

**Aerosols in the  
tropical and  
subtropical UT/LS**

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# Aerosols in the tropical and subtropical UT/LS: in-situ measurements of submicron particle abundance and volatility

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## Abstract

Processes occurring in the tropical upper troposphere and lower stratosphere (UT/LS) are of importance for the global climate, for the stratospheric dynamics and air chemistry, and they influence the global distribution of water vapour, trace gases and aerosols. The mechanisms underlying cloud formation and variability in the UT/LS are of scientific concern as these still are not adequately described and quantified by numerical models. Part of the reasons for this is the scarcity of detailed in-situ measurements in particular from the Tropical Transition Layer (TTL) within the UT/LS. In this contribution we provide measurements of particle number densities and the amounts of non-volatile particles in the submicron size range present in the UT/LS over Southern Brazil, West Africa, and Northern Australia. The data were collected in-situ on board of the Russian high altitude research aircraft M-55 "Geophysica" using the specialised COPAS (COndensation PARticle counting System) instrument during the TROCCINOX (Araçatuba, Brazil, February 2005), the SCOUT-O3 (Darwin, Australia, December 2005), and SCOUT-AMMA (Ouagadougou, Burkina Faso, August 2006) campaigns. The vertical profiles obtained are compared to those from previous measurements from the NASA DC-8 and NASA WB-57F over Costa Rica and other tropical locations between 1999 and 2007. The number density of the submicron particles as function of altitude was found to be remarkably constant (even back to 1987) over the tropical UT/LS altitude band such that a parameterisation suitable for models can be extracted from the measurements. At altitudes corresponding to potential temperatures above 430 K a slight increase of the number densities from 2005/2006 results from the data in comparison to the 1987 to 2007 measurements. The origins of this increase are unknown. By contrast the data from Northern hemispheric mid latitudes do not exhibit such an increase between 1999 and 2006. Vertical profiles of the non-volatile fraction of the submicron particles were also measured by a COPAS channel and are presented here. The resulting profiles of the non-volatile number density fraction show a pronounced maximum of 50% in the tropical TTL over Australia and West Africa. Below

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and above this fraction is much lower attaining values of 10% and smaller. In the lower stratosphere the fine particles mostly consist of sulphuric acid which is reflected in the low numbers of non-volatile residues measured by COPAS. Without detailed chemical composition measurements the reason for the increase of non-volatile particle fractions cannot yet be given. The long distance transfer flights to Brazil, Australia and West-Africa were executed during a time window of 17 months within a period of relative volcanic quiescence. Thus the data measured during these transfers represent a “snapshot picture” documenting the status of a significant part of the global UT/LS aerosol (with sizes below  $1\ \mu\text{m}$ ) at low concentration levels 15 years after the last major (i.e., the 1991 Mount Pinatubo) eruption. The corresponding latitudinal distributions of the measured particle number densities are also presented in this paper in order to provide input on the UT/LS background aerosol for modelling purposes.

## 1 Introduction

In the upper troposphere and lower stratosphere (UT/LS) aerosols influence the radiative budget and the actinic fluxes for photochemical reactions, serve as condensation nuclei for altocumulus and cirrus cloud formation (Krämer et al., 2009), and provide sites for heterogeneous chemical reactions (e.g., Borrmann et al., 1997).

Of particular importance for many of the aerosol and cloud related processes in the tropics is the so-called Tropical Transition Layer (TTL) within the UT/LS. It is defined as the altitude band between an upper “boundary level” somewhat above the thermal tropopause and the level of net zero radiative heating as lower “limiting surface” (Fueglistaler et al., 2009). This lower boundary often also is close to the level of neutral buoyancy where convection – with the exception of occasional overshooting – does not suffice to lift air parcels any higher. Thus the TTL is characterised by slow radiatively driven vertical ascent in contrast to the highly convective and dynamically active upper troposphere directly below. In the recent review paper by Flueglistaler et al. (2009) a “synthesis definition” for the TTL is given by specifying a lower TTL “boundary” level

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of  $\Theta=355$  K potential temperature (equivalent to 150 hPa ambient pressure, or 14 km of altitude) and a TTL top level with the corresponding values of 425 K, 70 hPa, and 18.5 km. Since the tropical tropopause typically is located at potential temperatures  $\Theta \approx 380$  K, or roughly 16 to 18 km altitude, it is situated within the TTL and does not constitute the upper TTL “boundary”. The latitudinal boundaries of the TTL are given by the subtropical jets and thus the TTL can be considered as sub-volume of the UT/LS region.

