Aircraft observations of the upper tropospheric fine particle aerosol in the Northern and Southern Hemispheres at midlatitudes

Andreas Minikin,[1] Andreas Petzold,[1] Johan Ström,[2] Radovan Krejci,[2] Marco Seifert,[2] Peter van Velthoven,[3] Hans Schlager,[1] and Ulrich Schumann[1]

Received 16 October 2002; revised 7 February 2003; accepted 14 February 2003; published 20 May 2003.

[1] As part of the project INCA (Interhemispheric Differences in Cirrus Properties from Anthropogenic Emissions) two aircraft field campaigns have been performed to study aerosol and cirrus cloud properties of the upper troposphere (UT) in the midlatitudes of the southern hemisphere (SH) and the northern hemisphere (NH). This paper focuses on the measurements of UT number concentrations and tropospheric vertical profiles of volatile and refractory Aitken as well as total accumulation mode particles. The results are discussed with respect to interhemispheric differences. Aerosol number concentrations in the NH are more variable and typically a factor of 2–3 higher. The SH data considerably extend the few existing UT *in situ* aerosol measurements in the SH midlatitudes. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry. **Citation:** Minikin, A., A. Petzold, J. Ström, R. Krejci, M. Seifert, P. van Velthoven, H. Schlager, and U. Schumann, Aircraft observations of the upper tropospheric fine particle aerosol in the Northern and Southern Hemispheres at midlatitudes, *Geophys. Res. Lett.*, *30*(10), 1503, doi:10.1029/2002GL016458, 2003.

1. Introduction

[2] Natural and man-made emissions of gaseous aerosol precursors, transported into the upper troposphere (UT) by synoptic scale lifting of air masses or deep convective processes, constitute the main source of UT aerosols. An additional source is the emission of particles and gaseous aerosol precursors from aircraft in the tropopause (TP) region. Despite the recognized importance in global climate the global distribution of UT aerosols and their microphysical, chemical and radiative properties are not well known, because the regional and temporal variability of aerosol occurrence is large and observational data are sparse [*Raes et al.*, 2000]. *In situ* measurements of UT aerosol properties, from aircraft and balloon platforms, are available from a number of experiments in the NH and tropical regions, partly extending into the SH subtropics (see Section 3). For the midlatitudes of the SH, however, with very few exceptions, experimental data were missing to date. To address this deficiency in observational data for UT properties of both, background aerosol and cirrus clouds, and to assess the impact of anthropogenic emissions on cirrus cloud properties, the project INCA (Interhemispheric Differences in Cirrus Properties from Anthropogenic Emissions) was designed to include two aircraft campaigns, one in each hemisphere. The two campaigns were performed within the same year (2000), at the same relative latitudes (50–60°), with the same instrumentation and in the same local season (autumn). In this paper, we focus on the interhemispheric differences in the UT "background" aerosol outside of clouds as found from the INCA experiments.

2. Flight Routes, Instruments and Data Selection

[3] The aircraft used in the INCA experiments, the DLR Falcon 20 (~13 km ceiling), was operated in the SH campaign out of Punta Arenas (53°S, Chile, March/April 2000) and in the NH campaign out of Prestwick (55.5°N, Scotland, Sept./Oct. 2000), see Figure 1. Both campaigns lasted for a 3–4 weeks time period in equivalent local season (autumn) comprising 9–10 measurement flights and 30–40 hours total flight time each. Between the time periods investigated, there was no interference of the UT background conditions by any major volcanic eruption or El Niño event.

[4] The aircraft carried an extensive set of particle and cloud physical properties related instruments (see http://www.pa.op.dlr.de/inca), of which only the ones relevant for this study will be introduced briefly in the following: A set of condensation particle (CN) counters (CPCs, TSI models 3760 and 3010) modified for aircraft use and set to different lower cut-off sizes was operated to obtain the total number concentrations of Aitken (>14 nm particle diameter) and ultrafine particles (>5 nm). One CPC was operated with its inlet line heated to 250°C to allow for determination of non-volatile (refractory) particle (>10 nm) fractions. The CPCs were connected to a sideward facing aerosol inlet (estimated upper size limit ~1 μm) in order to prevent from sampling artifacts caused by cirrus crystals entering the inlet. Accumulation mode particles (~0.1–1 μm) were measured in dry state with a wing-mounted PMS PCASP-100X [*Petzold et al.*, 2002]. To prevent cloud particles from entering the PCASP, a custom-made impaction plate was mounted inside the PCASP inlet leading to a modified size range of ~0.12–0.9 μm instead of nominally 0.12–3.5 μm. During the transfer flights to Chile and back (Figure 1) the aerosol instrumentation had to be operated in a configuration different from that described above. During the northbound flights the CPCs were operative but not the wing-mounted PCASP. During the southbound flights a different set of CPCs (volatile CN >6 nm, refractory CN

