Lindenberg Aerosol Characterization Experiment 1998 (LACE 98): Overview

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Abstract.

Back–scattering and absorption of solar radiation by aerosol particles are an important source of uncertainty in climate predictions. Integrated research on the radiative properties of aerosol may reduce this uncertainty. The Lindenberg Aerosol Characterization Experiment 1998 (LACE 98) contributes to this aim. LACE 98 took place between July 13 and August 12, 1998, near Berlin, Germany. The Lindenberg Meteorological Observatory (52.2° N, 14.1° E) was chosen as the central field site because of its long record with aerosol–optical–depth data. Measurements were performed from three aircraft, with one airborne and four ground–based lidars, and at a ground station. The meteorological situations in which intensive observations were carried out included clean and polluted air masses as characterized by low and high aerosol optical depths. This introductory paper gives an overview of the LACE 98 goals, instrumentation, meteorological and aerosol properties, and reports on the key findings as a guide to the results presented in the more detailed papers that follow. A very remarkable finding should be mentioned beforehand becaue of its unique character: on August 9–10, 1998, a free–tropopsheric aerosol layer was observed that originated from forest fires in western Canada.

1. Introduction

Because of the complex properties of atmospheric aerosol particles, present models of radiative forcing[1] of climate are far from complete and are highly uncertain. As pointed out by *Heintzenberg et al.* [1996], the principle (ultimate) goal of atmospheric aerosol research is the treatment of the aerosol as a prognostic variable in climate models and in models of global chemical cycles. Global monitoring of all chemically and climatically relevant aerosol properties is, however, not possible due to high spatial variability and the prohibitive costs of large numbers of surface stations and airborne sensors. Instead, the approach is to create locally comprehensive sets of data by combining measurements of physico–chemical aerosol properties with radiation measurements and models in so–called closure experiments (explained in section 3). These experiments provide

[1] Climate forcing is a change in heat balance (Watts per square meter) that is imposed upon the climate system by e.g., anthropogenic particles.

the basis for developing the parameterizations of aerosol properties and processes for climate models.

Several integrated field campaigns have been conducted. The Aerosol Characterization Experiment 1 (ACE 1) documented the chemical and physical characteristics of aerosol particles in a remote (clean) marine atmosphere (special issue [*J. Geophys. Res., Vol. 103, No. D13,* 1998]). ACE 2 extended these observations to the North Atlantic Ocean with emphasises on the anthropogenic perturbation of the background aerosol (special issue [*Tellus 52B, No. 2,* 2000]). The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) focussed specifically on the column–integrated radiative effects by one of the world's major plumes of urban/industrial haze (special issues [*J. Geophys. Res., Vol. 104, No. D2,* 1999 and Vol. 105, No. D8, 2000]). In contrast, the Smoke, Clouds, and Radiation–Brazil experiment (SCAR–B, special issue [*J. Geophys. Res. Vol. 103, No. D24,* 1998]) and the Indian Ocean Experiment (INDOEX, special issue [*J. Geophys. Res., Vol. 106, No. ...,* 2001]) concentrated on pollution emitted from less–developed regions. The recently conducted ACE Asia field project will provide new information regarding aerosol formation and fate in eastern Asia. In this context, LACE 98 was planned to extend the efforts of the aforementioned experiments to the highly industrialized central European region.

2. LACE 98 goals

The overall goal of integrated aerosol characterization experiments is to provide experimental data needed to properly describe aerosol particles in atmospheric models. A second focus is to test and improve parameterization schemes which are used in field experiments as well as in climate models to determine aerosol radiative effects from basic chemical and physical/optical properties of aerosol particles. Following this line, the specific tasks of LACE 98 were threefold: (a) Study of the interaction of the atmospheric aerosol system with the radiation field over a polluted central European site during the summer season, (b) quantification of the uncertainties of the optical properties of the particles and associated radiative effects, calculated from measured chemical composition and particle size distribution of the aerosol, and (c) quantification of the direct climatic effect of the observed, anthropogenic aerosol particles. To meet these three objectives, measurements of the aerosol radiative effects (solar irradiances) with vertical resolution are needed in conjunction with profile measurements of chemical, microphysical, and optical aerosol properties.

Another goal of LACE 98 was to introduce new aerosol analyses and measurement techniques and to demonstrate their potential [e.g., *Trimborn et al., Ebert et al., Fiebig et al.,* this issue]. As an example, for the first time, several advanced aerosol lidars were involved in a large aerosol field campaign [*Wandinger et al.,* this issue]. These lidars allow a comprehensive characterization of optical and microphysical properties. Unique aircraft/lidar comparisons could be realized during LACE 98.

A further goal was to measure the ground albedo spectrally resolved and daytime–dependent over the field site to provide the data needed to proof and improve procedures applied in spaceborne remote sensing over land [*Armbruster et al.*, 1999]. Compared to ACE 2, TARFOX, INDOEX, and ACE Asia, which also focussed on pollution plumes from highly industrialized regions, LACE 98 was the first field experiment that took place over a continental site. An important feature over land (heterogeneous terrain) is the spatially and temporally changing surface albedo characteristics which sensitively influences especially the upwelling shortwave radiation and thus the results of satellite remote sensing of the aerosol optical depth [*Veefkind et al.,* 2000; *Deuzé et al.,* 2001; *von Hoyningen–Huene et al.,* 2001] and the estimations of the impact of aerosols on climate [*Wendling et al.,* this issue]. Aerosol remote sensing over land is a fundamental requirement for a global view of aerosol sources, transport pattern, and the resulting radiative effects.

