Influence of clouds on aerosol particle number concentrations in the upper troposphere

A. Weigelt,1 M. Hermann,1 P. F. J. van Velthoven,2 C. A. M. Brenninkmeijer,3 G. Schlaf,3 A. Zahn,4 and A. Wiedensohler1

Received 10 January 2008; revised 28 August 2008; accepted 22 October 2008; published 7 January 2009.

[1] This work investigates statistically the influence of clouds on aerosol particles in the upper troposphere, from the tropics to midlatitudes. More than 7500 particle concentration values, collected with the Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container, were analyzed for the occurrence of cloud contacts using backward trajectories and satellite images from the International Satellite Cloud Climatology Project. The majority of high-altitude clouds over the Arabian Sea most likely contributed to the formation of new particles. At the contact, most clouds act as sink for Aitken and accumulation mode particles. However, as new particles grow these clouds act as indirect source for Aitken mode particles. A similar behavior was observed for clouds over the Caribbean. In contrast, no clear influence on nucleation mode particles was observed for most cloud contacts over the Middle East, southeastern Europe, and the midlatitude North Atlantic Ocean. However, in all three regions, the clouds act on average as sink for Aitken and accumulation mode particles. An analysis investigating the influence of the cloud contact duration showed that cloud contacts lasting 4 h or longer resulted in significantly higher number concentrations of nucleation and Aitken/accumulation mode particles than did short cloud contacts (1 h). This effect was observed over the Caribbean and southeastern Europe. Over the Arabian Sea, the Middle East, and the midlatitude North Atlantic Ocean, however, the duration of the cloud contact had no effect; it was only important whether a cloud contact had occurred.


1. Introduction

[2] Atmospheric aerosol particles affect the Earth’s climate by their direct influence on the radiation budget and their indirect influence on cloud processes [Twomey, 1977; Albrecht, 1989; Charlson and Heintzenberg, 1995; Brenguier et al., 2000]. However, current knowledge of atmospheric aerosol processes and aerosol life cycles is inadequate for the realistic modeling of global aerosol fields [Textor et al., 2007]. In particular, there is a lack of understanding of the effect aerosol particles have on cloud properties such as precipitation and cloud lifetime; thus, these properties are poorly represented in climate models [Ramanathan et al., 2001; Chen and Penner, 2005; Lohmann and Feichter, 2005; Intergovernmental Panel on Climate Change, 2007]. Aerosol particles do not only influence cloud microphysics, but inversely clouds also influence the particle number concentration and their chemical composition. Thus, the effect of aerosol particles on the cloud microphysics indirectly affects the particle transport and removal inside clouds, so that particles influence their own concentration and chemistry in the outflow regions of clouds [Cui and Carslaw, 2006]. In the free troposphere, clouds can create new particles through local homogeneous nucleation, can vertically transport aerosol particles from the boundary layer, or can be a sink for particles [e.g., Perry and Hobbs, 1994; Zhang et al., 1998; Ström et al., 1999; de Reus et al., 2001; Weber et al., 2001, Ekman et al., 2006; Kanawade and Tripathi, 2006]. These processes have particular importance in the upper troposphere (UT), where sources of primary particles (e.g., aircraft emissions) are rare. UT aerosol particles are important because of their influence on heterogeneous chemistry [e.g., Bell et al., 2005; Sövde et al., 2007] and on cirrus clouds, thereby affecting the radiation budget [Kärcher, 2003]. However, in the literature there are only a few experimental studies which investigated the influence of clouds on aerosol particles [e.g., Kleinman and Daum,
Two previous case studies have been performed to investigate the influence of clouds on particle number concentrations [de Reus et al., 2001; Kanawade and Tripathi, 2006]. These studies used aerosol and chemistry data from the INDOEX (Indian Ocean) and the TOPSE (North America) campaigns, and included backward trajectories of air parcels and satellite cloud data. The temperatures along the backward trajectories were compared with the cloud top temperatures obtained from satellite data; these results were used to determine the influence of clouds within the 48 h period prior to sampling. Both studies found an increase in the number of nucleation mode particles (in both cases defined as particles with diameters \(d_p\) below \(\leq 10\) nm) whenever there were cloud contacts within the prior 2 days. This increase could not be explained by vertical particle transport alone; hence, the authors concluded that local homogeneous particle nucleation was occurring in the outflow regions of clouds. Other analyses using cloud-resolving models also found that in the outflow regions of deep convective clouds homogeneous particle nucleation can occur [Zhang et al., 1998; Ekman et al., 2006]. These previous individual case studies were limited to small areas and short time periods. Until now there has not been a large enough data set available to allow a statistical analysis for the influence of clouds on the particle number concentration in the UT. This need has been partially met by the CARIBIC project (Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container, http://www.caribic-atmospheric.com). In CARIBIC, a passenger aircraft is used to acquire in situ and retrospective laboratory measurements of trace gases and aerosol particles in the UT and lowermost stratosphere (LMS) [Brenninkmeijer et al., 1999, 2007].

Data obtained from two CARIBIC flight routes were analyzed in the present study, the “Indian route” between Germany and Sri Lanka/Maldives in the Indian Ocean, and the “Caribbean route” between Germany and the Caribbean crossing the North Atlantic Ocean (Figure 1). Data were obtained mainly in the altitude range between 9 and 11 km, and comprised number concentrations for two different particle size ranges. Five-day backward trajectories were calculated every 3 min along the flight tracks to analyze the history of the measured air parcel. Using satellite images, each of these trajectories was examined for evidence of cloud contact within the past 48 h.

Altogether, more than 7500 averaged particle number concentration data points were measured on the two flight routes, covering a time period of more than 4 years. Thus, for the first time it became possible to perform a statistical analysis of the influence of clouds on the particle number concentration in the UT.

