Environ. Res. Lett. **3** (2008) 025010 (7pp) doi:10.1088/1748-9326/3/2/025010

Meridional gradients of light absorbing carbon over northern Europe

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Received 15 January 2008
Accepted for publication 12 May 2008
Published 30 May 2008
Online at stacks.iop.org/ERL/3/025010

Abstract

In situ measurements have been made in the upper troposphere of the properties of particles containing light absorbing carbon (LAC). These measurements, made in late November 2006 over northern Europe, show that the average LAC mass concentration varies between 1 and 5 ng m^{-3} over a latitude range 50° to 70°N, with maxima at 50° and 66°. The relative fraction of all particles larger than 0.1 μm that contain LAC decreases at higher latitudes. The derived extinction coefficient, which also increases with latitude, reaches a maximum of 1.4 Mm^{-1} at 66°.

The air mass histories associated with the LAC were evaluated with back trajectory analysis using wind field analysis from the European Center for Median-Range Weather Forecast (ECMWF). A positive correlation exists between the fraction of particles containing LAC and the maximum relative humidity (RH), minimum temperature and maximum number of hours of cloud experienced by the air mass in the 5–10 days prior to being sampled by the aircraft. Air masses arriving from lower altitudes and with trajectories over North America also had larger concentration fractions of LAC.

The average LAC mass is in good agreement with previous measurements made over North America for the same latitude range, and the span of values fits best with model predictions of LAC distributions that assume that the LAC transported from surface sources is hydrophobic.

Keywords: black carbon, light absorbing carbon, LAC cloud processing

1. Background

Black carbon (BC) particles, hereafter referred to as light absorbing carbon (LAC)[5] particles, are some of the most ubiquitous and radiatively active of all aerosol particles found in the atmosphere. Light absorbing carbon describes those atmospheric particles that change the thermodynamic structure of the atmosphere and contribute to regional and global climate change as a result of their optical properties (Turco *et al* 1990, Charlson *et al* 1992, Penner *et al* 1998, Koch 2000, Koch *et al* 2007, Bond and Bergstrom 2006). In the boundary layer they serve to absorb solar radiation and cool the Earth's surface, but in the upper troposphere they augment climate warming from greenhouse gases. In a recent review, Ramanathan and Carmichael (2008) estimate that the emissions of LAC are the second strongest contributor to current global warming after carbon dioxide emissions. The upper troposphere (UT) is particularly sensitive to the presence of LAC since these particles have a lifetime of days to weeks, depending upon their

[5] This was suggested by Bond and Bergstrom (2006) who recommended the nomenclature 'light absorbing carbon' for research related to the radiative impact of these types of particle.

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age and other chemical species that eventually deposit on the LAC surfaces.

The source of these particles may be aircraft (Pueschel *et al* 1992, 1997, Blake and Kato 1995) or transport from tropospheric sources (Cook and Wilson 1996, Liousse *et al* 1996, Cooke *et al* 2002, Hendricks *et al* 2004). Most of what we know about LAC in the UT comes from a very limited number of *in situ* measurements (Pueschel *et al* 1992, 1997, Blake and Kato 1995, Baumgardner *et al* 2004, Schwarz *et al* 2006) and from transport models (e.g., Cook and Wilson 1996, Liousse *et al* 1996, Koch 2000, Cooke *et al* 2002, Hendricks *et al* 2004, Koch *et al* 2007). These models require accurate inventories of LAC emissions and uncertain assumptions about the lifetime of these particles. Very limited validation of these models has been carried out, due to the paucity of LAC measurements in the upper atmosphere. Hendricks *et al* (2004) compared simulated distributions of LAC, assuming various removal scenarios of these particles, with the airborne measurements available at that time. They concluded that the values estimated from the model agreed reasonably well within the uncertainties of the measurements but were very sensitive to the removal mechanisms used in the simulation. They summarized by emphasizing the need for much more extensive measurements over larger geographical regions throughout all seasons. Here we present measurements of LAC in the upper troposphere over northern Europe, a region where no previous direct measurements have been made.