Several less extensive aerosol characterization experiments at continental European sites have been realized before LACE 98 [e.g., *ten Brink et al.,* 1996, 1997; *Mészáros et al.,* 1998]. In North America, large efforts are presently done in the framework of the Atmospheric Radiation Measurement (ARM) project [*Ferrare et al.,* 1998; *Kato et al.,* 2000] at the southern Great Plains, and at Bondville, Illinois, where long–term observations of chemical, physical and optical particle properties are performed [*Koloutsou–Vakakis et al.,* 2001],

3. Closure experiments

Closure studies [*Quinn et al.,* 1996] are of central importance in the investigations of the complex relationships between physical, chemical, and radiative properties of atmospheric aerosol particles and the uncertainties in the findings. In addition, closure studies help to determine the limits of the applied observational techniques, of the data analysis schemes, and the aerosol models applied. First closure studies were reported by *Russel et al.* [1979]. Concerning European aerosols,

one of the first closure experiments was presented by *Veefkind et al.* [1996].

As outlined by *Bates et al.* [1998], in closure experiments an overdetermined set of observations is obtained, and the measured value of a dependent variable is compared with the value that is calculated from the measured values of the independent variables using an appropriate model. The outcome of a closure experiment provides a direct evaluation of the combined uncertainty of the model and the measurements. If there is agreement between the measured and calculated values within the accepted level of uncertainty, the model may be a suitable representation of the observed system and appropriate for use as a component in other higher–order models. Poor agreement indicates that there are problems in the model or measurements that must be revealed and corrected for before proceeding further.

The closure experiments can be divided into local and column closure experiments. In a local closure experiment all measurements are made at a single location (in the same air parcel) simultaneously. The column closure experiment extends the local (zero–dimensional) closure to multiple heights in order to compare column–integrated and vertically resolved radiation measurements with in–situ (aircraft) and remote sensing (lidar, radiometer) observations of aerosol chemical, physical, and radiative properties. Figure 1 illustrates which areas of aerosol properties and radiative effects must be covered by observations and linked to each other by calculations (parameterizations) in the framework of the closure efforts.

Often the observations cannot be done simultaneously and within the same air parcel or column. One important task is therefore to analyze the available wheather charts and backward trajectory calculations in order to carefully select the cases for detailed closure studies.

The following local closure studies have to be addressed in integrated field campaigns in which the direct climatic effects of aerosols (caused by light scattering and absorption) are investigated. The first experiment deals with the consistency regarding the particle mass and chemical composition derived from a variety of size–resolved and size–integrated chemical and physical observations [*Neusüß et al.,* this issue]. Consistency between the mass, which is derived from the measurement of the number size distribution, and the mass, which is obtained from the chemical analyses and the gravimetric measurements, is a prerequisite for further closures studies regarding the optical effects of the particles.

In the second experiment, the directly measured light

scattering, backscattering, and absorption properties of dry aerosol particles (i.e., measured at low relative humidity) are compared to respective values predicted from the measured number size distribution and the size–resolved chemical composition (chemical mass size distribution) of the dry particles [*Wex et al.,* this issue]. Mie scattering theory is applied here, thus the particles are assumed to be spherical. Next to the number size distribution, the refractive index of the probed particle ensemble is the most important input parameter in the Mie scattering calculations. Thus, the approaches applied to quantify the particle refractive index must be analyzed too. Consequently, the refractive index derived from the measured chemical composition was compared with results of alternative (optical) approaches to estimate the refractive index [*Ebert et al.,* this issue].

The third experiment focusses on the response of the aerosol size and the light scattering properties to changes in relative humidity. Observed changes in the particle size and the optical effects are compared with those calculated from the measured (dry) aerosol size distribution, the size–resolved chemical characteristics, and assumed functional relationships between chemical composition and water uptake. [*Busch et al., Bundke et al.*, this issue].

The column closure studies then concentrate on the comparisons of measured profiles of the optical effects and radiative flux densities (irradiances), including the respective column–integrated effects, with the profile and column values that follow from the calculations based on the vertically resolved information on particle size distribution, chemical composition, and humidity dependence of the optical effects. [*Fiebig et al., Petzold et al., Wandinger et al., Wendisch et al., Wendling et al., Wex et al., Wieser et al.*, this issue]. These closure studies also include comparisons of in–situ (aircraft) measured microphysical properties, such as the surface–area weighted mean radius (effective radius) of the particles and the particle volume and surface–area concentrations, with respective solutions of inversion calculations based on the spectrally resolved lidar data of particle backscatter and extinction coefficients [*Wandinger et al.*, this issue].

In this way, closure studies provide a quality–checked, vertically–resolved aerosol data set that is appropriate to be used in the quantification of the impact of the aerosol distributions on the radiation budget [*Petzold et al., Wendisch et al., Wendling et al.,* this issue].