2. Data Set and Analytical Approach

2.1. CARIBIC Aerosol Data

The goal of the CARIBIC project is to acquire regular, detailed, and long-term measurements of trace gases and aerosol particles at flight altitudes encompassing the region of the UT/LMS (mainly 9–11 km [cf. Hermann et al., 2003]). To realize this goal, measurement instruments are installed in a standard air freight container, which is mounted and flown in the cargo bay of a long-range passenger aircraft once per month. During the first phase
of CARIBIC (CARIBIC-1, 1997–2002), an LTU International Airways Boeing 767–300ER served as the aircraft platform [Brenninkmeijer et al., 1999]. CARIBIC-2 started in November 2004 and uses a Lufthansa Airbus A340–600 carrying a newly developed container with the scientific instrumentation [Brenninkmeijer et al., 2007]. It is anticipated that CARIBIC-2 will operate at least until 2014. The present study uses measurements from CARIBIC-1, taken between 1997 and 2001 on the Indian and Caribbean route (Figure 1).

In CARIBIC-1, the number concentrations of nucleation mode particles (here 4 nm ≤ dp ≤ 12 nm, N4–12), and the number concentrations of particles with diameter larger than 12 nm (N12), were measured using modified Condensation Particle Counters (CPC, TSI models 7610) with different lower detection diameters [Hermann and Wiedensohler, 2001]. Normally, in the UT, the number concentration of Aitken mode particles (~10 nm ≤ dp ≤ ~100 nm) strongly dominates the number concentration of accumulation mode particles (~100 nm ≤ dp ≤ ~1000 nm) [Clarke and Kapustin, 2002; Heintzenberg et al., 2002]. Hence, the number concentration N12 can be considered to represent mainly Aitken mode particles. All raw particle concentrations were corrected for particle losses in the inlet and in the sampling line, for pressure-dependent CPC flow rates, for CPC counting efficiencies, and for coincidence in and in the sampling line, for pressure-dependent CPC flow rates.

Individual borders for the different regions are listed in Table 1. Measurements beyond these borders were disregarded to create a clear differentiation between the regions and to ensure that the results would be distinct. Since the influence of clouds was of primary interest, the position of the last cloud contact along the analyzed backward trajectory (compare sections 2.2 and 2.3) was used to attribute the measured values to the corresponding region. If no cloud contact occurred within the last 48 h, the actual measurement location was taken for the differentiation.

2.2. Backward Trajectories

Five-day backward trajectories were calculated along the CARIBIC flight tracks using the TRAJKS trajectory model from the Royal Netherlands Meteorological Institute (KNMI, http://www.knmi.nl/samenw/campaign_support/CARIBIC/). The input parameters for these calculations were taken “first guess” wind fields (6 h forecast, available only until September 2000), and from analyzed fields (after September 2000); both from the European Centre for Medium-Range Weather Forecasts (ECMWF) hybrid sigma-pressure model. The wind fields had a spatial resolution of 1° and a temporal resolution of 6 h. To determine the field values at each trajectory node, the model linearly interpolates the wind speed and direction to the node position (latitude and longitude), uses a logarithmic interpolation for the pressure, and a quadratic interpolation in time. The integration time step, and therefore the temporal resolution of the trajectories, was 1 h.

Errors in the trajectory can become large when going back in time several days, especially when encountering (convective) clouds or frontal systems. These errors are due to the assumptions in the trajectory models, and can also be due to the associated assumptions and errors in the input data derived from meteorological model calculations [cf. Scheele et al., 1996; Stohl, 1998; Stohl et al., 2001]. Because of these uncertainties in long-duration trajectories, we analyzed each backward trajectory only for the 2 days preceding the measurements, and stopped when the air parcel had its first cloud contact.

2.3. Analyses of Cloud Properties Using Satellite Data

The presence of clouds was verified using a data product provided by the International Satellite Cloud Climatology Project (ISCCP) (http://isccp.giss.nasa.gov/). This data set provides nearly global coverage in a common data format, with measurements taken by radiometers on the geostationary GOES and METEOSAT satellites, as well as the polar-orbiting NOAA satellites. The chosen data product (ISCCP-DX) includes data on the Cloud Top Temperature (CTT) and the Cloud Top Height (CTH) in pressure units, which could be directly compared with the trajectory height. Other cloud analysis products (e.g., from EUMETSAT or NOAA) offer only the CTT. Unfortunately, the ISCCP-DX product provides only at daytime a differentiation of cloud types. In order to have a uniform analysis, we did not use the cloud type information for the whole data set. The horizontal resolution of the data is 30 km in the tropical region and about 40 km at midlatitudes. The temporal resolution is 3 h. Data are available from July 1982 until June 2007, and can be downloaded from an ftp server ftp://eclipse.ncdc.noaa.gov/pub/isccp/dx/, or ordered from the

### Table 1. Latitudinal Borders of the Analyzed Regionsa

<table>
<thead>
<tr>
<th>Region</th>
<th>Arabian Sea</th>
<th>Middle East</th>
<th>Southeastern Europe</th>
<th>Caribbean Region</th>
<th>Midlatitude North Atlantic Ocean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Latitudinal Borders</td>
<td>&lt;22°N</td>
<td>28°–38°N</td>
<td>&gt;43°N</td>
<td>15°–30°N</td>
<td>&gt;43°N</td>
</tr>
</tbody>
</table>

*aIn order to derive more distinct and representative results, data collected in the intermediate regions were not analyzed.*

Aside from the aerosol particles, also trace gases such as ozone and carbon monoxide were measured within CARIBIC-1. Distortion is avoided by excluding the stratospheric samples using an ozone tropopause threshold value given by Zahn and Brenninkmeijer [2003]. Detailed information on the measurement system and the aerosol data set is given by Brenninkmeijer et al. [1999] and Hermann et al. [2003, 2008].

Since there may be regional differences in the encountered aerosol and cloud types, data were analyzed separately for the Arabian Sea, the Middle East, and southeastern Europe (Indian route), as well as the Caribbean region and the midlatitude North Atlantic Ocean (Caribbean route).
Atmospheric Science Data Center (http://eosweb.larc.nasa.gov/PRODOCS/isccp/table_isccp.html). In 1999, William B. Rossow stated [Rossow and Schiffer, 1999, p. 2266], “The ISCCP calibrations are now the most complete and self-consistent set of calibrations available for all of these radiometers.” Detailed information on the ISCCP calibration, the ISCCP products, and the ISCCP error estimation is given by Rossow and Schiffer [1991], Rossow and Garder [1993a, 1993b], Brest et al. [1997], Rossow and Schiffer [1999], and the ISCCP webpage (http://isccp.giss.nasa.gov/).