2. Measurement and analysis methodology

2.1. LAC properties

The measurements were made with a single particle Soot Photometer[6] (SP2) that employs a patented technique (Stephens *et al* 2003) combining the principles of light scattering, absorption and emission to derive the diameter, mass and incandescence temperature of individual aerosol particles in the diameter range (for the current study) from 150 to 600 nm (Baumgardner *et al* 2004, Schwarz *et al* 2006, Moteki and Kondo 2007). Particles enter the sample cavity of the SP2 (the typical sample flow rate for the instrument is 2 cm^3 s^{-1}), and while passing through the beam of a diode-pumped Nd:YAG laser (1.064 μm wavelength) they scatter and absorb light. Two cones of scattered light, 30°–60° and 120°–150°, are collected by the optics and focused on a photodetector that produces a voltage signal proportional to the scattering intensity. The particle diameter is derived from the scattered light using classical Mie theory. The reader must be cognizant that the SP2 measures individual aerosol particles only in the size range of 90–600 nm for the derived LAC mass diameter, D_{mass_eqv}, and 100–450 nm, depending on refractive index, for the optical diameter, $D_{optical}$.

Particles that contain material that absorbs light at the wavelength of the laser are heated and reach a temperature at which they incandesce and emit light at a wavelength that is a function of the temperature. Two detectors, each with filters for passing different wavelengths, measure the emitted light. The temperature is derived from the ratio of signals from these detectors (Baumgardner *et al* 2004, Schwarz *et al* 2006, Moteki and Kondo 2007). The intensity of the emitted light is proportional to the mass of the light absorbing material, and the temperature of incandescence identifies the composition.

Calibration of the SP2 is carried out with commercially available spherical particles. Monodispersed, polystyrene latex spheres are used to calibrate the scattering signals, and glassy carbon spheres of known density were size-selected with an electrostatic classifier for calibration of the incandescence signals. Incandescing particles are sized in the range of 3–300 femtograms per particle (120 to 650 nm mass equivalent diameter at 1.42 g cm^{-3} density). The optical diameter is derived from the scattering signal and ranges from 100 to approximately 400 nm, depending on the assumed refractive index. Given that the scattering and incandescence signals are recorded particle-by-particle, the SP2 is sensitive to very low concentrations of LAC.

A detailed description of the theory of operation, uncertainties in determining the size and mass of LAC, and measurement limitations are documented elsewhere (Schwarz *et al* 2006, Moteki and Kondo 2007). For the interpretation of the measurements presented in the present study, the estimated uncertainties in LAC mass, $D_{optical}$ and D_{mass_eqv} are ±20%, ±50% and ±30%, respectively.

2.2. Back trajectory analysis

Ten day back trajectories were initiated every ten seconds along the flight path, an interval of approximately two kilometres at the research velocity of the aircraft. The trajectories were calculated with the program LAGRANTO (Wernli and Davies 1997) based on wind fields from ECMWF analysis which are available at a three hour interval. To drive the trajectory calculations the analysis fields have been interpolated on a 0.6° latitude/longitude grid in horizontal and 91 pressure levels in the vertical. To minimize interpolation errors the trajectories were initially calculated backward in time until the next ECMWF time step and afterward calculated back in time for 10 days.

The objective of the back trajectory analysis is to evaluate the latitudinal variations in the LAC properties with respect to the environmental histories of the air masses through which the aircraft passed while making its measurements. As discussed above, the major sources for LAC in the UT are either aircraft or transport from ground based emissions, primarily combustion processes from industry and motorized vehicles or biomass burning. The major removal mechanism in the UT is wet deposition, i.e. cloud processing. Hence, the metrics that we have chosen to use for evaluating the meridional gradients in LAC are related to the geographical locations through which the air masses have passed and to the amount of cloud processing that might have occurred during their passage during varying meteorological conditions.