4. Instrumentation

LACE 98 took place from July 13 to August 12, 1998. The long–term operational Lindenberg Meteorological Observatory (MOL, German Weather Service, 52.2° N, 14.1° E) located about 50 km southeast of the eastern boundaries of Berlin in a typical non–urban landscape was chosen as main experimental site. MOL is a favorable site for two reasons. (1) The observatory is a equipped with a variety of advanced instruments for doing routine, quality–assured observations of meteorological, aerosol optical and radiation parameters. It is one of 27 (four in Europe) worldwide distributed stations of the Baseline Surface Radiation Network (BSRN). (2) Urban (aerosol plume of Berlin) as well as rural–like aerosol conditions for different pattern of air advection (from the ocean and the continent) can be studied at Lindenberg. Furthermore, the German Weather Service supported the project by providing weather forcasts (including personal communication as often as necessary), analysis charts, satellite imagery, detailed backward trajectory calculations, and logistics.

Table 1 lists the participating instituts. Table 2 gives an overview of the experimental activities, the used platforms, and field sites. The ground–based in–situ observations were performed on a grassy plane of MOL at Falkenberg, about 4 km south of Lindenberg. The instrumentation for the characterization of physical and optical particle properties was housed in two air-conditioned containers. The samplers for chemical analyses were placed in a shelter at the base of the sampling stack. A modified high–flow Anderson PM 10 inlet was used for the nephelometer measurements and impactor and filter sampling for mass and chemical analysis. A low–flow Anderson PM 10 inlet was employed for particle number concentration, number size distribution, volatility, hygroscopicity, and light absorption measurements. Both inlets were mounted 10 m above ground level to avoid near–ground aerosol contaminations.

The instruments for characterization of the physical particle properties and of the dependence of particle growth on relative humidity are presented in *Wex et al.* [this issue], *Bundke et al.* [this issue], and *Busch et al.* [this issue]. Special emphasis was put on observations under ambient conditions (horizontally pointing lidar, telephotometry) to allow comparisons with in–situ measurements of the optical properties of dry particles. The instruments and analyses applied in the complex characterization of the chemical properties of single particles and of particle ensembles are described in *Ebert et al., Trimborn et al., Wieser et al.,* and *Neusüß et al.* [this

issue].

Three airplanes carrying a sophisticated aerosol and radiation payload probed the entire troposphere in a complementary and coordinated way [*Wendisch et al., Fiebig et al., Petzold et al., Schröder et al., Wieser et al.,* this issue]. The aircraft were operated from a nearby airport from July 27 to August 12, 1998. The Partenavia P68B (instrumented by *enviscope* GmbH) chartered by the IFT, the Cessna of IWFUB, and the Falcon of DLR conducted a total of 29 flights. Seven vertically coordinated flights involving all three aircraft were made on five different days.

The Partenavia typically flew vertical ascents and descends which included triangular pattern at four to six height levels between 500 and 4000 m over the Falkenberg site. Most of these pattern were in the lowest tropospheric aerosol layer that usually coincides with the boundary layer at daytime. The maximum flight height reached by the Partenavia is 4.5 km [*Wendisch et al.,* this issue]. The Falcon mainly operated in the free troposphere, performing two 60 km straight legs in north–south direction at about six different height levels, one near and the other 20 km east of Lindenberg close to the border to Poland [*Petzold et al., Fiebig et al.,* this issue]. ¿From these two airborne platforms, the entire troposphere was characterized concerning aerosol properties and up– and downwelling solar irradiances. The in–flight coordination was simultaneously directed from the ground via radio operation based on actual lidar aerosol profile and radiosonde temperature and humidity observations.

One of the main objectives of the third aircraft was the daytime– (i.e., solar–zenith–angle–) dependent characterization of the spectral ground reflectance over the field sites (grassy land, fields, forests, and lakes). For this purpose, the Cessna flew up to four flight legs between 500 and 2500 m height over Falkenberg, Lindenberg, and nearby lakes and forests.

Four lidars were deployed at Lindenberg measuring aerosol properties, ozone, and water vapor concentrations. For the first time, three advanced aerosol lidars (two ground–based Raman lidars, one airborne High Spectral Resolution Lidar, HSRL) participated in a large campaign. In contrast to widely used backscatter lidars, advanced systems measure pure molecular backscatter signals and thus allow an unambigeous determination of the volume extinction coefficient of the particles under ambient conditions [*Ansmann et al.,* 1990; *Grund and Eloranta,* 1991]. The lidars measured vertical extinction profiles at up to three wavelengths simultaneously (290 nm, 355 nm, and 532 nm,

[*Wandinger et al.,* this issue]).

Combined lidar–aircraft observations are important for two reasons. (1) They provide a unique opportunity to proof, by cross checking, the quality of the airborne nephelometer observations and the lidar–derived extinction and backscatter profiles. (2) Comparisons with airborne observations of the physical particle properties in dry aerosol layers in the free troposphere are the most appropriate, often the only way to verify the lidar inversion results concerning particle size, and volume and surface–area concentrations [*Müller et al.,* 2000].

5. Meteorological and aerosol optical conditions

A brief overview of the atmospheric conditions is given here in order to facilitate the integration of the findings presented in the following papers into the meteorological context. Unsettled weather conditions prevailed during the four–week LACE 98 period. Low–pressure troughs and high–pressure ridges crossed the field site in quick succession. A stable high pressure system moving slowly from the North Sea to east Europe was only present during the last days of the experimental phase (August, 9–12). Because of the favorable weather conditions and the respective dense set of lidar/aircraft data, we called this phase the 'golden period' of LACE 98.

