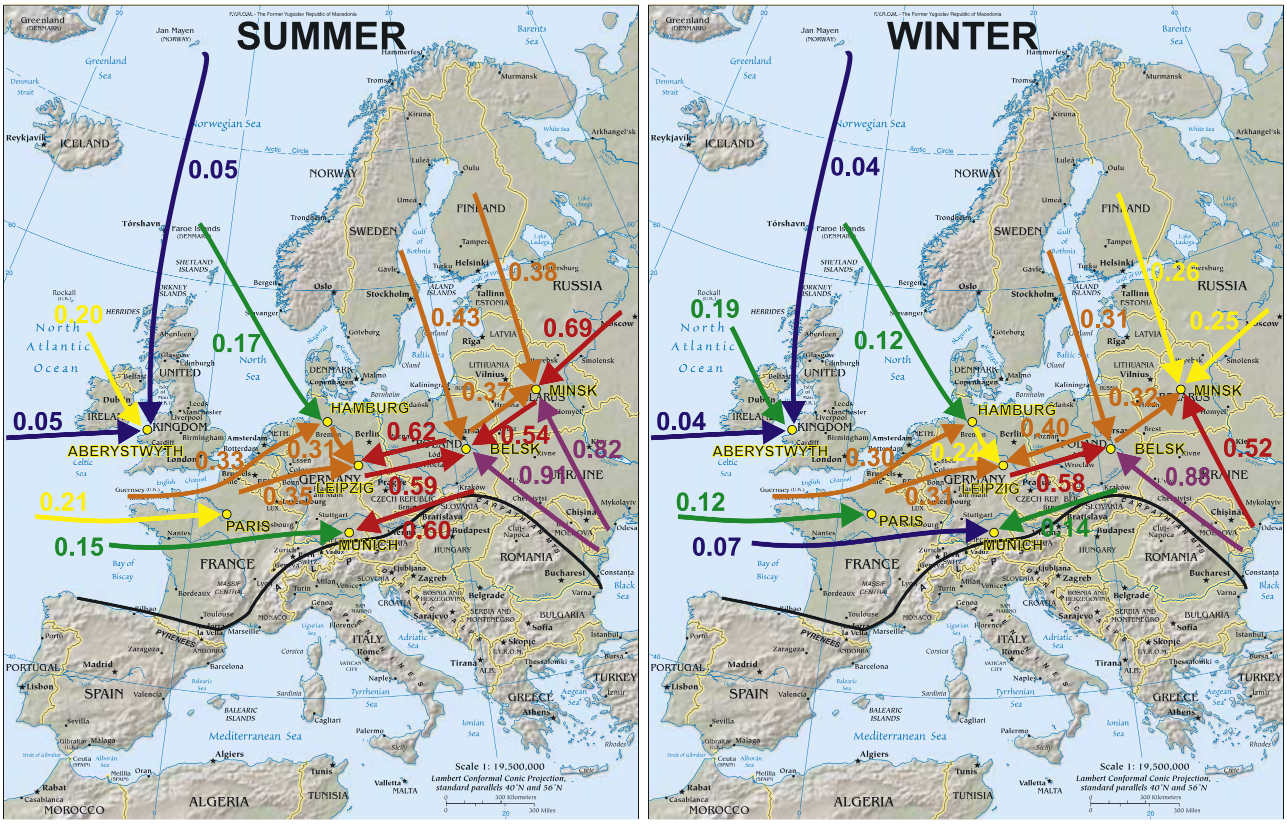

Figure 7. Overview of investigated airflows and the corresponding 355-nm optical depths for the summer and the winter halfyear. Blue, green, yellow, orange, red, and violet arrows indicate mean optical depths of 0–0.09, 0.1–0.19, 0.2–0.29, 0.3–0.49, 0.5–0.69, and >0.7, respectively. Optical depths (also listed in Table 2) are given in addition.

ficients are of the order 200 Mm^{-1} at 355 nm in central Europe for westerly flows, and decrease to 100 Mm^{-1} at Hamburg, when the air is advected from the North Sea. Under clean conditions at Aberystwyth the PBL mean extinction coefficients are below 50 Mm^{-1}, and thus much lower than typical values for Oceanic sites [*Franke et al.*, 2003]. The higher PBLs over Aberystwyth (1000–1500 m) cause this effect. Typical maritime boundary layers reach up to 500–900 m only.