[1]Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany.
[2]Institute of Applied Environmental Research, Stockholm University, Stockholm, Sweden.
[3]Royal Netherlands Meteorological Institute (KNMI), Atmospheric Composition Research, De Bilt, Netherlands.

0094-8276/03/2002GL016458$05.00

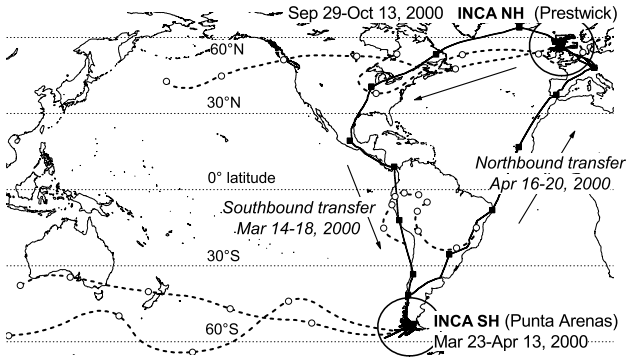

Figure 1. Flight routes and areas of operation of the DLR Falcon during the two INCA campaigns in March/April 2000 (SH) and Sept./Oct. 2000 (NH). Included are illustrative 5-day back trajectories, with endpoints at flight altitudes of 11–12 km.

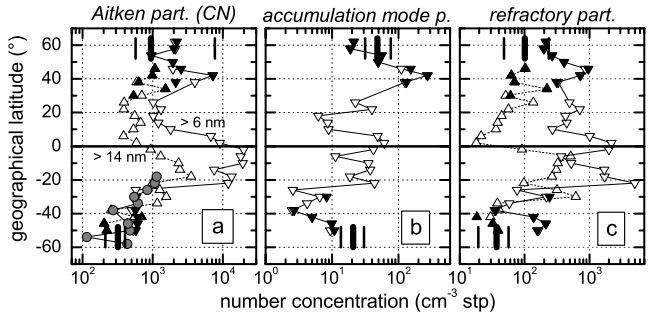

Figure 2. Meridional number concentration distribution of Aitken (a), accumulation mode (b) and refractory particles (c) in the UT during INCA transfer flights in March 2000 (southbound, ∇) and April (northbound, Δ), based on medians of 4° latitude bins for altitudes above 7 km. Included are the INCA SH and NH campaign medians, 25- and 75-percentiles (vertical lines) and data of the PEM TROPICS A experiment (see text, round symbols). Owing to the TP height being systematically higher at low latitudes, filled symbols indicate the subset of data which lie in the UT, here defined as the 3000 m layer below TP.

>18 nm) and a cabin-mounted optical particle counter, also a PCASP, were connected to a second, different aerosol inlet.

[5] The data set used in this study was carefully selected to represent cloud-free and UT air only. In-cloud situations were excluded using crystal density measurements of the FSSP-300 probe or the CVI system [*Gayet et al.*, 2002]. Stratospheric influences were omitted by excluding air masses with ozone mixing ratios above 75 ppbv. 5-day back trajectories were calculated for every 2 min of flight from the KNMI model using ECMWF analyzed wind data. TP heights were determined from ozone measurements and from potential vorticity maps (KNMI model). The number concentrations reported in this paper are calculated for standard conditions (denoted "stp") of 1013 hPa and 273 K.