The eight parameters that are derived from the back trajectory information are: (1) the maximum relative humidity (RH), (2) the number of hours in cloud, (3) the hours in the north Atlantic aircraft corridor, (4) the length of time between

[6] Droplet Measurement Technologies (DMT), Boulder, CO.

when the air is sampled from the aircraft and the last time the air mass contained cloud, (5) the hours over the eastern United States, (6) the maximum pressure from where the air mass ascended, (7) the number of hours the air was between 30°N and 40°N longitude, and (8) the number of hours the air was between 50°N and 60°N longitude. The hours in cloud is the amount of time that the ice water content, one of the ECMWF derived products, is greater than zero over the selected time period of the back trajectory. These parameters were calculated for periods from one to ten days prior to their intersection with the aircraft path along the back trajectories in order to assess the importance of short term versus long term histories of the air masses.

2.3. Field project description

The measurements were made during five flights from 23–28 November 2006, as part of the CIRRUS III field campaign, the third in a series of experiments to study the evolution of microphysical properties of cirrus in the UT over northern Europe. The aircraft was the Enviscope Lear Jet that was launched from the Hohn air force base in northern Germany. In addition to the SP2, a number of other particle and gas sensors were installed on the aircraft, including those to measure water vapor, total water, ozone, NO_y, condensation nuclei (CN), and cloud particle size spectra.

During all flights the ice water content (IWC) of the clouds was determined from the difference between the total water, i.e. gas and solid phase, and gas phase water. The Lyman-alpha hygrometer FISH (Fast *In situ* Stratospheric Hygrometer, Zöger *et al* 1999) is equipped with a forward facing inlet to sample the total water. Ice particles are over-sampled with an enhancement depending on altitude and cruising speed of the aircraft. Corrections are applied during post-flight analysis. Gas phase water was measured with the open path TDL OJSTER (MayComm Instruments, May and Webster 1993).

The FISH and the SP2 sampled from separate inlets that were located on either side of the fuselage. In addition, during taxi, takeoffs and landings the SP2 inlet was flushed to limit the number of LAC particles that might coat the inside of the inlet when in the heavily polluted boundary layer. In the present study only cloud-free air was analyzed in order to avoid counting artifact particles that are generated by ice particles when striking a sampling inlet (Murphy *et al* 2004). The total ice water values were used to determine when samples were being taken in cloud, and these are removed from the analysis when the ice water content was greater than a milligram per cubic metre.

Also avoided were those periods when the aircraft was passing through aircraft exhaust plumes. The average background number concentration of aerosol particles larger than 0.1 μm in the UT ranges from 10 to 100 cm^{-3}. In aircraft exhaust, depending on the age of the particles, this value increases by an order of magnitude or more. All samples were removed from the analysis when the total number concentration from the SP2 exceeded 100 cm^{-3}. Only measurements made at altitudes greater than an altitude of 9000 m were used for this study in order to limit the evaluation to the upper troposphere.

3. Results and discussion

3.1. Meridional trends in LAC properties

Figure 1 summarizes the LAC properties as a function of latitude for five of the six flights that were made during the campaign[7]. The solid lines represent the computed averages calculated every two degrees of latitude. The vertical bars are the standard deviations about the average.