In order to cover all backward trajectories from the two CARIBIC flight routes, it was necessary to use data from several geostationary weather satellites. For the Indian route, we used ISCCP data taken from METEOSAT-7 at 0°/C176 E and METEOSAT-5 at 63°/C176 E. For the Caribbean flights, we used also METEOSAT-7 and GOES-East at 75°/C176 W.

2.4. Cloud Contact Analysis Algorithm

All backward trajectories along the flight route were checked for cloud contact to determine whether a measured aerosol particle concentration had been affected by a cloud during the previous 2 days. A FORTRAN algorithm was developed which overlays the trajectories and the satellite images. If there is a cloud on a trajectory node, the algorithm compares the cloud top pressure to the trajectory pressure. If the cloud top is at the same altitude or higher compared with the trajectory node, the program marks the trajectory at this node with “cloud contact: yes” and notes the pressure difference between the cloud top and the trajectory. Otherwise, the trajectory node is marked with “cloud contact: no.” In most cases the CTH of deep convective clouds is underestimated by satellite measurements by approximately 1 km [Sherwood et al., 2004a, 2004b]. To account for this bias, the analysis program allows the trajectory to be up to 40 hPa (~1 km) above the cloud, but still to be marked as “cloud contact: yes.” The analysis was repeated using a margin of 20 hPa (~500 m), and yielded the same results as those shown in section 3 for a 40 hPa margin.

To account for the different spatial and temporal resolutions of the satellite cloud images (3 h) and the backward trajectories (1 h), a search function was incorporated to locate the image pixel closest to each trajectory node. This search function uses the wind direction provided by the trajectory data in order to account for the possible movement of a cloud or its outflow. If the cloud image was recorded earlier than the trajectory node, the program searches upwind for the closest pixel. If the cloud image was recorded after the trajectory node, the program searches downwind. The search starts at the trajectory node. If there is no cloud image pixel at the exact trajectory node coordinates (which is highly probable), the program searches a 0.1° square either with or opposed to the wind direction at the trajectory node (compare Figure 2). If still no pixel is found, the size of the square is stepwise incremented by 0.1°, until it reaches an upper extent of 0.5°. If there is still no pixel found, it is concluded that no information is available. Since the uncertainties increase with distance, the size of the search square is not extended beyond this limit. In only 5% of all analyzed trajectory nodes no adequate cloud pixels were found.

The cloud analysis program interpolates both in space and in time. If the time difference between the trajectory node and the satellite image for the cloud contact decision is more than 30 min, the program calculates a time-dependent average CTH (Figure 3). For the time interpo-
tion, the corresponding cloud image pixels that were recorded earlier and later than the trajectory node are checked for cloud occurrence. In case there is a cloud, a linear interpolation is made between the two pixels. This results in the computation of the averaged CTH at the trajectory node. Figure 3 shows an example in which the CTH at one pixel grows from about 700 hPa to 350 hPa. The trajectory node was recorded 60 min after the cloud image, and at this location the linear interpolation yields a cloud contact. If no interpolation was performed, the determination of no cloud contact would be made, since the closest image pixel in time would indicate that the cloud was too low. Admittedly, the assumption of a linear growth of CTH is simplistic. However, if all trajectory nodes are disregarded if they occur more than 30 min from the satellite images, then too much information would be lost or a false attribution could be made, resulting in even larger uncertainties.

We evaluated the linear time interpolation procedure by comparing our interpolated CTT data to noninterpolated satellite data with a temporal resolution of 15 min. For this purpose, we used satellite data of GOES-east from the National Oceanic and Atmospheric Administration (NOAA, http://www.class.noaa.gov/saa/products/welcome), which were processed for CTT values at the Max Planck Institute for Chemistry (MPI-C) in Mainz. All of the backward trajectories from one flight on the Caribbean route (4219 cases) were compared using both interpolated and noninterpolated data sets. In approximately 90% of these trajectories, there was agreement for the cloud contact decision. For these cases that were in agreement, the temperature difference between the two data sets was 0.6 K on average, with a standard deviation of 10.3 K. For the cases where the cloud contact decision disagreed, the mean temperature difference was 3.4 K with a standard deviation of 31.0 K. We found no systematic deviation in the data. We conclude that the agreement is satisfactory and that the interpolation mechanism works well.

By applying the entire cloud analysis algorithm to the backward trajectories, the following four questions could be answered for all measurement points: (1) Had the air parcel been in contact with a cloud within the previous 48 h? (2) If so, at what local time did it occur? (3) What was the duration of the last cloud contact? (4) How much time elapsed between the cloud contact and the measurement?

We will discuss these questions in the following sections, with the exception of the influence of the local time. The local time should have an effect on particle formation because of the availability of radiation energy...
to form OH and hence H$_2$SO$_4$ [Weber et al., 1997; Hermann et al., 2003; Berndt et al., 2005]. However, CARIBIC-1 does not provide enough measurement points for a statistically significant analysis of this effect. This analysis would require that the data set be divided according to cloud contact, local time of contact, and local time of measurement, resulting in data subsets that are too small to be representative.

3. Results and Discussion

3.1. Arabian Sea and the Caribbean Region

[19] Out of the 2475 tropospheric measurements obtained over the Arabian Sea, 1069 (43%) were influenced by cloud contact within the prior 48 h. Over the Caribbean region, 344 values (46%) were influenced and 397 measurements (54%) were not influenced by cloud contact. In both of these regions, deep convective clouds occur quite often because of the general atmospheric circulation (ISCCP, Data analysis to understand climate, http://isccp.giss.nasa.gov/climanal.html, 2005, accessed 8 January 2008). Hence, the cloud influence on the particle number concentration at 9–11 km altitude can be mainly attributed to deep convective clouds.

[20] Figure 4 compares the histograms of cloud-affected particle number concentrations (black columns) with unaffected concentrations (light gray columns) over the Arabian Sea. The fraction of nucleation mode particles was calculated using equation (1).