[38] Finally, we made an attempt to estimate the average effective aerosol mass flux (upward flux minus downward flux caused by removal processes such as washout and deposition) that is consistent with the optical-depth increase from Aberystwyth to Hamburg and Leipzig, and from Leipzig to Belsk. The measured increase of the 355-nm optical depth by 0.28, i.e., from 0.05 at Aberystwyth (western boundary of Europe) to about 0.33 at Hamburg and Leipzig (about 1200 km east of Aberystwyth), corresponds to an increase of the 500-nm optical depth by 0.17 (assuming an Ångström exponent of 1.6). If we now assume a specific extinction coefficient of 5 m^2/g at 500 nm [*Charlson et al.*, 1992] for sulfate aerosol, the column mass of anthropogenic particles increased from almost zero to 0.034 g/m^2 after the travel of 1000–1500-km from the Atlantic to central Europe (Leipzig), and, in terms of PBL mean aerosol mass concentration, to roughly 22 μg/m^3, assuming a PBL height of 1500 m (mean value for the whole EARLINET period at Leipzig). The increase of the column mass value correspond to an average effective flux of aerosol mass in the range of 0.11–0.17 μg/(m^2s) along the advection route. Here we assumed a mean wind speed (westerly winds) of 5 m/s along the 1000–1500-km path so that aerosol uptake could occur for 2.3–3.5 days. Note that aerosol water is included in the aerosol mass values because the lidar observations are conducted at ambient humidity conditions.

[39] The aerosol flux values estimated from the lidar data correspond to mean effective aerosol mass fluxes in the range of 8700–13000 Mg per year and 50 km × 50 km grid cell (between the Atlantic and central Europe). UNECE/EMEP (United Nations Economic Commission for Europe/Co-operative programme for monitoring and evaluation of the long-range transmissions of air pollutants in Europe) emission maps (emission database WebDab, http://webdab.emep.int [*UNECE*, 2003]) for particulate matter and sulfur dioxide suggest mean (upward) aerosol emissions in the range of 8000–20000 Mg per year and 50 km × 50 km grid cell when the air is advected from westerly directions towards central Europe.

[40] Based on the optical-depth increase from Leipzig to Belsk (600-km distance) of about 0.25 (355 nm) or 0.14 (500 nm) and by assuming again a specific particle light-extinction coefficient of 5 m^2/g and a mean wind

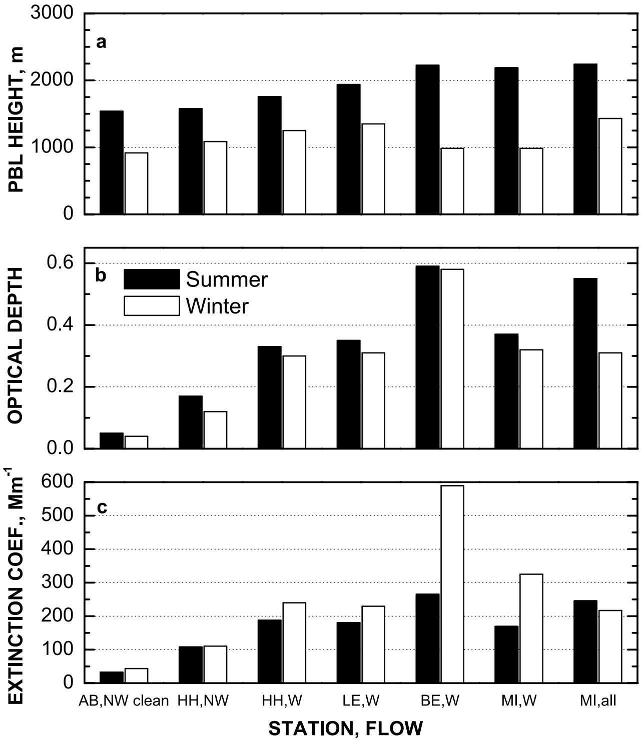

Figure 8. Development of the summer and winter mean boundary layer depth, 355-nm optical depth, and boundary layer mean extinction coefficient (mean optical depth (center) divided by the mean boundary layer depth (top)) with distance from the Atlantic Ocean. NW and W stand for northwesterly and westerly flows. Standard deviations of the optical depth and the PBL height are of the order of 30–50%.

speed of 5 m/s we end up with a further increase of the column aerosol mass by 0.03 g/m^2 after the travel from Leipzig to Belsk, a mean effective flux of aerosol mass of 0.25 μg/(m^{2}s), and mean effective aerosol emission of about 20000 Mg per year and 50 km × 50 km grid cell. EMEP data suggest mean aerosol mass emissions in the range of 15000–25000 Mg per year and 50 km × 50 km grid cell between Leipzig and Belsk.