Table 3 summarizes the general weather and aerosol conditions during LACE 98. Figure 2 shows the time series of the spectral columnar particle optical depth δ_λ for $\lambda \approx 540$ nm observed at Lindenberg with Sun and star photometers and the Ångström exponent $\mathring{a}$. The spectral aerosol optical depth δ_λ is defined as the column–integrated volume extinction coefficient of aerosol at a wavelength λ. The Ångström exponents $\mathring{a}$ [*Ångström,* 1961] in Table3, defined as $\delta_\lambda = \beta\lambda^{\mathring{a}}$ with wavelength λ in micrometer and the aerosol optical depth β for 1 μm, are determined from least–squares fits of the daily mean values of measured δ_λ (in a log–log scale) considering six Sun–photometer wavelengths from 451–855 nm or seven star–photometer wavelengths from 444–863 nm. At this site, aerosol optical depth spectral slopes generally adhere to an Ångström exponential law [*Weller and Leiterer,* 1988].

According to Table 3, in about 75% of all cases the air was advected from westerly directions (northwest to southwest) and the aerosol was of maritime, polar origin (mP, xP). The daytime mean aerosol optical depth at 551nm was typically larger than 0.2 with a mean value of 0.25 and a standard deviation of 0.09. The Ångström exponent $\mathring{a}$ exceeded 1.45 in more than 50%

of the cases which indicates the presence of considerable amounts of anthropogenic particles. For comparison, large particles, e.g., of maritime origin, lead to Ångström exponents below 0.5. The LACE 98 optical properties in Table3 fit well into the general picture given by *Weller et al.* [1998].

In Figure 2, cloud–screened values of the particle optical depth at $\lambda \approx 540$ nm are shown. The optical depth ranged from 0.02 to 0.55 during LACE 98. Significant changes in the optical depth within 12 hours were often observed, probably caused by urban influences. Air mass changes occurred slowly with typical time scales of days. The daytime and nighttime mean Ångström coefficients ranged from 0.8 to 1.7 and from 0.5 to 2.3, respectively. Most values (85%) were found between 1 and 1.7.

¿From Figure 3 it becomes obvious that the daytime– and nighttime–mean Ångström coefficients vary strongly with aerosol optical depth for values $\leq$0.2. Above 0.2, å was between 1.2 and 1.5 with only one exception. *Remer et al.* [1999] found a similar behavior of the Ångström exponent at the east coast of the United States. The values accumulated around 1.5 to 1.6 for particle optical depths larger than 0.2.

Plate 1 shows two examples of height–time distributions of the aerosol in terms of particle backscattering as measured with the IFT lidar at 532–nm wavelength. The two cases shown are studied in detail concerning their radiative properties [*Wendisch et al., Wendling et al,* this issue]. They represent a typically polluted case (August 1) and a clear–air case with very low particle optical depth (August 10).

On August 1, a two–aerosol–layer system was present. Such a layering often occurs over central Europe as our routine lidar measurements at Leipzig, Germany, show. The aerosol optical depth at 551 nm was approximately 0.27 in the morning of August 1 (time steps $<$150 in Plate 1) and the mean Ångström coefficient took values around 1.6. ¿From the profiles of the 180°–backscatter coefficient (scattering coefficient at 180° scattering angle and normalized to one steradian (sr) solid angle, unit: Mm^{-1} sr^{-1}) the optical depth of the lofted layer (above 1200 m height) was estimated to be 0.05–0.1. In this estimation, a ratio of the particle extinction coefficient to the particle backscatter coefficient of 50 (in sr) was assumed which is typical for polluted air [*Ansmann et al.,* 2001; *Wandinger et al.,* this issue].

According to the radiosonde profiles of relative humidity and potential temperature in Figure 4, the boundary layer reached up to 1200 m in the early afternoon at 1100 UTC (1300 local time). The backward trajectories

in Figure 5 indicate that the boundary layer air was advected from the North Sea, and crossed the Berlin area, whereas the aerosol aloft moved over the highly industrialized parts of western and central Europe.

On August 10, a boundary–layer development under cloudfree, low–wind conditions was documented. The particle optical depth of the boundary layer was about 0.05 at 550 nm. The air below 2000 m was advected from the Atlantic Ocean north of Great Britain, crossed southern Norway and Sweden, and the Baltic Sea before arriving at Lindenberg. Northerly winds prevailed at ground on that day. The relative humidity was well below 60% in the boundary layer most of the day, thus the aerosol particles were dry.

A thin, slowly descending aerosol layer was detected between 3 and 4 km on August 10 (particle optical depth of 0.02–0.05). According to backward trajectories this layer originated from forest fires in western Canada [*Wandinger et al.,* this issue; *Forster et al.,* 2001]. The layer was observed with lidar for about 36 hours on August 9-10, and initially extended from 4 to 8 km height. The aircraft probed the long–range–transported aerosol layer several times on these two days. Observations of this biomass–burning aerosol layer are discussed in detail in *Fiebig et al.* [this issue] and *Wandinger et al.,* [this issue].

