Aerosol states in the free troposphere at northern midlatitudes

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[1] A statistical analysis of in situ observations of aerosols in the midlatitude free troposphere and tropopause region in summer is presented. Vertical profiles of the number density and the associated variability of nuclei mode and larger aerosol particles are discussed. These data confirm that the upper troposphere is an efficient net source region of new particles. Aerosols are studied by analyzing size-segregated data, and it is suggested that observed extreme cases of aerosol evolution include a nucleation state, an accumulation state, and a cloud-processed state. This interpretation implies that aerosol sources (nucleation from the gas phase) are closely tied to aerosol sinks (cloud scavenging of aerosol particles). Processes that could modify the proposed aerosol life cycle are discussed. Number size and surface area distributions typical for the various aerosol states are examined. The dry aerosol surface area ranges from 1 to 20 μm² cm⁻³ in the free troposphere and from 2 to 13 μm² cm⁻³ in the tropopause region. Parameterizations of the aerosol size distributions and total surface area concentrations are provided to facilitate the use of the data in atmospheric models.

INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry


1. Introduction

[2] Atmospheric aerosols occur globally in the troposphere and stratosphere. They are highly variable in terms of abundance, chemical composition, and radiative properties. Aerosols perturb the Earth’s radiation budget due to scattering and absorption of radiation, act as precursors for liquid and ice clouds, serve as sites for heterogeneous chemical reactions, and thus contribute to radiative and climate forcing.

[3] While the importance of aerosols in the climate system is well recognized, significant gaps exist in characterizing and modeling aerosols in the free troposphere (FT) and tropopause (TP) region [Intergovernmental Panel on Climate Change (IPCC), 1996]. Stratospheric sulfuric acid particles, on average, are characterized by a stabilized accumulation mode with a total number of 10 (cm³-air)⁻¹ and a mean diameter near 140 nm [e.g., Junge et al., 1961; Hamill et al., 1997]. In contrast, tropospheric aerosols, especially above the boundary layer (BL), are only poorly defined both chemically and physically, even in terms of integral properties such as the total available surface area. They exhibit a large temporal and spatial variability of number and mass concentrations, partly depending on the origin of the air masses containing the aerosols [e.g., Hobbs, 1993].

[4] Long-term free tropospheric aerosol observations at midlatitudes [Hofmann, 1993] are hardly existent, because most campaigns have focused on case studies in order to study specific processes. In the framework of the International Global Atmospheric Chemistry (IGAC) project, a series of process-related field measurements aims at reducing pending uncertainties in quantifying the atmospheric impact of aerosols [Bates et al., 1998]. Other case studies, which also cover extended data sets and present detailed analyses of chemical composition and volatility of aerosols, do not explicitly report particle size distributions [e.g., Clarke, 1992; Murphy et al., 1998], focus on the formation of new aerosol particles [e.g., Clarke et al., 1998, 1999; de Reus et al., 1998, 1999], or investigated polar latitudes [e.g., Petzold et al., 2000]. Recent work by Raes et al. [2000] discusses the mechanisms determining aerosol size distributions in the global troposphere and puts aerosol processes into the context of atmospheric transport.

[5] In the present work, we present a statistical analysis of airborne aerosol measurements performed during the Lindenberg Aerosol Characterization Experiment (LACE 98) in July/August 1998 over Berlin in the region 13.5°–14.5°W and 51.5°–52.7°N. A description of the overall goal of this field experiment is provided by Ansmann et al. [2002]. Briefly, the main goal of the LACE 98 campaign was to characterize the chemical composition and optical properties of the continental atmospheric aerosol in summer in order to quantify its radiative effects. The main emphasis was clearly placed on the polluted aerosol in the BL, and for closure purposes, aerosols in the free troposphere were also measured. Chemical tracers were not measured.

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A discussion of aerosol properties relevant for radiative forcing during LACE 98 is presented by Petzold et al. [2002] and Fiebig et al. [2002]. Complementing these efforts, we discuss the vertical distribution and the life cycle of the average aerosol as observed in the FT (4–10 km) and TP region (10–12 km). We analyze the aerosol size and surface area distributions and offer an interpretation of the data in terms of freshly nucleated, aged, and cloud-processed aerosol particles as extreme states of the aerosol evolution. Thereby, we put the measurements performed during LACE 98 in a more general context of aerosol behavior.