3. Results and Discussion

3.1. Meridional Aerosol Distribution

[6] Figure 2 depicts the meridional cross sections of the aerosol number concentrations in the UT obtained during the transfer flights from Germany to Punta Arenas and back. Comparing these data has a few limitations: The measurements obviously represent the situation only on the days of flight. The southbound and northbound flights followed different routes (Figure 1), were 1 month apart, and utilized different instruments (Section 2). When inspecting the results of the two flight routes separately, one finds with respect to the midlatitude UT that number concentrations of Aitken, accumulation mode and refractory particles are consistently lower in the SH compared to the NH. Although the transfer flights do not link the same local season in the NH (spring) and SH (autumn) this interhemispheric difference agrees quite well with the INCA SH and NH campaign medians (see Section 3.2) also indicated in Figure 2. The pronounced decrease in UT CN number concentrations from the subtropics to midlatitudes found in the SH is remarkably similar to results of the 1996 PEM TROPICS A campaign [*Clarke and Kapustin*, 2002] (Figure 2). The CN data (>9 nm) used in this comparison (NASA GTE data archive at http://www-gte.larc.nasa.gov) represent several flights performed in the Easter Island region and south from there up to ~60°S latitude, a region ~4000 km west and upstream from the UT over Chile. In the tropics aerosol number concentrations above 7 km altitude increase by one order of magnitude (Figure 2). These elevated concentrations occur where 3-dimensional back trajectories originate from the central South American continent (examples in Figure 1) and have undergone systematic lifting by at least 100–300 hPa. The very high fraction of refractory particles, up to 50–60 % of total CN, may indicate a strong contribution of continental ground sources. Elevated aerosol concentrations in the tropics have been observed before [e.g., *Clarke et al.*, 1999; *de Reus et al.*, 2001]. They can result from intense convection over land, transporting both particles and aerosol precursor gases aloft, and from aerosol nucleation associated with convective clouds.

3.2. UT Aerosol: the NH Versus the SH

[7] A more representative view on UT aerosol number concentrations than from the meridional cross sections (Section 3.1) can be obtained from the statistics of the two 3–4 week midlatitude campaigns in the SH and NH (Table 1, frequency distributions in Figure 3). NH versus SH midlatitude median concentration ratios are in the order of 2–4. Highest ratios are observed for the smaller, Aitken and ultrafine particles, together with higher variability (10- and 90-percentiles in Table 1). Extending this analysis to the full data set including cloud passages does not change the overall statistical results essentially. The larger concentration ratio as

Table 1. Comparison of Aerosol Number Concentrations of the Midlatitude UT in the NH and SH During INCA

	Particle Number Concentration (cm^{-3} stp)				
	SH		NH		
Aerosol Category	Median	Range	Median	Range	NH/SH Ratio
Ultrafine (>5 nm)	350	180...830	1400	450...15000	4.1
Aitken (>14 nm)	240	130...400	770	290...9600	3.1
Accum. mode	17	6...34	40	16...90	2.4
Refractory	37	12...75	105	24...840	2.8

The UT is here defined as the 2000 m layer below the TP. The range is defined by the 10- and 90-percentiles. To obtain approximate ambient concentrations divide by a factor of 3.

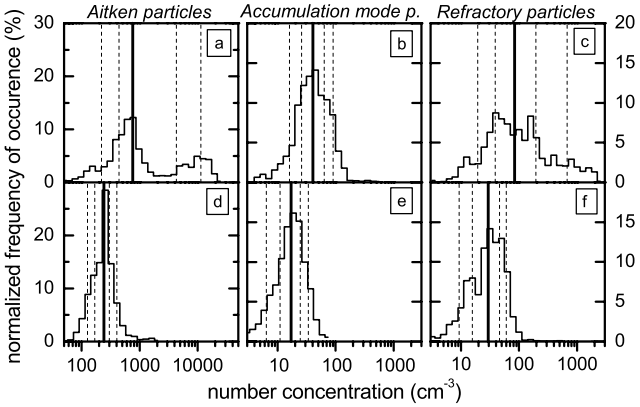

Figure 3. Frequency distributions of UT aerosol number concentrations of the NH (a–c) and SH (d–f). Median, 10-, 25-, 75- and 90-percentiles are indicated by vertical lines.

well as the higher variability of concentrations in the NH observed for smaller particles is partly a consequence of UT particle nucleation events being more frequent during the INCA measurements in the NH (not detailed here). The spatial distribution of (ultra)fine particles is more inhomogeneous and connected to recent, locally or regionally confined pollution events which occurred in the NH but not in the SH. This was in particular evident during two flights on October 12, 2000, where very high UT concentrations of Aitken particles (>10^4 cm^{-3}, contributing to the secondary maximum in the frequency distribution in Figure 3a) were clearly related to strong convection over the North Sea region.