[21] Figures 4a and 4c reveal a significant difference between the cases with and without cloud contact. Compared with the unaffected cases, the cloud-affected cases have a median concentration almost six times higher ($N_{4-12}$: 773 particles/cm$^3$ STP versus 126 particles/cm$^3$ STP) and a median relative fraction almost four times higher ($r_{FUFP}$: 17.6% versus 4.8%). Note the logarithmic x axis scale. This clear increase in the concentration of nucleation mode particles is likely caused by new particle formation due to tropical deep convective clouds over the Arabian Sea and the Bay of Bengal, where most of the trajectories came from [cf. Hermann et al., 2003]. This assumption will be discussed in detail below with the discussion of Figure 5. The histograms in Figures 4a and 4c further show that in more than 29% of the cases with no cloud contact during the prior 48 h, the absolute particle number concentration was below 90 particles/cm$^3$ STP ($r_{FUFP}$: below 1.8%) or even zero (first two bins). Similar to the case with the nucleation mode particles, the concentration of Aitken mode particles was increased whenever there was cloud contact within the prior 48 h (Figure 4b). However, the $N_{12}$ median is only about 30% higher than the unaffected background. The statistical “Mann-Whitney-U-test” (MWU-test) [Bosch, 1990; Pruscha, 2006] yielded significant differences for all compared histogram pairs in each graph of Figure 4.

[22] The large number of data points over the Arabian Sea allows a more detailed analysis to be performed. These data were plotted as mean and median number concentrations versus the elapsed time between the last cloud contact and the measurement (Figure 5). For a data set that is approximately normally distributed, the median is a representative value; however, the mean value can be strongly influenced by extreme values. Thus, the difference between the mean and the median could indicate the occurrence of extreme events. For the two graphs in Figure 5, the mean and median concentration at, e.g., 24 h were calculated using all of the measurements with a cloud contact occurring 24 h back in time or less. This “integral” time presentation form uses more data points and results in smoother curves compared with a representation using only data at a given time. However, the disadvantage to this form is that any potential dependence on cloud contact time is damped. A comparison between the two time scale representations shows a similar curve progression in each case; however, for the nonintegral time scale the curve is much noisier. The similar curve progression indicates that artifacts due to cloud droplet or ice crystal breakup during in-cloud measurements were minimal, since for nonintegral time scale representations,
the value at zero time lag does not show a step increase above the values with a nonzero time lag. With the integral presentation form, more cloud cases (>300) are used to calculate the mean and median for nonzero long time lags (right side of graph) than for short time lags (left side of graph). Over the Arabian Sea, southeastern Europe, and the midlatitude North Atlantic Ocean, at least 17 different cloud cases were available at time lag zero. Each cloud case includes three concentration values on average. In the Caribbean region only eight different cloud cases were available, because there were fewer flights in this region.

Figure 5 shows that in the UT over the Arabian Sea, the less time between the last cloud contact and the measurement, the higher the mean number concentration of nucleation mode particles and the lower the mean concentration of Aitken mode particles. These nucleation mode particles are likely formed in the outflow region of convective clouds. Because of coagulation and impaction scavenging, the lifetime of such small particles in a cloud is too short to survive transport from the boundary layer to the UT [Ekman et al., 2006]. In the outflow regions, temperature is low, relative humidity is high, actinic fluxes are high, preexisting particle surface area is low, and oxidized precursor gases may be available. All these conditions favor particle nucleation [Clarke and Kapustin, 2002; Williams et al., 2002; Twohy et al., 2002; Kulmala, 2003; Lee et al., 2004; Kojima et al., 2004; Kulmala et al., 2004, 2006].

The mean values of the cloud-affected \( N_{4-12} \) can be compared with the median values in Figure 5a. The median decreases slower than the mean with increasing time between the cloud contact and the measurement. The median value of \( N_{4-12} \) at time lag 47 h is about 20% lower than the median value at time lag zero. However, the cloud-affected median value of \( N_{4-12} \) is still over six times higher than the background median of \( N_{4-12} \). Hence, the strong influence of clouds is apparent. The difference between the mean and median values of \( N_{4-12} \) indicates that most of the tropical clouds over the Arabian Sea and the surrounding regions contribute to particle formation. Thereby a few clouds cause large particle bursts, which increase the overall number concentration to almost 4000 particles/cm\(^3\) STP. The mean value of \( N_{4-12} \) strongly decreases with increasing time difference, not only because of the condensational growth to Aitken mode particles, but also because of the enhanced coagulation in air masses with very high particle number concentrations.

At a first glance, the picture presented above seems to be consistent. However, on second thoughts it seems contradictory that the highest nucleation mode number concentrations were actually measured close to the clouds. If the particles had been freshly formed in the outflow regions of the clouds, they should have been too small (~1 nm) to be detectable by the CARIBIC CPCs (lower cutoff ~4 nm). Directly in or very close to the cloud, the concentration of nucleation mode particles should be minimal, because new particles would not have had enough time to grow to detectable sizes [Weber et al., 1997]. This paradox can be explained assuming that the CARIBIC measurements were conducted mainly in the downdraft regions around deep convective clouds. Outflow altitudes of tropical deep convective clouds are mostly higher than the cruising altitudes of commercial aircraft [Folkins and Martin, 2005]. Moreover, pilots invariably try hard to avoid contact with deep convective clouds because of the strong updrafts and the associated turbulence. Hence, it is very likely that in this region observed nucleation mode particles were formed in the outflow of deep convective clouds above flight altitudes but were measured in the downdraft regions of these clouds. Nucleation mode particles would have had an age on the order of several tens of minutes already and hence should have had enough time to grow to detectable sizes. Because the aircraft fly probably below the anvil of the deep convective clouds, the analysis algorithm determines a cloud contact at short time lags. The results obtained for the Caribbean region support this theory.

The picture of new particle formation in the spatial and temporal vicinity of clouds is supported by the behavior of the Aitken mode particles. First, clouds act as particle sinks (as seen by the points on the left side of Figure 5b), which leads to mean and median number concentrations below the nonaffected background (dotted and dashed lines). However, as the time between the cloud contact and the measurement increases, the Aitken mode number concentrations increase above the background values. This increase is likely caused by condensational and coagulation growth from the nucleation mode size to Aitken mode size. As this behavior shows, deep convective clouds over the Arabian Sea act as an indirect source for Aitken mode particles. Since the mean and the median values for the number concentrations behave in parallel, the described sink and source effects seem to be effective for most of the tropical clouds over the Arabian Sea.