An interesting feature appearing during the period August 9–12, 1998, was the decoupling of the air mass transport below and above about 500 m. Below 500 m height, the airflow turned from north on August 10 over east to southeast on August 11. Above 500 m height, the air was advected from the polluted western and southwesten parts of Europe on August 11–12. The conditions became increasingly stagnent. High particle extinction coefficients (>100 Mm^{-1} at 532 nm) and high ozone concentrations were found at Lindenberg in the layer between 1000 and 3000 m on August 11–12. The particle optical depth at 532 nm increased from 0.05 (August 10, late evening) to 0.35 (August 11, early morning) according to lidar observations. The column observations on August 11 document heavily polluted conditions.

6. Key findings

This section briefly highlights several key results. The summary may serve as a guide through the LACE 98 papers.

1) Gravimetrically derived mass size distribution, the corresponding size-segregated chemical composition and log-normal parameters of the number/volume distribution have been determined simultaneously for various

maritime and continental air mass types [*Neusüß et al.*, this issue]. Concerning mass closure a linear correlation was found between the gravimetrically determined and the size–distribution–related mass concentrations. However, the latter approach delivered 50% higher mass concentrations for submicrometer particles. These differences are almost independent of aerosol type (maritime or continental particles). A combination of the incomplete information of both physical properties (density, shape), and measurement uncertainties (sizing of the DMA, impactor losses and impactor cut–offs) cause the observed systematic differences in mass concentration.

2) Hygroscopic properties of atmospheric aerosols have been measured simultaneously in the Aitken and accumulation size range with a HTDMA and in the supermicron particle range with the SoFA system [*Busch et al.,* this issue]. The growth factors are consistent with findings regarding the humidity dependence of particle scattering and 180° backscattering derived from optical measurements [*Bundke et al.*, *Wandinger et al.*, this issue].

3) On the basis of a complete characterization (chemical composition, size distribution, thermodynamic phase, particle morphology) of single particles measured between 0.1 and 3 μm particle diameter during LACE 98, an attempt was made to determine the real and imaginary parts of the particle refractive index at 550 nm wavelength as a function of particle size [*Ebert et al.*, this issue]. Average values are presented considering the relative volume concentrations of six particle groups, namely of ammonium sulfates, calcium sulfates, carbonates, metal oxides, sea-salt (including aged sea-salt), silicates, and carbon-rich particles. The calculations are in good agreement with results from optical measurements. Deviations were between 0.01 and 0.04 for the real part and between 0.005 and 0.03 for the imaginary part. According to the chemical analysis the real and imaginary parts of the refractive index varied between 1.51 and 1.58 and 0.03 and 0.06, respectively, for the cases chosen for the comparison. Questions remain to what extend these results are applicable to aerosols at ambient conditions because the probed particles were dried before the analysis and therefore lost some amount of the water and other volatile species.

4) Deviations of the order of $\pm$20% (relative deviation between measured and calculated values) were achieved in closure studies concerning the volume scattering coefficient of the particles. The investigations were based on both ground–based and aircraft observations of measured particle number size distributions, chemical com-

position, and scattering and absorption properties of the particles [*Wex et al.,* this issue].

5) Considerably large discrepancies (30%–170%) were found for the volume absorption coefficients [*Wex et al.,* this issue]. The calculated values were always larger than the measured ones. The calculations are most sensitive to uncertainties in the determined fraction of absorbing material in the aerosol particles and to uncertainties in the measured particle size distribution. The uncertainties in the absorption coefficients propagated into considerable uncertainties (of the order of 0.1) of the derived single scattering albedo (scattering–to–extinction ratio).

6) Comparisons between optical properties deduced from the profiles of the measured aerosol microphysical properties and the particle aerosol optical depth determined with Sun/star photometers and backscatter coefficients measured with lidar yielded deviations of less than 30% [*Petzold et al., Fiebig et al., Wendisch et al.,* this issue].

7) Good agreement was also found in the comparison of lidar–derived microphysical aerosol properties and in–situ measured values [*Wandinger et al.,* this issue]. Deviations range between 10%–30% for the effective radius, and the volume and surface–area concentrations. The multiwavelength lidars allowed an almost complete characterization of aerosol properties needed for the estimation of the climate impact of the tropospheric aerosol distributions.

8) On the basis of the quality–assured lidar–aircraft data ([*Fiebig et al., Petzold et al.,* this issue], the measured humidity growth factors [*Busch et al., Fiebig et al.,* this issue], and determined refractive index characteristics [*Ebert et al., Bundke et al., Wex et al.,* this issue], the relationship between the physical, chemical, and optical properties in the tropospheric column and the downwelling and upwelling irradiances were investigated. Agreement (within the measurement error bars) between measured and calculated *global* solar irradiances were found for ground–based downwelling irradiances and airborne down– and upwelling pyranometer measurements near the surface [*Wendling et al., Wendisch et al.,* this issue]. At higher altitudes the airborne downwelling solar irradiance measurements revealed to be significantly overestimated by respective calculations [*Wendling et al.,* this issue]. Also, significant differences between the measured and calculated solar and spectral *diffuse* and *direct* surface insolations were found. Because the approach was based on a well–defined and consistent input data set, it is concluded that the obvious inconsistencies are linked to funda-

mental problems in understanding of the solar radiative transfer in the atmosphere. The underestimation of atmospheric absorption by the model may have caused part of the discrepancies.

9) The radiative aerosol forcing at the tropopause obtained from the measurements and calculations ranges between - 4 W m^2 for the clean conditions on August 10 and - 16 W m^2 for the polluted situation on August 1 [*Wendling et al.,* this issue]. Typical continental aerosol distributions over Europe mainly cools the Earth/Atmosphere system. *Wendling et al.* [this issue] show the importance of including horizontal surface reflectance inhomogeneities in the radiative transfer calculations.