2. Experiment and Data Analysis

The 10 flights carried out during LACE 98 with the DLR research aircraft Falcon comprise about 17 hours of aerosol sampling within the FT and the TP region on which the analysis of the vertical profiles in section 3.1 is based. Six flight missions (M1–M6), or about 7 hours of sampling, have been selected for the illustration of aerosol states and potential transformation processes discussed in section 3.2. Only flight sequences under clean air conditions were analyzed. Investigations on the processing of aerosols in clouds were beyond the scope of this study. Therefore in-cloud measurements have been removed from the data sets (see below). More detailed information on the flights and respective weather conditions are given by Petzold et al. [2002] and Ansmann et al. [2002].

Constant altitude flight legs were flown during the missions, probing the tropospheric column by 5–7 U-shaped patterns with average leg lengths of about 60 km extending from Lindenberg near Berlin to the Polish border. Westerly air inflow from Canada and the Azores (altitudes above 7 km) and from the British Isles, Iceland, and Greenland (at 4 km altitude) was prevailing during the flights, 72 hour back trajectories are shown by Ansmann et al. [2002]. Sampling has been conducted above the BL in areas outside the flight corridors, so that very fresh contributions from aircraft exhaust plumes and from surface sources can be excluded. Also, we have excluded the observation of an extended forest fire aerosol layer, which moved in during 9 and 10 August [Fiebig et al., 2002]. We refer to section 3.3 for a more detailed discussion.

Aerosol size distributions within the diameter range of 3 nm to 20 μm were deduced by combining measurements of condensation nucleus counters (CNCs, modified TSI type 3025, 3760A) with data from optical spectrometers Passive Aerosol Spectrometer Probe (PMS PCASP-100) and Forward Scattering Spectrometer Probe (PMS FSSP-300) [e.g., Strapp et al., 1992]. Data were recorded at 1 Hz frequency.

The CNCs operated at lower size detection limits of nominally 3, 5, and 14 nm, required air samples (flow ~20 cm³ s⁻¹ each) directed to the pressurized aircraft cabin through a back-facing interstitial inlet [Schroöder and Ström, 1997]. Active sampling resulted in about 30–80 K readings, assigning the resulting values to the corresponding size intervals. We also derived the concentrations of ultrafine condensation nuclei (UCN), n₃–n₁₄ (or n₅–n₁₄ when n₃ measurements were not available), and the UCN ratio, UCN/n₁₄. UCN and the UCN ratio serve as indicators for new particle formation and growth [e.g., Clarke, 1992]. The FSSP-300 data were basically treated as described by Schroöder et al. [2000]. However, in the context of the present work, the FSSP was only used as a cloud sensor in order to remove in-cloud data from the complete data set. Data were removed whenever the integral FSSP concentrations exceeded 2–3 cm⁻³.

3. Results and Discussion

3.1. Vertical Distributions

Figure 1 illustrates the vertical profiles and the variability ranges of n₅, n₁₄, and UCN particle concentrations and the UCN ratio. The thin lines depict percentiles counting 5%, 25%, 50% (the median values), 75% and 95% of the particles, and the thick lines represent the arithmetic mean concentration values. The position of the thermal TP was determined within a few 100 m uncertainty for all individual flight missions by using the Falcon temperature measurements and data from nearby radiosondes (relative humidity was not measured in situ). The horizontal dotted lines in each figure mark the upper boundary of the continental BL, where arithmetic mean and median concen-
Concentrations are also displayed. Up to about 5 km altitude, the concentrations should be read as a function of the pressure altitude. Above 5 km, all curves refer to altitudes relative to the TP. All concentrations are given at a pressure of 1013 hPa and a temperature of 293 K.