Homogeneous nucleation in the vicinity of convective clouds has been already observed in previous studies [e.g., Schröder and Ström, 1997; Wang et al., 2000; de Reus et al., 2001; Twohy et al., 2002; Clement et al., 2002; Lee et al., 2004; Kulmala et al., 2004; Kojima et al., 2004; Kulmala et al., 2006]. Bertram et al. [2007] derived a curve for the concentration of nucleation mode particles (which were defined as 3 nm ≤ \( d_p \) ≤ 10 nm) with an increasing time difference between convective events and the measurement, similar to that shown in Figure 5a. However, as the current study is based on a much larger data set, it yields statistically representative results.

Similar to the results found over the Arabian Sea, the median values of \( N_{4-12} \), \( N_{12} \), and \( \text{rF}_{\text{UFP}} \) over the Caribbean region are significant increased if a cloud contact occurred within the prior 48 h (Figure 6). The medians of \( N_{4-12} \) and \( \text{rF}_{\text{UFP}} \) for cloud-affected particle concentrations are increased by a factor of five and two, respectively, over the medians for particle concentrations not affected by clouds (\( N_{4-12} \): 1597 particles/cm\(^3\) STP versus 333 particles/cm\(^3\) STP; \( \text{rF}_{\text{UFP}} \): 18.6% versus 8.6%). This increase indicates that deep convective clouds are a source for new particles in the UT of the Caribbean region. The cloud-affected median value for \( N_{12} \) was increased by more than 75% (7220 particles/cm\(^3\) STP versus 4066 particles/cm\(^3\) STP). Hence, these clouds also act as a source for Aitken mode particles.

Similar to Figure 5 for the Arabian Sea, Figure 7 shows the measured particle number concentrations for the Caribbean region plotted against the elapsed time between the cloud contact and the measurement. Note that the scale on the y axes of Figures 5, 7a, and 7b are different. The mean of \( N_{4-12} \) behaves similarly to the mean over
the Arabian Sea, however, at much higher concentrations. The shorter the time elapsed between the cloud contact and the measurement, the higher the observed particle number concentrations (Figure 7a). The median values show the same behavior as the mean values, indicating that the majority of clouds contribute to homogeneous nucleation. However, with a time difference less than 11 h, the mean concentration decreases significantly. This could be due to the amount of time needed to grow freshly nucleated particles (~1 nm) to a detectable size (>4 nm). Ridley et al. [2004] showed that the strongest transport by convective clouds over the Gulf of Mexico and the southern USA occurs at an altitude of 9–11 km, matching the altitude of the CARIBIC measurements. Hence, the freshly nucleated particles were too small to be detected with the CARIBIC system directly after the cloud contact. The large time lag of

![Figure 7. Dependency of particle number concentrations on the time between the last cloud contact and the measurement, like in Figure 5 but for the Caribbean region. Note the different scales at the y axis.](image-url)
up to 11 h needed to grow these new particles to detectable sizes might also be due to a lesser growth speed as compared to the Arabian Sea (e.g., because of lower precursor gas concentrations).

[30] As the time difference increases between the cloud contact and the measurement, the mean and median values for $N_{12}$ in the Caribbean region increase significantly above the background values (Figure 7b). However, at a time lag of 2 h, the median value is at the background level, which contrasts to the same situation for the Arabian Sea (because there were only three different cloud situations available, the time lag of 0 h is not shown in Figure 7). This difference seems to indicate that contrary to the clouds over the Arabian Sea the analyzed clouds over the Caribbean region on average do not act as a immediate sink, but as an indirect source for $N_{12}$ by condensational and coagulation growth of freshly nucleated particles. The mean values of $N_{4-12}$ and $N_{12}$ show a much more pronounced behavior than the medians, indicating that some clouds have a stronger contribution to the nucleation of new particles than the majority. The maximum of the mean values of $N_{4-12}$ and $N_{12}$ was observed about 11 h after cloud contact (7258 particles/cm$^3$ STP and 13625 particles/cm$^3$ STP).

3.2. Middle East

[31] Because of the general atmospheric circulation (Hadley and Ferrell cells), the subtropical region of the Middle East is dominated by a downward welling air motion [Hupfer and Kuttler, 1998]. Consequently, fewer and predominantly lower clouds are observed in the Middle East (ISCCP, Data analysis to understand climate, http://isccp.giss.nasa.gov/climanal.html, 2005, accessed 8 January 2008).

[32] No clear difference can be seen between the 201 cloud-influenced and 1436 unaffected values (Figure 8a). However, when calculating the median for each histogram, a small difference appears. Over southeastern Europe and the midlatitude North Atlantic Ocean, the respective numbers are 708 (56%) and 548 (44%).

[33] Comparing the cloud-affected $N_{4-12}$ values for both flight routes (Figures 9a and 9c) with the unaffected $N_{4-12}$ values, no clear influence of clouds is apparent. However, when calculating the median for each histogram, a small difference appears. Over southeastern Europe, as well as over the midlatitude North Atlantic Ocean, the medians of the cloud-affected nucleation mode particle number concentrations are about 10% higher than the medians of the unaffected concentrations. This small difference also appears in the results of the MWU test. This result suggests that, on average, clouds over southeastern Europe and the surrounding areas, as well as over the midlatitude North Atlantic Ocean, slightly increase the particle number concentration of nucleation mode particles. However, over the North Atlantic, the cloud-affected mean of $N_{4-12}$ was
increased by ~80% above the unaffected mean, whereas over southeastern Europe the cloud-affected mean increased by only ~3%. This difference indicates that some clouds over the midlatitude North Atlantic Ocean, but not the majority, had a stronger influence on nucleation mode particles than those clouds over southeastern Europe.