10) From the aircraft–lidar observations it was found that the single scattering albedo was always larger than 0.9 in the polluted boundary layer and between 0.8 and 0.9 in the biomass–burning aerosol layer in the free troposphere. From the ground–based observations the single scattering albedo was derived to be, on average, 0.8.

Finally, it is worthwhile to mention that, in most cases, we were not able to bring ground–based aerosol data in consistent agreement with aircraft and lidar measurements. Even in cases of a well–mixed boundary layer as on August 10 (afternoon), significant differences in the extensive quantities such as mass concentration, particle number concentration, or particle scattering coefficient were usually found. However, both in–situ measurements at ground as well as the column observations with aircraft and lidar are required as complementary parts of aerosol field experiments. Only at ground overdetermined aerosol data sets can be obtained for performing the fundamental aerosol closure studies. Only the column observations allow a trustworthy quantification of the climatic impact of observed aerosol distribution on the radiation field.

7. Conclusion

A comprehensive aerosol data set for a continental European site is now available. On the basis of complex, complementary observations at ground, with aircraft and remote sensing, the chemical, physical, optical, and radiative properties of typical central European, summertime aerosol distributions are described. These data are a valuable contribution to the global aerosol climatology. They can be used as input parameters in atmospheric models and as ground–truth values in applications of spaceborne aerosol remote sensing over land.

In this sense, LACE 98 contributed to the field of research that deals with the impact of aerosols on climate and the respective modelling approaches. A con-

siderable number of closure studies have been done to investigate the relationships between physico–chemical particle properties and the radiation field and to quantify the radiative impact of the observed aerosol distributions as well as the uncertainties in the observations and determinations. The potential and limits of a variety of new and established aerosol observational and analysis techniques were demonstrated.

Problems in the characterization of the radiative effects of aerosol at ambient conditions mainly arise from the uncertainties in the determined particle absorption properties. The fact that most in–situ measurements are done under low, nonambient humidity conditions causes another source of uncertainty. Radiative closure studies demonstrated that more work is needed in order to improve the knowledge of radiative transfer in the atmosphere. Spaceborne remote sensing over land still remains a challenging task keeping in mind the large (dominant) influence of the surface albedo on the radiation field at short wavelengths.

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Figure 2. Sun photometer (open circles) and star photometer observations (closed circles) of the particle optical depth [*Leiterer et al.,* 1998] at 551 and 533 nm, respectively, and of the Ångström exponent. The Ångström exponent is calculated from optical–depth values averaged over the entire daytime (Sun photometer) and nighttime periods (star photometer). Data affected by clouds are removed.

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Figure 5. 4–day backward trajectories arriving at Lindenberg at 950, 850, 700, and 400 hPa on August 1, 1998 at 0600 UTC.

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Plate 1. Particle backscatter coefficient (scattering coefficient at 180°) at 532–nm wavelength determined from lidar return signals. Rayleigh scattering and atmospheric transmission effects are removed by means of actual radiosonde (air density) and Sun photometer data (particle optical depth). Temporal and vertical resolution is 30 s and 15 m, respectively. The observations were taken on 1 August 1998 between 0549 UTC and 0945 UTC (top) and on 10 August 1998 between 0621 UTC and 1841 UTC (bottom). Data below 700–800–m height are not reliable.

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Table 1. Participating institutions

Institution	
DFD	Deutsches Fernerkundungsdatenzentrum, DLR[1], Neustrelitz
DLR	Institut für Physik der Atmosphäre, DLR, Oberpfaffenhofen
FMTUD	Fachbereich Materialwissenschaften, TU[2] Darmstadt
GSF	Gesellschaft für Strahlenforschung, München
IFT	Institut für Troposphärenforschung, Leipzig
IMGF	Institut für Meteorologie und Geophysik, Universität Frankfurt
IMP	Institut für Mathematik, Universität Potsdam
IMTUD	Institut für Mineralogie, TU Darmstadt
IPCW	Institut für Physikalische Chemie, Universität Würzburg
IPAM	Institut für Physik der Atmosphäre, Universität Mainz
IPH	Institut für Physik, Universität Hohenheim
IPW	Institut für Physik, Universität Wien
IUB	Institut für Umweltphysik, Universität Bremen
IWFUB	Institut für Weltraumwissenschaften, Freie Universität Berlin
MIM	Meteorologisches Institut, Universität München
MOL	Meteorologisches Observatorium Lindenberg, DWD[3]
MOP	Meteorologisches Observatorium Postdam, DWD
MPIC	Max–Planck–Institut für Chemie, Mainz
MPIM	Max–Planck–Institut für Meteorologie, Hamburg

[1]DLR = Deutsches Zentrum für Luft– und Raumfahrt, [2]TU = Technische Universität, [3]DWD = Deutscher Wetterdienst

Table 2. Experimental contributions, field sites, and measurement platforms. Falkenberg (Fal) and Lindenberg (Lin) were the field sites, Falcon (F), Partenavia (P), and Cessna (C) the aircraft involved. d_P denotes the dry particle diameter, δ_λ stands for the particle optical depth, and λ is the wavelength.