The $n_5$, $n_{14}$, and UCN profiles exhibit typical, Z-shaped structures. The exponential decline from the BL into the FT is well-known [e.g., Jaenicke, 1992] and can be attributed to aerosol sources at the Earth’s surface. In the absence of further aerosol sinks or sources, we would expect a continuous decrease of particle concentrations with altitude. However, the distinct local concentration minima at 4–5 km altitude seen in the integral and Aitken profiles combined with the appearance of secondary concentration maxima about 1–2 km below the TP seen in the integral and UCN profiles shows that the upper troposphere is effectively decoupled from the surface. While the minimum likely results from scavenging of aerosols by lower tropospheric clouds, the maximum close to the TP is most likely caused by new particle formation processes, as we argue below. Above the TP, the concentrations rapidly decline again. Note, however, that concentration levels in the lowermost stratosphere exceed those typically observed in the lower stratosphere higher up (not shown), underlining the notion that the lowermost stratosphere is a mixing region exhibiting both tropospheric and stratospheric character.

Figure 1 shows a four-fold increase of the average integral aerosol number concentrations, from about 750 cm$^{-3}$ in the lower FT to about 3000 cm$^{-3}$ in the upper FT. This increase with altitude is strongest for the smallest particles: the arithmetic mean (median) UCN concentrations raise from 100 (20) cm$^{-3}$ to 2000 (1000) cm$^{-3}$ by a factor of 20 (50), while the larger Aitken particles just double their concentrations, from 600 to 1200 cm$^{-3}$. The percentiles of the UCN ratio illustrate that the mean particle sizes shift toward smaller values with increasing altitude. The median and arithmetic mean UCN ratios (0.05 and 0.1) are small near the minimum in the lower troposphere, but reach 2 about 2 km below the TP, which means that half of the total aerosol number is made up by nuclei mode particles. These
observations confirm quantitatively that the upper troposphere and TP region provides an efficient net source (at least by number) of new aerosol particles.

[17] Nucleation events in the atmosphere predominantly affect UCN concentrations and UCN ratios, while the larger particles in the Aitken size range are much less affected. Such nucleation events (bursts) are responsible for the large differences between arithmetic mean and median values seen in the two lower panels in Figure 1. In particular, the average UCN and UCN ratio profiles exceed the 75% percentile profiles throughout large regions of the FT. In contrast, arithmetic means and medians (the 50% percentile profiles) of the larger Aitken particles deviate much less from each other. This is consistent with very high concentrations appearing in the nucleation mode. Similar measurements have been interpreted in detail by Schroeder and Strom [1997].

[18] At the TP, the differences between median and arithmetic mean profiles as well as the overall variability, indicated by the differences between the 5% and 95% percentiles, reach their smallest values. This indicates a decreasing variance of the width of the particle size spectra. The reduced particle formation activity reflects the decreasing concentrations of water vapor and other aerosol precursor gases across the TP. Because the atmospheric residence times increase markedly with altitude in the TP region, a more aged aerosol develops there compared to the rest of the FT.

[19] From the flown constant altitude legs, there is also information available on the horizontal variability of the UCN and Aitken mode aerosols. In the upper FT at 7.3 to 7.8 km altitude (about 4 to 5 km below the TP) the UCN variability ranges from 15% to 60%, while the corresponding variability of the Aitken mode aerosol is 46% to 15%. There is no significant correlation observable between lateral variabilities of nucleation mode and Aitken mode. The situation might be different at altitudes closer to the TP where the production rate of new particles reaches its maximum value, see Figure 1.

3.2. Aerosol States

[20] We arrange the cloud-free aerosol data for the FT in a scatterplot (Figure 2a), showing ambient number densities of aerosol particles above \( D > 100 \) nm (rouly proportional to the total aerosol surface area) versus those between 3 and 100 nm (an indicator for recent new particle production). The state MED represents the typical background aerosol. Possible limitations of this interpretation are discussed in the text.

Figure 2. a: Scatterplot of free tropospheric aerosol particle concentrations (ambient values) taken between 4 and 10 km altitude under clear sky conditions over central Europe in summer. Number densities of particles with \( D > 100 \) nm (roughly proportional to the total aerosol surface area) versus those between 3 and 100 nm (an indicator for recent new particle production). b: Schematic illustrating the proposed map of likely states of tropospheric aerosols: NUC: fresh, nucleated state, ACC: aged, accumulated state, SCA: cloud-processed state. The state MED represents the typical background aerosol. Further, a most typical state is defined (circle in Figure 2b), which is described in more detail in the text.