5. Summary

[41] The air mass modification over the European continent north of the Pyrenees and Alps was investigated in terms of aerosol optical properties. For the first time, a seasonal-dependent, height-resolved aerosol study was performed, on the basis of regular EARLINET measurements and of analytical backward trajectories. Typical airflows from northwest and west to southeast and east showed a strong increase in aerosol load for air masses which enter Europe from maritime sites and cross highly populated, industrialized regions.

[42] A mean 355-nm optical depth of 0.05 and a mean PBL height of 1.5 km was found for clean maritime summer conditions at Aberystwyth. The particle optical depth and PBL height increased with increasing distance from the North Atlantic. Mean PBL heights of 1.9–2.3 km were observed at the continental sites of Leipzig, Belsk, and Minsk for the summer halfyear. Mean PBL heights for the winter halfyear were mostly between 0.7 and 1.3 km at the seven EARLINET sites. Summer mean 355-nm optical depths increased from 0.05 (clean maritime) over values around 0.2 (Aberystwyth, slightly polluted; Hamburg, northwesterly flow; Paris, westerly flow) and 0.35 (Hamburg, Leipzig, westerly flow) to values of 0.6 (Belsk, westerly flow). Winter mean optical depths were lower by about 10%–30% than the respective summer values. In the case of easterly air mass transports during the summer halfyear, the 355-nm optical depth increased significantly and reached values of 0.5–0.7 at most of the central European sites. According to the EARLINET observations, Poland and southeastern Europe are strong aerosol source regions, whereas the highly industrialized and populated western European region contributes moderately to the European aerosol burden. The comparably low particle optical depth usually found at the Munich site indicates the sensitive influence of the orography on local haze conditions.

[43] From the optical measurements, effective aerosol mass emissions were estimated. The mean effective aerosol mass fluxes of the order of 10000 Mg (western Europe) and 20000 Mg (Leipzig-Belsk) per year and 50 km × 50 km grid cell were found to be in reasonable agreement with EMEP emission data.

[44] **Acknowledgments.** The financial support of this work by the European Commission under grant EVR1-CT1999-40003 is gratefully acknowledged. Analytical backward trajectories are kindly provided by the German Weather Service on a regular schedule. We thank all coworkers of the EARLINET project for the encouraging and fruitful cooperation.

References

Ansmann, A., F. Wagner, D. Althausen, D. Müller, A. Herber, and U. Wandinger (2001), European pollution outbreaks during ACE 2: Lofted aerosol plumes observed with Raman lidar at the Portuguese coast, *J. Geophys. Res.*, *106*, 20,725–20,734.

Ansmann, A., F. Wagner, D. Müller, D. Althausen, A. Herber, W. von Hoyningen-Huene, and U. Wandinger (2002), European pollution outbreaks during ACE 2: Optical properties inferred from multiwavelength lidar and star-Sun photometry, *J. Geophys. Res.*, *107*(D15), 4259, doi:10.1029/2001JD001109.

Böckmann, C., et al. (2004), Aerosol lidar intercomparison in the framework of the EARLINET project. 2. Aerosol backscatter algorithms, *Appl. Opt.*, *43*, 977–989.

Bösenberg, J., et al. (2003), EARLINET: A European Aerosol Research Lidar Network to establish an aerosol climatology, *MPI Rep. 348*, Max-Planck-Inst. für Meteorol., Hamburg, Germany.

Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakley Jr., J. E. Hansen, and D. J. Hofmann (1992), Climate forcing by anthropogenic aerosols, *Science*, *255*, 423–430.

Franke, K., A. Ansmann, D. Müller, D. Althausen, F. Wagner, and R. Scheele (2001), One-year observations of particle lidar ratio over the tropical Indian Ocean with Raman lidar, *Geophys. Res. Lett.*, *28*, 4559–4562.

Franke, K., A. Ansmann, D. Müller, D. Althausen, C. Venkataraman, M. S. Reddy, F. Wagner, and R. Scheele (2003), Optical properties of the Indo-Asian haze layer over the tropical Indian Ocean, *J. Geophys. Res.*, *108*(D2), 4059, doi:10.1029/2002JD002473.

Kaufman, Y. J., A. Smirnov, B. N. Holben, and O. Dubovik (2001), Baseline maritime aerosol: Methodology to derive the optical thickness and scattering properties, *Geophys. Res. Lett.*, *28*, 3251–3254.