The number concentration of LAC particles (black curve in figure 1(A)) has a maximum of 1.2 cm^{-3} at 50°, and then decreases by a factor of three to an average value of 0.4 ± 0.2 throughout the range of latitudes. The standard deviation is quite large for the concentration at 50°, however, so it is probably biased by a few very large values that are not necessarily representative of LAC concentrations at that latitude. The average fraction of total particles measured by the SP2 that contain LAC is 0.08 ± 0.04 (blue curve, figure 1(A)). This fraction reaches a maximum of 0.3 at 52° latitude but also has a very large standard deviation at this location, so cannot be considered significantly larger, but rather that there were some very large concentrations of LAC in this region during one of the flights that dominate the statistics. The LAC mass concentration (black curve in figure 1(B)) has a bimodal trend with maxima at 50° and 66°. The standard deviations indicate a large degree of inhomogeneity in the LAC spatial distribution such that even though there are general trends as suggested by the averages, higher levels of LAC mass are found throughout the range of latitudes. The trends in the fraction of LAC mass found in all the particles detected by the SP2 (blue curve, figure 1(B)) is similar to the concentration fraction, with a maximum at 52° and average of 0.04 ± 0.02. The general trend is to decrease with latitudes further north, but the large standard deviations prohibit attaching statistical significance to this trend.

Figure 1(C) shows the meridional trends in the mass equivalent diameter (black curves) and the extinction coefficient that is derived from the size distribution of all particles measured with the SP2, those with and without LAC. The diameter has a trend that follows that of the LAC mass concentrations, with similarly large standard deviations. The derived extinctions, however, have a more distinctive trend that maximizes at 66° latitude with a value of 1.3 ± 0.2 Mm^{-1} that is three times the minimum at 52° of 0.4 ± 0.4 Mm^{-1}, a statistically significant difference.

3.2. Comparison with previous studies

There are limited previous *in situ* observations of LAC in the UT and none of them measured LAC mass directly. Blake and Kato (1995) and Pueschel *et al* (1997) published measurements of BC mass concentrations made between 1992 and 1994 over a range of latitudes, mostly above North America. Wire impactor samples were analyzed with electron microscopy to estimate the BC mass, which varied from 0.01 to about 1 ng m^{-3}.

[7] The measurements from the first flight were discarded because of a leak in the SP2 air distribution system.