Institution	Instrument	Derived quantities	Ground	Aircraft
IFT	Filter + 5–stage impactor:	Total and size resolved: Particle mass,	Fal	
	Thermographic Carbon Analysis (TGCA),	Organic and elemental carbon,		
	Capillary Electrophoresis (CE)	Inorganic and organic ions		
	Twin Differential Mobility Particle Sizer (TDMPS)	Particle number size distribution (d_P:3–800 nm)	Fal	
	Volatile Tandem Differential Mobility Analyzer (VTDMA)	Volatile volume fraction (d_P:15–150 nm)	Fal	
	Aerodynamic Particle Sizer (APS)	Particle number size distribution (d_P:0.8–10 μm)	Fal	
	Condensation Particle Counter (CPC)	Particle number concentration ($d_P > 10$ nm)	Fal	P
	Ultrafine Condensation Particle Counter (UCPC)	Particle number concentration ($d_P > 3$ nm)	Fal	P
	Passive Cavity Aerosol Spectrometer Probe (PCASP–X)	Particle number size distribution (d_P:0.1–10 μm)	Fal	P
	Nephelometer	Particle scattering and back–scatt. coef. (λ=450,550,700 nm)	Fal	P
	Particle/Soot Absorption Photometer (PSAP)	Particle absorption coefficient (λ=565 nm)		P
	Multiwavelength lidar	Particle 180°–backscat. coef. (λ=355,400,532,710,800,1064 nm), Part. extinct. coef. (λ=355,532 nm), Particle volume and surface conc., Particle effective radius, Particle refractive index, Water–vapor mixing ratio	Lin	
	Sun photometer	δ_λ (λ:351–1062 nm)	Lin	
	Precision Spectral Pyranometer (PSP)	Up– and downwelling solar irradiances (λ:0.3–3 μm)		P
	Ultraviolet Radiometer (TUVR)	Up– and downwelling UV irradiances (λ:0.3–0.4 μm)		P
	Spectroradiometer	Global, direct, and diffuse spectral downwelling irradiances (λ:0.5–0.92 μm)	Fal	
MPIC FMTUD IMTUD	5–stage impactor/filter: Total Reflectance X–Ray-Fluorescence (TRXF), CE, High Resolution Scanning Electron Microscope (SEM), Transmission Electron Microscope (TEM), Laser Microscope Mass Analyzer (LAMMA)	Size–resolved particle mass, Morphology of particles, Bulk chemical properties, Chemical composition of single particles, Elemental composition	Fal	F,C
	Nephelometer	Particle scatt. coef. (λ=550 nm)		F
IPCW	Laser Mass Analyzer for Particles in the Airborne State (LAMPAS)	Chemical composition of single particles (aerosol type classification, 5 size ranges, d_P: 0.2–1.5 μm)	Fal	

Table 2. (continued)

Institution	Instrument	Derived quantities	Ground	Aircraft
IPH	5–stage impactor: LAMMA, HRSEM	Chemical composition of single particles (aerosol type classification, 5 size ranges, d_P: 0.15–15 μm)	Fal	P,C
	Optical Particle Counter (OPC)	Particle number size distribution (d_P:0.3–20 μm)		C
GSF	Hygroscopy Tandem DMA (HTDMA)	Hygroscopic growth of particles (d_P: 0.03–0.3 μm)	Fal	
IPAM	Water Soluble Fraction of Large and Giant Aerosol Particles (SoFA)	Water soluble fraction of particles (7 size ranges, d_P: 0.4–4 μm)	Fal	
IMGF	Filter probes + Polar Aerosol Photometer	Visible–spectrum mean value: Particle extinction coefficient, Scattering coefficient, Absorption coefficient, Single scattering albedo, Complex refractive index, Asymmetry parameter of the scattering phase function, Volume soot content	Fal	
	Radiation Balance Photometer	Vector of radiative flux density, Diffusive and reflected radiation	Fal	
IPW	Absorption photometer/ Integrating plate	Particle absorption coefficient (λ=450,550,650 nm)	Fal	
	Telephotometer	Particle extinction coefficient (λ:400–750 nm)	Fal	
	Nephelometer	Particle scatt. coef. (λ=633 nm)	Fal	
IUB	Sun photometer	δ_λ (λ:365–1021 nm), Column particle size distribution	Lin	
MOL	Sun photmeter	δ_λ (λ:380–1054 nm)	Lin	
	Star photometer	δ_λ (λ:390–1045 nm)	Lin	
	Radiosonde (every 3–6 hours)	Relative humidity, temperature, pressure, wind	Lin	
	High–precision radiosonde	Relative humidity	Lin	
	Baseline Surface Radiation Network (BSRN)	Downwelling solar (λ:0.3–4 μm) and longwave irradiances (λ:4–50 μm)	Lin	
MOP	Sun photometer	δ_λ (λ:369–1023 nm)	Fal	C
MIM	Sun photometer	δ_λ (λ:370–1020 nm),	Lin	
	Multiwavelength lidar	Particle 180°–backscat. coef. (λ=355,532,1064 nm), Horizontal–path–integrated particle extinction coefficient (λ=355,532,1064 nm), Particle depolarization ratio	Lin	
MPIM	UV Aerosol/O_3 Differential Absorption Lidar (DIAL)	Particle 180°–backscat. coef. (λ=320,351 nm), Part. extinct. coef. (λ=292,351 nm), Ozon concentration	Lin	
	H_2O DIAL	Particle 180°–backsc. coef. (λ=729 nm),	Lin	