Kottmeier, C., and B. Fay (1998), Trajectories in the Antarctic lower troposphere, *J. Geophys. Res.*, *103*, 10,947–10,959.

Matthias, V., et al. (2004a), Aerosol lidar intercomparison in the framework of the EARLINET project. 1. Instruments, *Appl. Opt.*, *43*, 961–976.

Matthias, V., et al. (2004b), Vertical aerosol distribution over Europe: Statistical analysis of Raman lidar data from 10 EARLINET stations, *J. Geophys. Res.*, *109*, D18201, doi:10.1029/2004JD004638.

Mattis, I., A. Ansmann, D. Müller, U. Wandinger, and D. Althausen (2004), Multiyear aerosol observations with dual-wavelength Raman lidar in the framework of EARLINET, *J. Geophys. Res.*, *109*, D13203, doi:10.1029/2004JD004600.

Müller, D., I. Mattis, U. Wandinger, D. Althausen, A. Ansmann, O. Dubovik, S. Eckhardt, and A. Stohl (2003), Saharan dust over a central European EARLINET–AERONET site: Combined observations with Raman lidar and Sun photometer, *J. Geophys. Res.*, *108*(D12), 4345, doi:10.1029/2002JD002918.

Pappalardo, G., et al. (2004), Aerosol lidar intercomparison in the framework of the EARLINET project: 3. Raman lidar algorithm for aerosol extinction, backscatter, and lidar ratio, *Appl. Opt.*, *43*, 5370–5385.

Remer, L. A., and Y. J. Kaufman (1998), Dynamic aerosol model: Urban/industrial aerosol, *J. Geophys. Res.*, *103*, 13,859–13,871.

Remer, L. A., Y. J. Kaufman, and B. N. Holben (1999), Interannual variation of ambient aerosol characteristics on the east coast of the United States, *J. Geophys. Res.*, *104*, 2223–2231.

United Nations Economic Commission for Europe (UNECE) (2003), Present state of emission data, *Rep. EB.AIR/GE1/2004/10*, Geneva.

von Hoyningen-Huene, W., M. Freitag, and J. B. Burrows (2003), Retrieval of aerosol optical thickness over land surfaces from top-of-the–atmosphere radiance, *J. Geophys. Res.*, *108*(D9), 4260, doi:10.1029/2001JD002018.

Wandinger, U., et al. (2002), Optical and microphysical characterization of biomass-burning and industrial-pollution aerosols from multiwavelength lidar and aircraft measurements, *J. Geophys. Res.*, *107*(D21), 8125, doi:10.1029/2000JD000202.

Wendisch, M., et al. (2004), Airborne measurements of areal spectral surface albedo over different sea and land surfaces, *J. Geophys. Res.*, *109*, D08203, doi:10.1029/2003JD004392.

A. Ansmann, I. Mattis, M. Tesche, and U. Wandinger, Leibniz Institute for Tropospheric Research, Permoserstr. 15, 04318 Leipzig, Germany. (albert@tropos.de; ina@tropos.de; ulla@tropos.de)

J. Bösenberg and H. Linné, Max Planck Institute for Meteorology, Bundesstr. 53, 20146 Hamburg, Germany. (boesenberg@dkrz.de; linne@dkrz.de)

A. Chaikovski, Institute of Physics, National Academy of Sciences of Belarus, 68, F. Scarina Ave., 220072 Minsk, Belarus. (chaikov@dragon.bas-net.by)

V. Freudenthaler and M. Wiegner, Meteorologisches Institut der Ludwig Maximilians-Universität, Theresienstraße 37, 80333 München, Germany. (volker.freudenthaler@meteo.physik.uni-muenchen.de; m.wiegner@meteo.physik.uni-muenchen.de)

L. Komguem and G. Vaughan, Physics Department, University of Wales, Aberystwyth, Ceredigion SY23 3BZ, UK. (llk@aber.ac.uk; gxv@aber.ac.uk)

V. Matthias, Institute for Coastal Research, Max-Planck-Strasse 1, 21502 Geesthacht, Germany. (volker.matthias@gkss.de)

J. Pelon and L. Sauvage, Service d'Aéronomie, Centre National de la Recherche Scientifique, Université Pierre et Marie Curie, Boîte 102, 4 place Jussieu, 75252 Paris Cedex, France. (jacques.pelon@aero.jussieu.fr; sauvage@aero.jussieu.fr)

P. Sobolewski, Institute of Geophysics, Polish Academy of Sciences, Ks. Janusza 64, 01-452 Warsaw, Poland. (pss@ra.onet.pl)