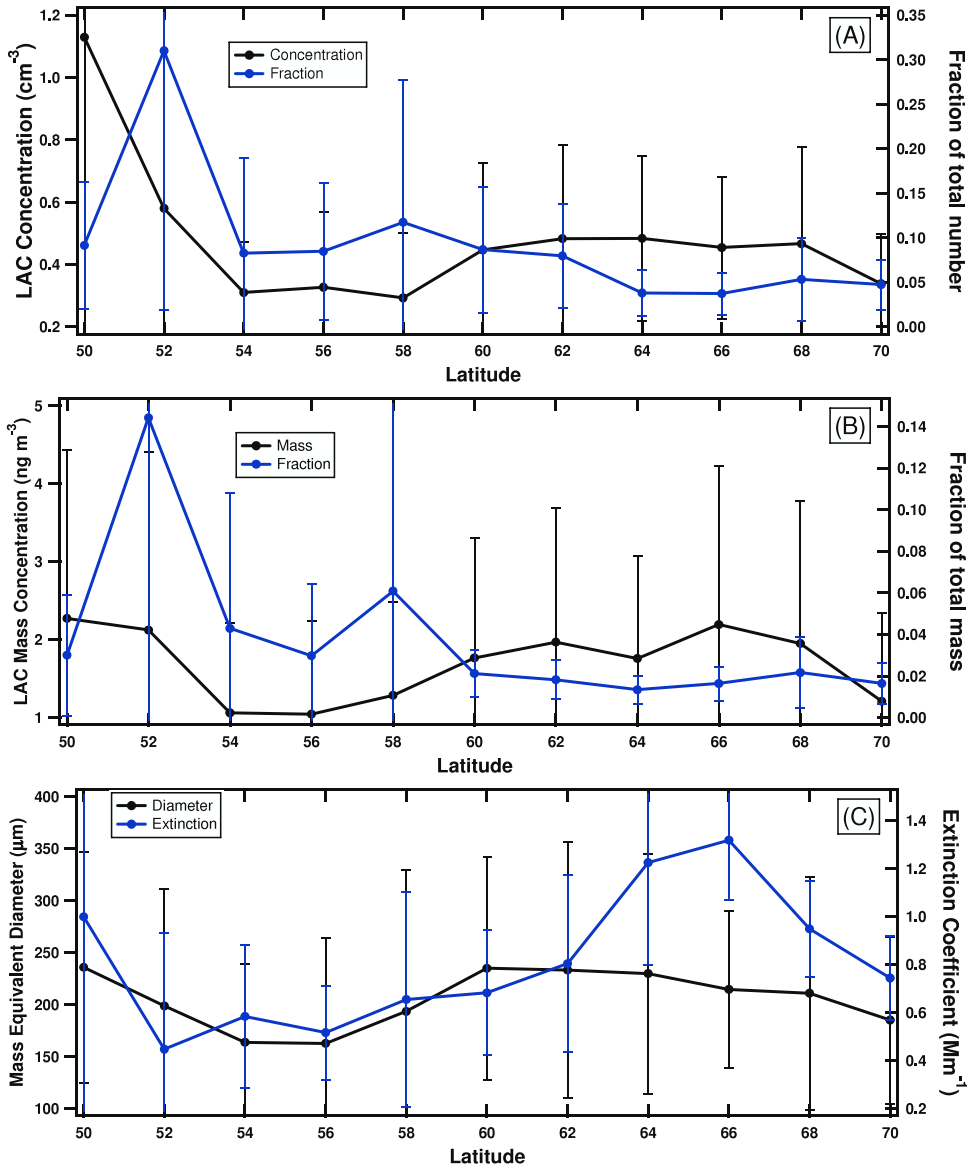

Figure 1. The meridional trends in LAC properties are shown here as averages (solid lines) and standard deviations (vertical bars). The number concentration and concentration fractions are seen in (A), the mass concentration and mass mixing fraction in (B), and the mass equivalent diameter and extinction coefficient in (C).

In 2003 the SP2 was deployed for the first time on the NASA DC-8 research aircraft during the second SAGE III Ozone Loss and Validation Experiment (SOLVE II) as part of the field campaign based in Kiruna, Sweden (67.8 °N, 20.3 °E) to evaluate ozone loss processes in the polar vortex. Measurements of LAC were made from 60° to 90° in late January and early February when the polar vortex was well developed. Outside of the vortex, between 60° and 66° the value of the LAC mass concentration varied from 0 to 5 ng m^{-3}. The concentration fraction of the LAC varied between 10 and 20%.

More recently, the NOAA[8] SP2 was flown on the NASA[9] WB-57F high-altitude research aircraft in November 2004 from Houston, Texas (Schwarz *et al* 2006). Two flights were made that measured LAC over the central US (29°–38°) and reached altitudes of 18.7 km. In the 9–12 km range, similar to that measured during the CIRRUS III campaign, the SP2 measured 0.2–2 ng m^{-3} of LAC mass.

The global distribution of BC has been modeled by Hendricks *et al* (2004) using the ECHAM4 global climate model (Roeckner *et al* 1996) and various emission inventories of BC with different assumptions about the e-folding lifetime of the BC particles were used to simulate the removal of BC particles by dry and wet deposition. When the BC particles are assumed to be hydrophilic the average BC mass concentration over northern Europe at 250 mb (approximately 10 km), the average flight altitude of the Enviscope research aircraft, is 0.5 ng m^{-3}. When the particles are assumed to be hydrophobic the average mass is 2 ng m^{-3}.

Hence, the values of LAC mass measured during CIRRUS III are consistent, i.e. within the same range, with those found

[8] National Oceanic and Atmospheric Administration.
[9] National Aeronautics and Space Administration.

Table 1. Spearman–Rank correlation coefficients. (Note: the number in parentheses is the number of days backwards along the air mass trajectory where the correlation was found. Only correlation coefficients with statistical significance >99% are listed.)