The aerosols in the tropical upper troposphere and TTL most likely contribute significantly to the particles of the global stratospheric Junge layer (Brock et al., 1995), and may influence the transport of water vapour into the stratosphere (Corti et al., 2008). Water vapour contained in the upwelling air is removed in significant amounts within the TTL through freeze drying in thin visible, subvisible and opaque cirrus. This is the basis of the stratospheric water vapour tape recorder phenomenon (Mote et al., 1996; Randel et al., 2001; Randel et al., 2004). The formation of cirrus and subvisible cirrus is dependent on the properties of the aerosol (Thomas et al., 2002; Peter et al., 2003; Luo et al., 2003; Stefanutti et al., 2004) although the exact mechanisms leading to nucleation and cloud formation in the TTL still are unknown (Froyd et al., 2009a). From in-situ cloud and submicron aerosol particle data De Reus et al. (2009) showed that in the tropical upper troposphere typically one out of 3000 aerosol particles can be activated to form an ice particle with occasional values as high as one of 300 and as low as 1/30 000. In a very recent publication Froyd et al. (2009b) present air-borne mass spectrometric composition measurements of ice residues from tropical cirrus and subvisible cirrus clouds. They conclude that most ice residuals consisted of internal mixtures of neutralised sulphate and some organics (similar to liquid sulphuric acid droplets) and that mineral dust or other heterogeneous nuclei do not play a role for cirrus formation.

According to current belief, the major source for aerosol particles in the lower stratosphere is gas-to-particle conversion of precursor gases like OCS, CS<sub>2</sub>, and SO<sub>2</sub> ultimately forming binary sulphuric acid/water droplets (Thomason and Peter, 2006;

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Wilson et al., 2008). According to Brock et al. (1995) fine and ultrafine particles present or formed in the tropical upper troposphere can be transported into the stratosphere and recent evidence suggests that organic components may be involved here also (Ekman et al., 2008; Froyd et al., 2009a). This is supported by mass-spectrometric in-situ measurements from Murphy et al., 1998, who found that “organic material was almost always present” in the UT/LS particles. In addition, the UT/LS particle mass spectra from Murphy et al. (1998) indicated the presence of metals like iron, magnesium, and even mercury. Also Nielsen et al. (2007) detected solid particles of unknown origin in the tropical lower stratosphere. A study based on numerical simulation of observed in-situ data indicate that ion induced nucleation in the UT/LS facilitates gas to particle conversion for the  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  system (Lee et al., 2003; Lovejoy et al., 2004) and thus may be an important source of UT/LS aerosols. According to Lee et al. (2004) inside tropical upper tropospheric cirrus clouds new particle nucleation events can occur. However, the significance of this source for the global UT/LS aerosol and the underlying mechanisms are still unknown. Based on recent laser ablation mass spectrometric in-situ measurements in the TTL over Costa Rica, Froyd et al. (2009a) showed that significant numbers of particles with sizes above 500 nm contain oxidised organic compounds. They also conclude that the chemical composition of these larger particles in the tropical TTL depends on tropical dynamics, regional sources (implying boundary layer processes as well) and the occurrence of overshooting cumulonimbus clouds. Furthermore, evidence has been presented by Popp et al. (2006) and Voigt et al. (2008) suggesting that nitric acid trihydrate (NAT) particles are present in the tropical transition layer. Based on numerical simulations of the microphysics and thermodynamics and on global modelling, it was hypothesised that these may even form a “tropical tropopause NAT belt” although evidence for this is still lacking.

A comprehensive, recent overview of the mechanisms underlying stratospheric aerosol microphysics and chemistry, as well as observations, volcanic influence, climatology, trends, and modelling of stratospheric aerosols including references to the relevant literature were presented by Thomason and Peter (2006) in the WRCP SPARC

Assessment Report No. 4. Further detailed information is available in Chapter 3 of WMO (1999).

Summarising, the aerosols in the upper troposphere and lower stratosphere seem to originate 1.) from gas-to-particle conversion processes, possibly involving organic substances, and possibly influenced by ions induced from cosmic rays, as well as 2.) from lifting processes (like deep convection and overshooting convection) which transport gases and particulate materials from the boundary layer to the UT/LS. 3.) Cirrus clouds also may play a role as sources for submicron particles. 4.) The aerosol present in the tropical UT may form a reservoir of particles amenable for transport into the lower stratosphere. 5.) With respect to the chemical composition of the aerosol particles, sulphuric acid-water solutions are assumed to be the major components in the lower stratosphere while more complex composition including organic materials may be found in the TTL. Also metals, possibly soot and biomass burning residues can play a role for the chemical composition.