[36] There was no significant shift due to the influence of clouds on the Aitken mode particle concentration over southeastern Europe (Figure 9b). The same result was also indicated by the MWU test. The absence of a shift might indicate that the clouds over this region do not affect the number concentration of Aitken mode particles. Conversely, this absence could be caused by the different influences of the various cloud types superimposing within this graph such that their effects cancel out (compare discussion at the end of this section). The N_{12} histograms, recorded over the midlatitude North Atlantic Ocean, significantly differ from each other (Figure 9d). Data with an occurrence of cloud contact within the prior 48 h had a 32% lower median value for the particle number concentration. This reduction indicates that clouds over the midlatitude North Atlantic Ocean act as a sink for Aitken mode particles, on average.

[37] Comparing the time dependence plots of both regions, it can be seen that the mean values of N_{4–12} differ in their temporal behavior (Figures 10a and 10c). Over southeastern Europe, the mean N_{4–12} is about 40% below the unaffected background with zero time lag. As time increases between the cloud contact and the measurement, the mean concentration increases and reaches the background level after about 14 h. The reduced value at zero time lag indicates that clouds over southeastern Europe act as a sink for preexisting nucleation mode particles. Thereafter, on average about 14 h are required before the mean concentration rises back to the background level. However, over the midlatitude North Atlantic Ocean, the mean value of N_{4–12} was already at background level at zero time lag; it subsequently increased to ~175% above the background level after a 5 h time lag. As the time difference increases, the concentration decreases likely because of coagulation and condensational growth. As already discussed in relation to Figures 5 and 7, 5 h might be the amount of time needed to grow freshly nucleated particles to detectable size. Compared with the Caribbean region, the maximum concentration over the North Atlantic Ocean occurs at a somewhat earlier time lag (5 h for the North Atlantic versus 11 h for the Caribbean), indicating that less time was needed to grow the new particles to detectable size. This shorter time requirement might be due to the higher occurrence of condensable precursor gases at midlatitudes. However, because the median values of N_{4–12} over the midlatitude

**Figure 9.** Histograms of particle number concentrations, like in Figure 4 but for northern midlatitudes. (a and b) Over southeastern Europe, 626 cloud influenced air masses were compared to 790 not influenced values. (c and d) Over the midlatitude North Atlantic Ocean the respective numbers are 708 and 548.
North Atlantic Ocean remain almost constant as the time difference increases, the effect described above was only caused by a few strong events. The median values of $N_{4-12}$ over southeastern Europe show a time dependency and will be discussed below. Since deep convective clouds led to particle formation over the Arabian Sea and over the Caribbean region, it is likely that the same effect would be seen at midlatitudes. Particle nucleation by midlatitude deep convective clouds has been reported by Schröder and Strom [1997], Wang et al. [2000], Clement et al. [2002], Twohy et al. [2002], and Young et al. [2007].

While there is no clear influence of clouds on the Aitken mode particle number concentration over southeastern Europe (Figure 9b), a time dependency for this concentration becomes apparent (Figure 10b). The shorter the elapsed time between the cloud contact and the measurement, the lower the mean and median values of $N_{12}$, for time lags above 8 h. For smaller time differences, the median significantly increases up to almost background level as the time lag decreases to zero. This behavior was also observed for the median value of $N_{4-12}$ in this region. Hence, the concentration of nucleation and Aitken mode particles was probably not directly reduced by the cloud contact. The reason for this behavior is not obvious. However, as the median decreases stepwise with increasing time difference, a statistical artifact is not likely. One explanation is that very thin cirrus clouds occur undetected after the detected cloud contact, thereby reducing the number concentration. Jensen et al. [2001] showed that supersaturation over ice occurs quite often at the midlatitude UT (this appears in up to 45% of their data). Jensen et al. [2001, p. 17, 263] state that “the fraction of the sky that could be converted by low optical depth ice clouds formed in moderately supersaturated regions is potentially very large.” From the plot of the median values, it can be seen that about 26 h are required to increase $N_{4-12}$ to the background level; for $N_{12}$ even 48 h are necessary.

In agreement with Figure 9d, Figure 10d affirms that the Aitken mode particle concentration over the midlatitude North Atlantic Ocean was significantly reduced by the majority of clouds. However, in contrast to the measurements along the Indian route (Figure 10b), no decrease in the median was observed with decreasing time difference. Nevertheless, along both flight routes, most clouds at midlatitudes seem to act as a sink for particles larger than 12 nm.

Since there is a wide spectrum of cloud types at midlatitudes and these different types of clouds can influence the particle number concentration in different ways, it is difficult to interpret the mean influence of all clouds on the particle number concentration. Unfortunately, our analysis provides no differentiation of cloud types because of the limited information within the ISCCP-DX data set (compare section 2.3). Hence, the curves displayed in Figure 10 might lose distinction because of the varying

---

**Figure 10.** Dependency of particle number concentrations on the time between the last cloud contact and the measurement, like in Figure 5 but (a and b) for southeastern Europe and (c and d) for the midlatitude North Atlantic Ocean. Note the different scales at the y axes of $N_{4-12}$ and $N_{12}$.
effects that stratiform clouds or convective clouds could have on \( N_{4-12} \). Differentiating between the cloud types in future work would be possible by using satellite data from the second generation METEOSAT (compare section 4). Plotting the data of each individual point in Figure 10 in a separate histogram, a bimodal structure becomes visible for most of the points. Figure 11a shows as an example the histogram of all measurements, which contributed to the data point of cloud contacts 3 to 5 h before measurement in Figure 10b. The bimodal structure at midlatitudes supports the assumption that different cloud types have different influences. Repeating the analysis for the same data point over the Arabian Sea, where deep convective clouds occur quite often (compare section 3.1), results in a monomodal structure (Figure 11b). However, only small numbers of data points were available for the histograms (~60 each). Hence, caution is needed to interpret these results.

3.4. Analysis of the Cloud Contact Duration

The cloud analysis algorithm can be used to compute the duration of the last cloud contact, with a temporal resolution of 1 h (compare section 2.4). With this information, the cloud-influenced measurements for both flight routes in each region were further categorized into two groups, on the basis of the contact length. The first group contains the cases where the last cloud contact persisted for 4 h or more (long cloud contact). The second group contains the cases where the last cloud contact lasted for 1 h (short cloud contact). These categories were selected to clearly discriminate between the short and long cloud contacts. Any interpretation of this comparison should account for the large uncertainty in the cloud contact duration result, caused by the large uncertainty in the backward trajectories inside clouds [Scheele et al., 1996; Stohl, 1998; Stohl et al., 2001].