[8] Analysis of 5-day back trajectories for the SH campaign revealed that UT air originated almost always from very far west without major changes in altitude, indicating that the UT air gets very effectively zonally transported within the UT at SH midlatitudes. In the NH the circulation is also dominated by zonal transport from west to east, but in some cases connected with an upward transport of boundary layer air potentially carrying pollutants from the North American continent. Comparing to the SH situation, this enhanced variability of transport patterns (zonal and vertical) in the NH may contribute to the larger range of number concentrations of all aerosol types observed in the NH (Figure 3).

[9] Other quite extensive data sets on volatile CN concentrations of the NH midlatitude UT have been obtained from measurements on commercial airliners: *Detwiler et al.* [2000] found CN concentrations in the UT of 250–1000 cm^{-3} (North Atlantic, 1977–1979) which agrees well with the INCA observations. *Hermann et al.* [2002] report on significantly higher NH midlatitude concentrations (Europe/Mediterranean, 1997–2000) but discuss that their data might be biased towards higher values because their measurements were made along major flight routes. The INCA NH measurements were made partly close, but not directly inside the North Atlantic Flight Corridor. An enhancement of CN background concentration in flight corridors was previously found to occur only in the direct vicinity of the flight routes and estimated to be below 200 cm^{-3} [*Anderson et al.*, 1999; *Schröder et al.*, 2000]. Therefore we expect the INCA NH values, and consequently the NH/SH concentration ratios (Table 1) to be reasonably representative of the background situation in the Atlantic/European sector of the NH. This is further supported, for example, by observations of mean CN concentrations of 770–970 [*Petzold et al.*, 2000] and 250–1000 cm^{-3} stp [*Heintzenberg et al.*, 1991] in the tropopause region over northern Europe. In the Pacific region *Clarke and Kapustin* [2002] show that in comparison to the SH volatile CN are similar or even lower in the NH whereas refractory particles are enhanced by a factor of two in the midlatitudinal free troposphere (FT) above 3 km. *Rosen et al.* [1997] find a similar aerosol background in balloon-borne backscattersonde profiles at two sites at 41°N (Wyoming) and 45°S (New Zealand). Though the measurements are few and difficult to compare, this may indicate that midlatitude interhemispheric differences in the UT aerosol load are not zonally uniform around the globe.

[10] While the INCA measurements compared the same local season in both hemispheres, earlier studies give some hints on the possible seasonality of the UT aerosol: For optically active particles *Kristament et al.* [1993] and *Liley et al.* [2001] find in the tropical dry season (July–September) enhanced aerosol abundance in the FT of the SH. *Rosen et al.* [1997] find weaker seasonal variations for the midlatitude UT, both in the SH and the NH, with higher aerosol backscatter in the local winter/spring half year. For Aitken particles *Hermann et al.* [2002] report on a two times lower concentration in winter north of 42°N. For the SH the seasonality of CN remains unknown. Overall, the seasonality of UT aerosol is not well established from *in situ* observations, but it appears that local autumn, the time of the INCA measurements, does not represent an extreme seasonal case.

3.3. Vertical Aerosol Profiles

[11] The composite vertical aerosol distributions presented in Figure 4 point out the different role of aerosol surface sources in both hemispheres. Aerosol components in the SH show all weak vertical gradients in the FT indicating that the troposphere of the SH is well mixed above ~2 km and that aerosol on average will be relatively aged. This is consistent with the expected lack of major surface sources of aerosol and gaseous precursors in the SH [*Rosen et al.*, 1997]. In the NH, in contrast to the SH, accumulation mode and refractory particles show a distinct and steady decrease from the surface towards the UT. Therefore, concentration differences between both hemispheres as discussed in Section 3.2 are even more pronounced for the middle and lower troposphere. This type of vertical profile reflects qualitatively the minor importance of UT sources versus surface sources for accumulation mode and refractory particles. For the Aitken particles the NH vertical profile is characterized by a relative maximum in the upper FT as observed and discussed previously [e.g., *Singh et al.*, 2002; *Schröder et al.*, 2002; *Clarke and Kapustin*, 2002]. Figure 4 includes the composite vertical profiles obtained during the LACE 98 campaign in summer at a continental site in Germany [*Petzold et al.*, 2002] showing a good overall agreement between the observed vertical distributions given the differences in season and measurement region.