Back trajectory variables	LAC conc.	LAC conc. fraction	LAC mass	LAC mass fraction	Mass equivalent diameter
Maximum RH	−0.4 (1)	0.6 (⩾2)			
Hours in cloud	−0.5 (1)	0.7 (8)		−0.7 (1)	
Hours in traffic corridor	−0.4 (6)			0.5 (1)	−0.4 (⩾7)
Time to last cloud		−0.7 (8)		−0.5 (⩾9)	
Hours over eastern USA	−0.5 (⩾7)	0.4 (⩾8)	0.5 (⩾7)		−0.4 (⩾7)
Maximum pressure	0.5 (4)	0.7 (10)	0.4 (4)	0.4 (5)	
Hours 30–40°N	−0.4 (10)	0.6 (⩾6)	−0.6 (3)	−0.5 (3)	
Hours 50–60°N	0.4 (4)	−0.5 (⩾6)	0.5 (3)		

in the UT in other geographical locations in the northern hemisphere; however, given the large variability shown in the current study, we wish to explore in greater detail if these variations are related to the air mass history. This is discussed in the following section.

3.3. Back trajectory analysis

The LAC properties were correlated with the eight parameters derived from the air mass back trajectories, those that were described in section 2.2. The scatterplot between the number fraction of LAC and the maximum RH and atmospheric pressure, shown in figure 2, illustrates two of the relationships that were found between some of the LAC characteristics and air mass history. Table 1 summarizes these correlations, whereby the statistic is the Spearman–Rank correlation coefficient and is only reported if its level of significance is greater than 99%. The numbers in parentheses indicate the number of days over which the back trajectory was calculated before this level of significance was achieved. The numbers with '⩽' or '⩾' before them indicate that all the days previously or afterward, respectively, were significantly correlated. Blank cells indicate that the correlations were not significant.

The LAC number concentrations and concentration fractions were correlated with seven out of eight of the back trajectory parameters, followed by the mass fraction (5 out of 8) and mass concentration (4 out of 8). The equivalent mass diameter was weakly correlated with only two parameters, the hours in the north Atlantic flight corridor and numbers of hours over the eastern US. As seen in figure 2 and in the correlations, the concentration fraction of LAC increased as the maximum RH and pressure increased. There is also a strong positive correlation between the concentration fraction and the hours in cloud previous to intersecting the aircraft track. The positive correlation between LAC number fraction and the number of hours over the eastern US and in the latitude band from 30 to 40°N suggests that these air masses are probably the same. The negative correlations between concentration fraction and the last time in cloud, as well as hours in the latitude band from 50 to 60°N, indicates that there are relatively more LAC particles in air masses that have recently encountered cloud and fewer when the air mass has come from the more northern latitude.

The positive and negative correlations between the other LAC properties and back trajectory parameters listed in the table can be similarly linked, although their interpretation is not necessarily unambiguous as correlation does not necessarily imply causation. Under the assumption that LAC containing particles are produced from either surface based or airborne combustion sources, and are removed by clouds and precipitation, we analyze the correlations as they relate to the production and removal processes.

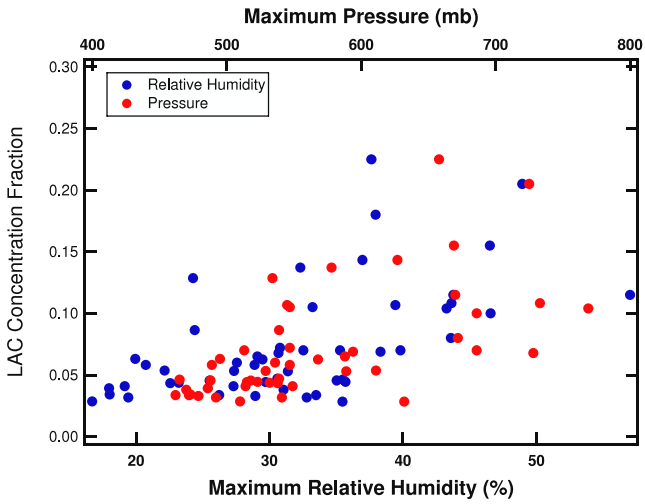

Figure 2. The average fraction of particles that contain LAC are compared here with the maximum relative humidity and pressure that was encountered by the air masses ten days before reaching the location where the LAC was measured.