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3.2. Aerosol States

[20] We arrange the cloud-free aerosol data for the FT in a scatterplot (Figure 2a), showing ambient number densities of aerosol particles above \( (n_{100}) \) versus below \( (n_3 - n_{100}) \) 100 nm diameter. The former is a crude but nevertheless applicable measure for the total aerosol surface area density, which is usually dominated by aerosols in the accumulation mode containing "aged" particles; see also Figure 3 later in this paper. The latter is a measure for "fresh" particles in the ultrafine and Aitken (hereafter nucleation) size range that have recently formed and grow into the accumulation mode size range. We presume that adjacent data points in Figure 2a represent comparable microphysical states of the aerosol, although the data were not necessarily obtained at the same time and location. We preferred to relate number densities of particles instead of surface densities, since for the calculation of surface densities of the ambient aerosol from particle size distributions measured in the dry state, additional information on ambient relative humidity and humidity growth factors is needed. This information was not available in the required high temporal and spatial resolution, so we restricted the data analysis to the more simplified but more robust approach of relating number densities.

[21] Later we will examine extreme states of the aerosol evolution. These are shown as boxes in Figure 2b. Each box comprises 100–300 single measurements, corresponding to several percent of the data or about 5 min measuring time. This ensures that, at the same time, these states are well defined and that the PCASP measurements (especially from the large size channels) are statistically robust. Further, a most typical state is defined (circle in Figure 2b), which
comprises 1% of all data points that lie within ±5% of the statistical median values of \( n_3 - n_{100} \) and \( n_{100} \).

[22] Interestingly, the vast majority of data points in Figure 2a form a triangle in this specific graphical representation. The fresh aerosol concentrations show the largest variability (over two orders of magnitude), but only at the lowest concentrations of aged particles, that is, at the base of the triangle. When \( n_{100} \) increases, this variability decreases, forming the two sides of the triangle. The data points within the triangle very likely result from transitions between the extreme states at the edges, and from the variability of environmental conditions (temperature, humidity, concentration of condensable gases, cloud occurrence frequency, among others) encountered during the measurements.

[23] We offer an explanation of these measurements with the help of the schematic depicted in Figure 2b. Box NUC comprises typical concentrations \( n_3 - n_{100} \approx 2500 \text{ cm}^{-3} \) of particles from the nucleation state. These are not yet affected by larger, preexisting particles. The low available aerosol surface area paves the way for new particle formation from the vapor phase. Box ACC represents a fully developed, accumulated aerosol state with \( n_{100} \approx 200 \text{ cm}^{-3} \). Such aged particles result from the collection of part of the smaller, fresh particles, convective transport of aerosols from surface sources and perhaps from condensational growth of NUC particles, following the path NUC → ACC. The presence of large surface area concentrations of aged particles suppresses new particle formation. Finally, box SCA represents an aerosol state with number densities of only \( n_{100} \approx 20 \text{ cm}^{-3} \), presumably reduced via scavenging by large (μm-sized) cloud particles. The circle MED encompasses median concentrations \( n_{100} \approx 60 \text{ cm}^{-3} \) and \( n_3 - n_{100} \approx 300 \text{ cm}^{-3} \). In this state, which was mostly present during the measurements, the aerosol is subject to various degrees of aging or cloud processes. We can think of other processes that could alter the explanation given above (see Figure 2b), including tropopause folds inducing nucleation by mixing of air parcels, long-range transport of well-aged air devoid of aerosol precursor species, or air masses containing particles produced by aircraft.

[24] The scheme presented in Figures 2a and 2b does of course not exclude the existence of specific aerosol states outside the triangle under certain conditions. On the other hand, we propose that the triangle bounded by the extreme states is representing the majority of existing states of the upper FT aerosol.