4. Summary and Conclusions

[12] This study based on aircraft measurements made with identical instrumentation during the INCA experiments in the SH and NH aimed at gaining insight into interhemi-

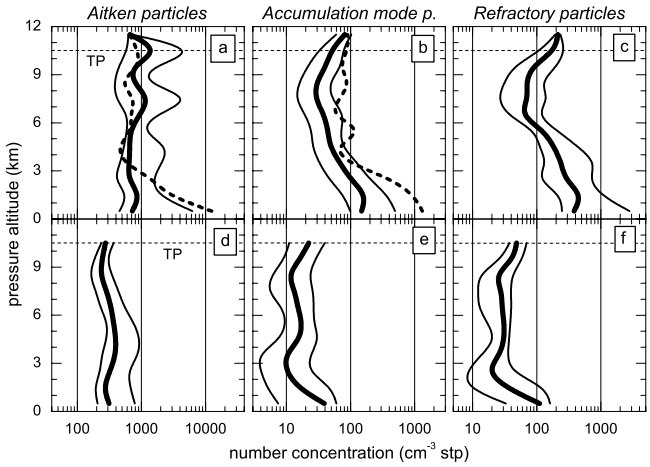

Figure 4. Tropospheric vertical profiles (median, 25- and 75-percentiles) of aerosol number concentrations in the NH (a–c) and SH (d–f) based on binning into 1000 m altitude intervals. The pressure altitude scale of each flight was normalized in order to match the actual TP heights to the mean TP height of 10.5 km (valid for both INCA campaigns).The dotted lines refer to data of the LACE 98 campaign (see text).

spheric differences in UT aerosol properties. As expected, the midlatitudes (50°–60°) of the SH (South America) proved to be a pristine region, with UT number concentrations of Aitken, refractory and accumulation mode particles being on average a factor of 3.1, 2.4 and 2.8, higher in the NH (eastern North Atlantic). Vertical aerosol profiles differ significantly between both hemispheres, most likely because of larger anthropogenic emissions in connection to more widespread convection processes [*Wang et al.*, 2000] and larger land mass in the NH.

[13] Refractory aerosol particles in the UT may initiate cirrus formation via heterogenous freezing processes. Their ambient concentration was found to be in the order of 35 cm^{-3} in the NH and 12 cm^{-3} in the SH (converted from Table 1). This is a fairly high number concentration, compared to the typical ice crystal densities in cirrus clouds of 0.5–5 cm^{-3} observed during INCA [*Gayet et al.*, 2002]. Therefore, the role of heterogenous freezing in cirrus formation, in particular in the NH, remains to be further investigated.

[14] The model results of *Raes et al.* [2000] show midlatitude UT ambient concentrations (at 250 hPa in July) of ~50 (SH) and ~100 cm^{-3} (NH) for accumulation mode and ~10^4 cm^{-3} (SH and NH) for Aitken mode aerosol. As *Raes et al.* [2000] discuss, the model appears to over-predict Aitken mode particle concentrations in the UT. The observations made during INCA may not represent UT zonal means, but they show notably lower number concentrations than the simulation, even in the NH, where variability was largest. Compared to our results in Table 1 the model overestimates aerosol number concentrations by an order of magnitude, not only for Aitken but also for accumulation mode particles. This shows that global models need further improvement and observational validation to accurately predict the concentration level and large-scale distribution of aerosols in the UT.

[15] **Acknowledgments.** This work was partially supported by the Commission of the European Community (contract no. EVK2-CT-1999-00039). We thank H. Rüba, M. Fiebig, P. Stock, J. Baehr for essential help with instrumentation and data, and B. Kärcher for helpful comments on the manuscript. Excellent field support was provided by R. Busen and the flight department of DLR.

References

Anderson, B. E., et al., An assessment of aircraft as a source of particles to the upper troposphere, *Geophys. Res. Lett.*, *27*, 369–372, 1999.

Clarke, A. D., and V. N. Kapustin, A Pacific aerosol survey. part I: A decade of data on particle production, transport, evolution, and mixing in troposphere, *J. Atmos. Sci.*, *59*, 363–382, 2002.

Clarke, A. D., et al., Nucleation in the equatorial free troposphere: Favorable environments during PEM-Tropics, *J. Geophys. Res.*, *104*, 5735–5744, 1999.

de Reus, M., H. Fischer, R. Krejci, R. Scheele, J. Ström, and J. Williams, Vertical and horizontal distribution of the aerosol number concentration and size distribution over the northern Indian Ocean, *J. Geophys. Res.*, *106*, 28,629–28,641, 2001.