Over the Arabian Sea and the midlatitude North Atlantic Ocean, neither the mean nor the median of the measured particle number concentrations of any size depended strongly on the duration of the last cloud contact (Table 2). Any effect from the clouds depended only on whether cloud contact occurred or not. However, over the Caribbean and southeastern Europe, there is a dependency on cloud contact duration. In these regions, there were up to 110% higher mean and median values of \( N_{12} \) with long cloud contacts compared with short cloud contacts (Table 2). Furthermore, the long cloud contacts caused a significantly higher median \( N_{4-12} \) than the short cloud contacts. Conversely, over the Middle East, there was a dependence on cloud contact duration for the mean values, but not the median values of number concentration. For the Middle East, the mean values of the nucleation and Aitken mode particles were about 72% and 53% lower, respectively, with long cloud contacts than with short cloud contacts. Unfortunately, only 16 long cloud contact cases were available over the Middle East, and hence, the result is not definite.

Currently, we have no clear explanation for the various dependencies on cloud contact duration seen in different regions. These differences may be due to various cloud types, or to the uncertainties of the trajectories inside clouds, particularly in deep convective clouds occurring in

![Histograms of Aitken mode particle number concentrations (\( N_{12} \)), like in Figure 4 but using only data at which the last cloud contact was 3 to 5 h before the measurement. (a) Over southeastern Europe, 53 cloud influenced and 790 not influenced data points were used. (b) Over the Arabian Sea the respective numbers are 69 and 1416.](image)

Table 2. Dependency of the Particle Number Concentration in the UT on the Duration of the Last Cloud Contact

<table>
<thead>
<tr>
<th>Geographic Region</th>
<th>( N_{4-12} )</th>
<th>( N_{4-12} )</th>
<th>( N_{12} )</th>
<th>( N_{12} )</th>
<th>Number of Data Points</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arabian Sea</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \geq 4 ) h</td>
<td>1869</td>
<td>789</td>
<td>3728</td>
<td>2898</td>
<td>246</td>
</tr>
<tr>
<td>1 h</td>
<td>1554</td>
<td>766</td>
<td>4367</td>
<td>3531</td>
<td>543</td>
</tr>
<tr>
<td>Caribbean region</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \geq 4 ) h</td>
<td>5360</td>
<td>3499</td>
<td>14528</td>
<td>11608</td>
<td>74</td>
</tr>
<tr>
<td>1 h</td>
<td>3700</td>
<td>1214</td>
<td>10511</td>
<td>6598</td>
<td>171</td>
</tr>
<tr>
<td>Middle East</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \geq 4 ) h</td>
<td>189</td>
<td>131</td>
<td>1237</td>
<td>857</td>
<td>16</td>
</tr>
<tr>
<td>1 h</td>
<td>672</td>
<td>170</td>
<td>2610</td>
<td>1249</td>
<td>146</td>
</tr>
<tr>
<td>Southeast Europe</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \geq 4 ) h</td>
<td>3399</td>
<td>1274</td>
<td>8770</td>
<td>3841</td>
<td>109</td>
</tr>
<tr>
<td>1 h</td>
<td>2386</td>
<td>747</td>
<td>4162</td>
<td>2437</td>
<td>348</td>
</tr>
<tr>
<td>Midlatitude North Atlantic Ocean</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \geq 4 ) h</td>
<td>918</td>
<td>207</td>
<td>2380</td>
<td>1672</td>
<td>169</td>
</tr>
<tr>
<td>1 h</td>
<td>831</td>
<td>206</td>
<td>3509</td>
<td>1935</td>
<td>360</td>
</tr>
</tbody>
</table>

*For each climatic region the measurements were separated into “long” (\( \geq 4 \) h) and “short” (1 h) cloud contacts, respectively. All concentrations were converted to standard conditions (STP: 1013.25 hPa and 273.15 K) and are given in particles/cm³ STP.
Table 3. Summary of the Average Influence of Clouds on the Medians of the Particle Number Concentration of Nucleation (N$_{4−12}$) and Aitken Plus Accumulation Mode Particles (N$_{12}$) in the UT in the Individual Geographic Regions

<table>
<thead>
<tr>
<th>Geographic Region</th>
<th>Average Influence on N$_{4−12}$</th>
<th>Average Influence on N$_{12}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arabian Sea</td>
<td>source</td>
<td>sink in first instance</td>
</tr>
<tr>
<td></td>
<td></td>
<td>and indirect source</td>
</tr>
<tr>
<td></td>
<td></td>
<td>by growth of</td>
</tr>
<tr>
<td></td>
<td></td>
<td>nucleation mode particles</td>
</tr>
<tr>
<td>Caribbean region</td>
<td>source</td>
<td>indirect source</td>
</tr>
<tr>
<td></td>
<td></td>
<td>by growth of</td>
</tr>
<tr>
<td></td>
<td></td>
<td>nucleation mode particles</td>
</tr>
<tr>
<td>Middle East</td>
<td>no effect</td>
<td>sink</td>
</tr>
<tr>
<td>Southeastern Europe</td>
<td>sink for preexisting particles</td>
<td>sink</td>
</tr>
<tr>
<td>Midlatitude North Atlantic Ocean</td>
<td>no effect</td>
<td>sink</td>
</tr>
</tbody>
</table>