3.3. Representativeness of the Data Set

[25] The question arises whether the data points in Figure 2a are truly independent in a statistical sense. To mention one example, data points located in the state SCA may not be uniquely related to cloud processing, as we have conjectured, but might be produced by a stratospheric intrusion. Table 1 summarizes the frequencies of occurrence of the extreme aerosol states NUC, ACC, and SCA for each analyzed flight with respect to the total set of clear-sky data points. Only the extreme aerosol states were considered in Table 1 because they form the enveloping triangle, while the state MED is representative for almost all aerosol states corresponding to the inner area of the triangle. Obviously, all extreme states were observed during each flight, but to various extent. A high observation frequency of SCA was always connected to the presence of clouds (flights M3, M4 [Petzold et al., 2002]), while under clear sky conditions the ACC state was observed more frequently. Nucleation events occurred with the lowest frequency, except during the flight M4. However, the data presented in Table 1 do not fit into a general pattern which connects the proposed extreme aerosol states to each other in a direct way. To give an example, a transition from NUC to ACC rather follows a path through the MED state instead of a direct transition along the triangle boundary.

[26] Since LACE 98 was designed as an aerosol column closure experiment, no Lagrangian experiments were conducted during the campaign. (Recall the basic objectives of LACE 98 as summarized by Ansman et al. [2002].) But this type of experiment would be the only one to establish direct connections between extreme aerosol states in a sense of process studies.

[27] On the other hand, the analyzed flights were conducted during a three-week period at the same place probing the atmospheric column from the TP down to the BL. In terms of aerosol optical depth, which is a measure of the atmospheric aerosol burden, the LACE 98 period has turned out to be representative for a midlatitude continental site during summer [Petzold et al., 2002]. During the intense LACE 98 period there was no major convection activity in the Lindenberg area, which excludes a strong impact of fresh emissions from nearby ground sources on the FT. Air masses in the FT were transported by westerly flow either from SW Europe (M1, M2) or across the North Sea (M4–M6), so that both cases of continental and marine influence on the air masses are represented in the data set. In this sense, the data give a good example of the variability of aerosol properties at a continental site.

3.4. Size and Surface Area Distributions

[28] The upper row in Figure 3 shows the number size distributions for the median (MED) and extreme (NUC, ACC, SCA) states as observed in the FT (3a) and the TP region (3b). Particles in the coarse mode \( (D > 1 \text{ μm}) \) are not discussed further, because their concentrations are generally too low to affect the total surface area density. As described above, each spectrum is an average over several 100 single measurements.

[29] Comparing Figures 3a and 3b reveal further interesting details. In particular, the spectrum of aged particles from the ACC state near 100 nm appears to be narrower in the TP region than in the FT, accompanied by an ultrafine mode containing much fewer particles (box ACC in Figure 2b shifts to the left). In the TP region, fresh particles seem to be more abundant in the MED and SCA states than in the FT.

Table 1. Contributions (%) of Individual Missions With Flight Altitudes 4–10 km During LACE 98 to the Aerosol Stages in Figure 2

<table>
<thead>
<tr>
<th></th>
<th>NUC</th>
<th>ACC</th>
<th>SCA</th>
</tr>
</thead>
<tbody>
<tr>
<td>M1</td>
<td>1.4</td>
<td>12.0</td>
<td>2.4</td>
</tr>
<tr>
<td>M2</td>
<td>5.3</td>
<td>17.4</td>
<td>13.1</td>
</tr>
<tr>
<td>M3</td>
<td>0.3</td>
<td>2.6</td>
<td>53.3</td>
</tr>
<tr>
<td>M4</td>
<td>43.6</td>
<td>2.9</td>
<td>15.6</td>
</tr>
<tr>
<td>M5</td>
<td>0.7</td>
<td>21.0</td>
<td>0.7</td>
</tr>
<tr>
<td>M6</td>
<td>0.2</td>
<td>1.0</td>
<td>2.1</td>
</tr>
</tbody>
</table>
In the MED case, this indicates that new particle formation rates maximize in the midlatitude TP region [Hofmann, 1993], as also observed in the tropics and extratropics [Brock et al., 1995]. In the SCA case, this may indicate that in the TP region, the scavenging timescales of small particles by clouds are longer (presumably due to lower cloud surface areas) and/or that new particle formation sets in earlier after cloud processing than in the FT. Finally, NUC and SCA distributions are remarkably similar above 100 nm and differ mainly in terms of total number density.

The lower row in Figure 3 displays the spectral distributions of the specific aerosol surface area corresponding to the number spectra. In terms of surface area, the nucleation modes are much less pronounced in the states MED and ACC where the accumulation modes dominate the overall spectrum.