Detwiler, A. G., L. R. Johnson, and A. G. Schauer, Exploratory analysis of the distribtution of condensation nuclei in the Northern Hemisphere upper troposphere and lower stratosphere during the late 1970s, *J. Geophys. Res.*, *105*, 9265–9282, 2000.

Gayet, J.-F., F. Auriol, A. Minikin, J. Ström, M. Seifert, R. Krejci, A. Petzold, G. Febvre, and U. Schumann, Quantitative measurement of the microphysical and optical properties of cirrus clouds with four different in situ probes: Evidence of small ice crystals, *Geophys. Res. Lett.*, *29*(24), 2230, doi:10.1029/2001GL014342, 2002.

Heintzenberg, J., J. Ström, J. A. Ogren, and H.-P. Fimpel, Vertical profiles of aerosol properties in the summer troposphere of central Europe, Scandinavia and the Svalbard region, *Atmos. Environ., Part A*, *25*, 621–627, 1991.

Hermann, M., J. Heintzenberg, A. Wiedensohler, A. Zahn, G. Heinrich, and C. A. M. Brenninkmeijer, Meridional distributions of aerosol particle number concentrations in the upper troposphere and lower stratosphere obtained by CARIBIC flights, *J. Geophys. Res.*, *108*(D3), 4114, 10.1029/2001JD001077, 2002.

Kristament, I. S., M. J. Harvey, and J. B. Liley, A seasonal cycle in the southwest Pacific free tropospheric aerosol concentration, *J. Geophys. Res.*, *98*, 16,829–16,837, 1993.

Liley, J. B., J. M. Rosen, N. T. Kjome, N. B. Jones, and C. P. Rinsland, Springtime enhancement of upper tropospheric aerosol at 45S, *Geophys. Res. Lett.*, *28*, 1495–1498, 2001.

Petzold, A., C. Hoell, B. Kärcher, J. Beuermann, C. Schiller, H. Ziereis, and H. Schlager, In situ observations of aerosol properties above ice saturation in the polar tropopause region, *J. Geophys. Res.*, *105*, 29,387–29,395, 2000.

Petzold, A., M. Fiebig, H. Flentje, A. Keil, U. Leiterer, F. Schröder, A. Stifter, M. Wendisch, and P. Wendling, Vertical variability of aerosol properties observed at a continental site during LACE 98, *J. Geophys. Res.*, *107*(D21), 8128, doi:10.1029/2001JD001043, 2002.

Raes, F., R. Van Dingen, E. Vignati, J. Wilson, J.-P. Putaud, J. H. Seinfeld, and P. Adams, Formation and cycling of aerosols in the global troposphere, *Atmos. Environ.*, *34*, 4215–4540, 2000.

Rosen, J. M., N. T. Kjome, and J. B. Liley, Tropospheric aerosol backscatter at a mitlatitude site in the Northern and Southern Hemispheres, *J. Geophys. Res.*, *102*, 21,329–21,339, 1997.

Schröder, F., C. A. Brock, R. Baumann, A. Petzold, R. Busen, P. Schulte, and M. Fiebig, In situ studies on volatile jet exhaust particle emissions: Impact of fuel sulfur content and environmental conditions on nuclei mode aerosol, *J. Geophys. Res.*, *105*, 19,941–19,954, 2000.

Schröder, F., B. Kärcher, M. Fiebig, and A. Petzold, Aerosol states in the free troposphere, *J. Geophys. Res.*, *107*(D21), 8126, doi:10.1029/2000JD000194, 2002.

Singh, H. B., et al., Global distribution and sources of volatile and nonvolatile aerosol in the remote troposphere, *J. Geophys. Res.*, *107*(D11), 4121, doi:10.1029/2001JD000486, 2002.

Wang, Y., et al., Evidence of convection as a major source of condensation nuclei in the northern midlatitude upper troposphere, *Geophys. Res. Lett.*, *27*, 369–372, 2000.

R. Krejci, M. Seifert, and J. Ström, Institute of Applied Environmental Research, Stockholm University, Stockholm, Sweden.

A. Minikin, A. Petzold, H. Schlager, and U. Schumann, DLR, Institut für Physik der Atmosphäre, Oberpfaffenhofen, Wessling, 82234 Germany. (andreas.minikin@dlr.de)

P. van Velthoven, Royal Netherlands Meteorological Institute (KNMI), Atmospheric Composition Research, De Bilt, Netherlands.