The number and mass concentrations are larger with air masses that originate nearer the surface (higher pressures) and that arrive from more northern latitudes. The number fraction of LAC, however, decreases with latitude. This is consistent with biomass burning, i.e. forest fires, in Canada where such sources produce copious concentrations of particles, those with and without LAC. This source would produce higher LAC number and mass but smaller relative numbers of LAC.

Another large source of LAC particles is pollution from the population centers along the eastern US coast. This is a likely source of the positive correlations between numbers of hours over the eastern US and the LAC concentration fraction and LAC mass concentration. At odds with these correlations, however, is the negative correlation of the number concentration with the same parameter, i.e. the more time the air mass had been over that region of the US, the lower the

LAC concentration. It should be noted, however, that neither the positive or negative correlations were significant until the back trajectories had been integrated back at least seven days. This suggests that the air had probably been much further west prior to passing over the east coast and the correlations may have been unrelated to the air having passed over that region. Pollution from the east coast would only be a factor if transported into the upper troposphere prior to reaching Europe. Further analysis is needed to separate those air masses ascending from the east coast from those that passed over at a higher altitude.

The number of hours the air mass in the north Atlantic flight corridor was chosen as an indicator of aircraft as a potential source of LAC over Europe. The negative correlations with LAC concentration and mass equivalent diameter and positive correlation with the LAC mass fraction seem contradictory. If the aircraft emissions are relatively fresh, then we would expect smaller mass diameters and probably a higher mass fraction; however, fresh emissions should also increase the number concentration, not decrease it as observed. As a result, these comparisons, as currently derived, do not show aircraft emissions as a significant source of LAC particles over Europe.

4. Summary

The properties of LAC particles, when sufficiently far from local sources, are hypothesized to be related to the history of the air mass in which they are found. The measurements from five flights in five days is a very limited data set and may not be statistically representative of the LAC in the upper troposphere over Europe; however, given that the LAC mass concentrations were consistent with measurements from previous experiments, in other regions of the world, made with other instruments by other investigators, we can assume that the present measurements are not atypical, even with the observed variations.

The meridional trends of average LAC properties and the associated variances are perhaps only representative of this time of the year; however, the statistically significant correlations observed with certain characteristics of the air masses in which they are found are consistent with biomass burning in Canada as a primary source of particles that contain LAC. The eastern coast of the US also seems to be a potential source of LAC particles but the current analysis produces ambiguous results that will have to be resolved with additional stratification of the back trajectories.

These measurements have provided further insight into how LAC particles are transported and transformed in the UT but underscore the need for much more detailed measurements with a wider set of microphysical and chemical instrumentation to better evaluate the LAC properties and their impact on the regional and global climate. To paraphrase one of the conclusions of the latest report of the IPCC (Forster and Ramaswamy *et al* 2007), there is no consensus as to the impact on climate of LAC with respect to CO_2 ('efficacy') and this may represent problems with the stratospherically adjusted definition of radiative forcing. Measurements such as those presented here will help to constrain the range of LAC that is predicted by global aerosol models that in turn can be used to better parameterize the LAC aerosol optical properties in global climate models. At the same time, further work is needed to put these measurements into the context of aerosol lifetime and the relative importance of different processes for removing LAC from the atmosphere.

Acknowledgments

The authors would like to thank Rolf Maser and Dieter Schnell of Enviscope for their excellent support during the project, the flight crews who flew and maintained the aircraft, and Peter Spichtinger of ETH. We would also like to thank Nicole Spelten and Reimar Bauer of FZ Jülich, who processed the measurements used in the derivation of the ice water content.

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