Table 2. (continued)

Institution	Instrument	Derived quantities	Ground	Aircraft
DLR	PCASP–100X	Water vapor concentration, Particle number size distribution (dry, d_P:0.12–3 μm)		F
	Forward Scattering Spectrometer Probe (FSSP–300)	Particle number size distribution (ambient, d_P:0.3–20 μm)		F
	CPC	Particle number concentration ($d_P > 5$ nm)		F
	CPC	Particle number concentration ($d_P > 14$ nm)		F
	Ultrafine Condensational Particle Counter (UCPC)	Particle number concentration ($d_P > 3$ nm)		F
	PCASP–X	Particle size distribution (d_g:0.1–10 μm)		F
	PSAP	Particle absorption coefficient (λ=565 nm)		F
	Multiwavelength lidar	Particle 180°–backscat. coef. (λ=355,532,1064 nm), Particle extinct. coef. (λ=532 nm) Particle depolarization ratio,		F
	PSP	Up and downwelling solar irradiances (λ:0.3–3 μm)		F
	Pyrgeometer	Longwave irradiances (λ:5–50 μm)		F
IWFUB	Compact Airborne Spectrographic Imager (CASI)	Upwelling irradiances, Surface albedo (λ:481–905 nm)		C
	FUB's Integrated Spectrographic System (FUBISS)	Zenith irradiances (λ:600–1000 nm)		C
	Sun photometer	δ_λ (λ:413–862 nm)		C
	Sun photometer	δ_λ (λ:368–1024 nm)	Fal	

Table 3. Airflow, aerosol, and cloud conditions during LACE 98. Traj950 and Traj850 roughly indicate mean direction of horizontal air transport (estimated from backward trajectories) during the last 48 hours before arrival at MOL (0600 UTC) at 950–hPa level (500–m height) and 850–hPa level(1500–m height), respectively. $\delta_{551\mathrm{nm}}$ and å denote the daytime means of the particle optical depth at 551 nm and of the Ångström exponent derived from Sun photometer observations. The air mass types (AMT) were taken from the Berliner Wetterkarte (Freie Universität Berlin, Meteorologisches Institut). Clouds are separated in four classes. The observations were made at the IFT lidar site. The LACE 98 morning (am) and afternoon (pm) flights (Falcon (F), Partenavia (P), Cessna (C)) are given in addition.

Date	Traj950	Traj850	$\delta_{551\mathrm{nm}}$	å	AMT[1]	Clouds	Flights
July 13	SW	SW	–	–	mS	Cu	
July 14	W	W	0.20	1.57	mP	Cu	
July 15	W	W	0.24	1.52	mP	Cu,Ac	
July 16	W	W	0.37	1.46	mP	Cu,Ac	
July 17	SW	SW	0.19	1.26	xPs	Cu,Sc	
July 18	SW	SW	0.24	1.47	mS	Cu,Ci	
July 19	W	W	0.17	1.24	mP	Cu,Ci	
July 20	W	W	0.17	1.17	mM	Ci	
July 21	SW	SW	0.25	1.52	xS	Cu,Ac,Ci	
July 22	W	SW	–	–	mS	Cu,Ac,Ci	
July 23	W	SW	0.35	1.57	mS	Cu,Ci	
July 24	SW	SW	–	–	mPs	Cu,Ci	
July 25	NW	W	0.29	1.56	mP	Cu,Sc,Ac	
July 26	NW	W	0.42	1.68	mP	Cu,Ac,Ci	
July 27	E	E	0.44	1.69	xPs	Cu,Ac,Ci	
July 28	E	SW	0.37	1.62	xPs	Cu,Ac	
July 29	NW	NW	0.36	1.30	xPs	Cu,Sc	
July 30	SW	W	–	–	xPs	Cu,Sc,Ci	
July 31	SW	SW	0.15	1.27	xPs	Cu	am(FPC), pm(P)
August 1	NW	W	0.25	1.51	xPs	Cu,Ci	am(FPC)
August 2	E	SE	0.29	1.57	xPs	Cu,Sc,Ac,Ci	
August 3	NE	SW	0.21	1.45	xPs	Cu,Sc,Ac,Ci	
August 4	E	SW	0.22	1.35	xPs	Cu,Sc,Ac,Ci	
August 5	NW	NW	0.13	0.81	mP	Cu,Sc,Ac	
August 6	NW	NW	0.16	0.85	mP	Cu,Ci	am(PC)
August 7	W	W	0.20	0.84	mS	Cu,Sc,Ac,Ci	pm(F), night(F)
August 8	NW	NW	0.27	1.37	mP	Cu,Ci	pm(C)
August 9	NW	NW	0.19	1.27	mA	Cu,Ac,Ci	pm(FPC), night(F)
August 10	N	N	0.09	1.14	xP		am(FPC), pm(FPC)
August 11	E	W	–	–	cPs	Cu,Ac,Ci	am(P), pm(FPC)
August 12	SE	SW	–	–	cPs	Cu,Ci	am(FPC)

[1]m=maritime, c=continental, x=maritime/continental, A=arctic air, P: subpolar air, Ps=aged and warmed subpolar air, S=subtropical air, M=midlatitudinal (european) air

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