[44] The effect of clouds on the number concentration of nucleation mode particles (N$_{4−12}$) and of Aitken plus accumulation mode particles (N$_{12}$) in the upper troposphere was analyzed using a combination of in situ measurements, backward trajectories, and satellite pictures. The aerosol measurements acquired in the CARIBIC project (http://www.caribic-atmospheric.com) [Brennkimeijer et al., 1999, 2007] were divided into five regions: the Arabian Sea, the Caribbean, the Middle East, southeastern Europe, and the midlatitude North Atlantic Ocean. An analysis algorithm was developed to check the backward trajectories (calculated by KNMI, de Bilt, Netherlands) for the occurrence of cloud contact within 48 h prior to measurement. If there was cloud contact, the algorithm notes the duration of the last cloud contact and the time lag to the measurement. The satellite data were obtained from the International Satellite Cloud Climatology Project (ISCCP) (http://isccp.giss.nasa.gov/). Unfortunately, this ISCCP DX data set does not distinguish between different cloud types; hence, only the average influence of clouds could be discerned. Potential errors in the results could arise because of the assumptions in the analysis algorithm and in the input data set. To reduce the potential error from the backward trajectories, they were analyzed only up to the last cloud contact, and stopped in either case at 48 h. Nevertheless, errors might still arise in the duration of the last cloud contact, because of the uncertainty of trajectories inside clouds. The satellite images are another potential source for uncertainties, since they underestimate the CTH by approximately 1 km [Sherwood et al., 2004a, 2004b]. This error was addressed by incorporating the bias into the analysis algorithm (compare section 2.4). To account for the different spatial and temporal resolutions of the satellite image data and the trajectories, the cloud contact analysis algorithm contains a search function dependent on the wind direction, which locates the closest pixel to a trajectory node and linearly interpolates between temporally adjacent images. Nevertheless the developed algorithm seems to perform well, as a comparison with another high-resolution satellite data set has shown. The results of the analysis are summarized in Table 3, and are valid for the majority of the analyzed clouds.

[45] Over the Arabian Sea and over the Caribbean, the mean and median values of the number concentration of nucleation mode particles increased if cloud contact occurred within 48 h prior to the measurement. Moreover, the shorter the time difference between the measurement and the cloud contact, the higher the mean value of N$_{4−12}$ and the lower the mean and median values of N$_{12}$. This behavior indicates that the majority of the clouds (mostly deep convective) over both regions act as a source of new particles. Furthermore, the clouds over the Arabian Sea most likely act as a sink for Aitken and accumulation mode particles in the first instance. However, they also act as an indirect source for Aitken mode particles by the growth of the freshly nucleated particles with time. In contrast, over the Caribbean region, the clouds seem not to act as a immediate sink, but rather as an indirect source for N$_{12}$. Because of the favorable conditions the formation of new particles is assumed to take place in the outflow regions of the clouds. Thereby few clouds contribute to large particle bursts, increasing the overall mean concentration of N$_{4−12}$ over the Arabian Sea to four times the median. Over the Caribbean region, these large particle bursts cause the mean to double in value over the median.

[46] In contrast to the Arabian Sea and the Caribbean, the analyzed clouds over the Middle East seem not to act as a source for new particles. This might be due to the less frequent occurrence of deep convection, and to the lower concentrations of precursor gases. The cloud contacts significantly reduce the Aitken and accumulation mode particle number concentration.

[47] Over southeastern Europe and the midlatitude North Atlantic Ocean, N$_{12}$ seems to be significantly reduced by the majority of the detected clouds. Additionally, the clouds over southeastern Europe do probably act as a sink for preexisting nucleation mode particles. Over the midlatitude North Atlantic Ocean, the majority of the clouds do not influence N$_{4−12}$. Nevertheless, a few clouds provide such a strong contribution to the formation of new particles that the overall mean of N$_{4−12}$ is increased up to 175% above the background level. Since several types of clouds occur at midlatitudes, there could be varying effects on the aerosol particles. Additional work is needed to distinguish the influence due to specific cloud types.

[48] Further analysis has shown that long-duration cloud contacts (≥4h) over southeastern Europe and the Caribbean have increased the mean and median values of N$_{12}$ up to 110% higher than values associated with short-duration cloud contacts (1h). Also, the concentration of nucleation mode particles increased because of long cloud contacts over both regions. However, over the Arabian Sea and the Middle East, the duration of the cloud contact had no effect; it was only relevant whether any cloud contact occurred. Over the midlatitude North Atlantic Ocean, no dependency on the cloud contact duration was discerned. The interpretation of these different behaviors is still open.

[49] Distinguishing between the different cloud types in the analysis method would increase the understanding of
how clouds affect the particle number concentration, particularly at midlatitudes. Unfortunately, the ISCCP data set provides information on cloud optical thickness (used to distinguish cloud types) only during the daytime. However, second generation METEOSAT satellites (starting with METEOSAT-8) routinely offer the cloud optical thickness, and therefore, the cloud classification [EU-METASAT, 2007]. In future work, these data could be applied to the measurements obtained from CARIBIC-2 (2004–2014). Moreover the second phase of CARIBIC provides more trace gas information than phase one as well as the particle size distribution obtained with an optical particle counter [Brenninkmeijer et al., 2007]. With this additional information, a more detailed analysis of the influence of clouds on aerosol particles in the UT/LMS can be made in future work.

References

[50] Acknowledgments. We thank LTU International Airways, Lufthansa Passage, and Lufthansa Technik for enabling CARIBIC. Furthermore we thank ISCCP, NOAA, and EUMETSAT for providing satellite pictures. Sincere thanks to Dominik Brunner (EMPA) for his help in evaluating the analysis algorithms and to Jörg Aumüller (DWD) for his help in decoding the EUMETSAT data. Special thanks to Paul Carter from Langley Atmospheric Sciences Data Center for his help with the ISCCP data. CARIBIC was funded by the German Ministry for Research and Education within the AFO2000 research program (07 ATF 17) and by ISCCP data. We thank LTU International Airways, Lufthansa Passage, and Lufthansa Technik for enabling CARIBIC. Further-


C. A. M. Brenninkmeijer and G. Schlaf, Atmospheric Chemistry Division, Max Planck Institute for Chemistry, D-55128 Mainz, Germany. M. Hermann, A. Weigelt, and A. Wiedensohler, Physics Department, Leibniz Institute for Tropospheric Research, Permoserstrasse 15, D-04318 Leipzig, Germany. (weigelt@tropos.de)

P. F. J. van Velthoven, Royal Netherlands Meteorological Institute, NL-3730 AE de Bilt, Netherlands.

A. Zahn, Institute for Meteorology and Climate Research, Research Center Karlsruhe, D-76021 Karlsruhe, Germany.