[30] The lower row in Figure 3 displays the spectral distributions of the specific aerosol surface area corresponding to the number spectra. In terms of surface area, the nucleation modes are much less pronounced in the states MED and ACC where the accumulation modes dominate the overall spectrum.

[31] We note that the spectral measurements have an important limitation: the CNC data do not give a size resolution and we cannot rule out the existence of two aerosol modes between 30 and 100 nm. (The lines in Figure 3 connecting the few data points there have only been drawn to guide the eye.) This lack of size resolution in the Aitken range has also been a problem in previous measurements [e.g., Hofmann, 1993], and may lead to uncertainties in derived aerosol parameters when only one mode is fitted as the size distribution below 100 nm.

3.5. Parameterizations

[32] To facilitate the use of our data in models, we have parameterized the spectra from Figures 3a and 3b in the form of bimodal lognormal size distributions. Lognormal modes have been chosen because they allow sufficiently accurate fits and have convenient analytic properties. Each function is
Table 2. Summary of Aerosol Parameters Fitted to Observed Size Distributions

<table>
<thead>
<tr>
<th>State</th>
<th>MED</th>
<th>NUC</th>
<th>ACC</th>
<th>SCA</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_1$, cm$^{-3}$</td>
<td>140</td>
<td>2500</td>
<td>200</td>
<td>60</td>
</tr>
<tr>
<td>$D_1$, nm</td>
<td>18</td>
<td>9</td>
<td>15</td>
<td>9</td>
</tr>
<tr>
<td>$\sigma_1$</td>
<td>1.7</td>
<td>1.7</td>
<td>2</td>
<td>1.7</td>
</tr>
<tr>
<td>$n_2$, cm$^{-3}$</td>
<td>220</td>
<td>60</td>
<td>300</td>
<td>60</td>
</tr>
<tr>
<td>$D_2$, nm</td>
<td>75</td>
<td>55</td>
<td>120</td>
<td>55</td>
</tr>
<tr>
<td>$\sigma_2$</td>
<td>1.8</td>
<td>1.85</td>
<td>1.6</td>
<td>1.85</td>
</tr>
<tr>
<td>$A$, µm$^2$ cm$^{-3}$</td>
<td>7.5 (6.6)</td>
<td>2.3 (2.9)</td>
<td>22.3 (20.1)</td>
<td>1.3 (1.2)</td>
</tr>
</tbody>
</table>

Lognormal nucleation (subscript 1) and accumulation (2) modes are fitted to the CNC data and PCSAP dry particle spectra ($D < 1$ µm) for each state MED, NUC, ACC, SCA, as defined in Figure 2b. Values $D_1$ and $\sigma_1$ are uncertain due to the poor spectral resolution below 100 nm (see Figure 3); we use educated guesses. Total ambient surface area densities $A$ are computed from the fitted spectra and directly inferred from the observations (in brackets). Measurement uncertainty for $A$ estimated to be ±30%, except for the NUC states and for state SCA in the tropopause region, where it could be larger.

characterized by a total (ambient) number density $n$, a geometric-mean number diameter $D$, and a geometric standard deviation $\sigma$ [e.g., Seinfeld and Pandis, 1998]. These parameters have been fitted to match the observed number spectra and total surface areas $A$ for $D < 1$ µm within the overall experimental uncertainty (±30%) and are summarized in Table 2.

The bulk of accumulation mode particles (subscript 2) can be described rather accurately with one mode between 65 and 110 nm. This mode dominates the total surface area, except for the cases FT-NUC, TP-NUC, and TP-SCA; see Figures 3c and 3d. Concerning the nucleation mode (subscript 1), the parameterizations extend the fits to the regime below 100 nm, where the CNC data do not yield a sufficiently fine size resolution. This small particle mode is constrained in terms of $n$ in cases where it dominates the total number (FT-NUC, TP-NUC, TP-SCA, TP-MED). In all other cases it must be considered highly uncertain; see also the discussion at the end of section 3.4.

In the FT, the ambient specific surface area $A$ of the most representative state MED is ~7 µm$^2$ cm$^{-3}$, in good agreement with earlier measurements [Schröder and Ström, 1997]. As extreme cases, the SCA and ACC states are characterized by minimum and maximum values of ~1 µm$^2$ cm$^{-3}$ and ~20 µm$^2$ cm$^{-3}$, respectively. The high number of small particles in the NUC state raises $A$ slightly above the value for state SCA. In the TP region, $A$ ranges from ~2 to 13 µm$^2$ cm$^{-3}$, with $A$(MED) ~ 5 µm$^2$ cm$^{-3}$, which is along the lines of a theoretical study exploring particle properties at the northern midlatitude TP [Kärcher and Solomon, 1999]. The corresponding range of the total particle number density ($n_1 + n_2$) is in agreement with the observed values reported by de Reus et al. [1998].

Comparing the ratios $A$(SCA)/$A$(MED), we infer that clouds less effectively scavenge aerosols in the TP region (1.8/5.1 = 0.35) than in the FT (1.2/6.6 = 0.18); recall the discussion of Figure 3. While nucleation typically produces detectable particle numbers in the range 1000–3000 cm$^{-3}$, the lowest particle numbers are typically found after cloud scavenging, if our interpretation of the data shown in Figure 2 is correct.

For an altitude-resolving survey of aerosol microphysical properties obtained during the column closure experiment we refer to Petzold et al. [2002], where a set of parameterized tropospheric aerosol distributions is presented for 5 individual missions during LACE 98. Those represent size-segregated, average aerosol budgets relevant to radiative forcing and cannot necessarily be compared to the typical median state MED as reported here.

4. Concluding Remarks

4.1. Summary

We attempted to contribute to a better understanding of the free tropospheric aerosol—a severely undersampled portion of the atmospheric aerosol—by examining summer-time airborne measurements conducted over Central Europe. We discussed vertical profiles, deduced the size-resolved and integral number and specific surface area densities, and explored the variability of these quantities in the free troposphere and the tropopause region.

After having documented the physical properties of the aerosol, we have conjectured regarding possible processes causing the observed properties. By addressing nucleation, growth, and cloud processing, we suggest that our observations may represent the background aerosol state as well as extreme states describing the life cycle of particles. Our interpretation supports the few previous measurements and model calculations suggesting that the upper troposphere is a region where the formation of aerosol particles from the gas phase occurs at times. Further, we have offered a consistent explanation of how the presence of clouds reduces the accumulation aerosol reservoir and illustrated how the disappearance of clouds paves the way for new particle formation. Together, the measurements support the notion that the life cycles of aerosols and clouds are strongly linked.

By parameterizing the size distributions related to typical and extreme microphysical states, we provided a data set of particle properties that can be used in models simulating the aerosol dynamics in the troposphere. For example, our data should aid in improving aerosol modules employed in large-scale models required to quantify the role of aerosols in the climate system and to study the link between aerosols and clouds.

4.2. Outlook

We presented a plausible explanation of the data in terms of a typical free tropospheric aerosol life cycle. However, due to the lack of experimental information regarding in particular the photochemical properties of the sampled air masses, we cannot completely rule out that...
some of the postulated, extreme aerosol states were dominated by a few untypical events, although we believe that the likelihood for such a statistical aberration is small. This belief partly builds on observations from other field campaigns that show quite similar features and can be interpreted accordingly [Schröder, 2000].

[41] For these reasons, some conjectures regarding the aerosol life cycle remain unsupported by evidence, and this work cannot necessarily be put into the same class in a statistical or climatological sense as the repetitive, 20-year aerosol profile database produced by Hofmann [1993]. For example, those data show a very distinct seasonal cycle in aerosol properties, while our measurements cannot easily be placed in a seasonal or hemispheric context.

[42] To create a truly climatological, global database of aerosol properties, more measurements at different locations and seasons are needed. In each campaign, concentrations of chemical tracers such as ozone, CO, NO, and NOx, and relative humidity should be recorded simultaneously with the aerosol data to be able to trace back the origin of the probed air masses. Better resolved particle measurements in the Aiken range are needed, for example by using several CNCS with different lower cut-off sizes operating in parallel [Brock et al., 2000], to study the ultrafine particle size distribution in more detail. Complimentary field studies are necessary which — besides the microphysical aspects as examined here — address the chemical composition and the radiative properties of aerosols.

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