It is quite clear that the mechanisms controlling particle production, ageing and composition and also the global spatial and temporal distribution of the sources are to a large extent unknown or at least un-quantified for the tropical UT/LS and in particular the TTL. Obviously this short (and incomplete) survey of the recently gained knowledge demonstrates that an increasingly complex picture evolves with respect to the microphysics and dynamics of the tropical UT/LS and in particular the TTL aerosols. This implies considerable difficulties when it comes to numerical simulation and the need for more detailed measurements is evident.

The aims of this study are: 1.) To provide in-situ data for submicron particle number densities at altitudes above 14 km during the time of relative volcanic quiescence (here: from January 2005 until August 2006) although at the end of this period a small eruption played a role. The data include, in addition to mission flights, transfer flights of the Russian M-55 high altitude research aircraft “Geophysica” from Europe to Brazil (TROCCINOX, 2005), to Northern Australia (SCOUT-O3, 2005), and to West Africa (SCOUT-AMMA, 2006). 2.) To relate these measurements to 1999–2007 data from

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other tropical locations like Costa Rica and to older data from Hawaii. 3.) To show vertical profile in-situ data of submicron particle number concentrations up to 20 km altitude from Brazil, West Africa and Northern Australia, and 4.) to present and discuss results from measurements of non-volatile residues.

5 The main instruments used to obtain the data are specialised condensation particle counters (CPCs). For the M-55 “Geophysica” this is the COPAS (CONDensation Particle counter System; see Weigel et al., 2009) while an NMASS instrument was operated by the University of Denver on the NASA DC-8 and NASA WB-57F. Similar instruments were implemented on the German DLR Falcon-20 and a balloon borne  
10 CPC was utilised by the University of Wyoming. The Russian M-55 “Geophysica” and the NASA WB-57F are high altitude research aircraft capable of reaching 21 km and 16 km, respectively, while the balloons ascend to  $\approx 30$  km. Thus in-situ measurements in the tropical UT/LS and TTL could be conducted in Brazil, Costa Rica, Northern Australia and Western Africa.

## 15 2 Methodology and field experiments

### 2.1 Instrumentation for submicron particle measurements

The aerosol number concentration of ambient aerosol particles with size diameters between 6 nm and roughly  $1 \mu\text{m}$  was measured using two independent CONDensation Particle counter Systems (COPAS) installed at two different locations on board of the  
20 M-55 “Geophysica”. Each COPAS unit consists of two CPC channels designed for automated low pressure measurements on a fast moving platform of the particle number concentration (Weigel et al., 2009) utilising Fluorinert (FC-43) as condensable substance. Three of the four channels sample the ambient aerosol but are operated with different internal temperature settings, such that the cut-off diameters (in terms of 50%  
25 detection efficiency) are 6, 10 and 14 nm, respectively. The design of the aerosol inlet system for COPAS implies an upper size limit for the sampled particles of roughly  $1 \mu\text{m}$

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(Weigel et al., 2009). The total aerosol number concentration in  $\text{cm}^{-3}$  for particles with diameters larger than 6, 10 and 14 nm is denoted as  $N_6$ ,  $N_{10}$  and  $N_{14}$  throughout this paper, while the particle mixing ratios (in number of particles per milligram of air) are given in lower case as  $n_6$ ,  $n_{10}$  and  $n_{14}$ . The fourth channel heats the sampled air containing the ambient aerosol particles to  $250^\circ\text{C}$  prior to the detection of the grown droplets. This way volatile ambient particles completely evaporate (or shrink out of the detectable size range of the COPAS instrument) and only particles containing non-volatile residues larger than 10 nm (as 50% detection level cut-off diameter) are counted as number density  $N_{10nv}$  ( $\text{cm}^{-3}$ ) or given as number mixing ratio ( $\text{mg}^{-1}$ )  $n_{10nv}$ . For stratospheric measurements this implies that binary solution droplets of sulphuric acid and water or ternary  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}/\text{HNO}_3$  droplets completely evaporate resulting in zero counts of this channel. Only if, for example, particles from meteoric ablation and re-condensation are contained inside these growing droplets their metallic or mineral core will not evaporate and the residual particles are detected as non-zero counts. Curtius et al. (2005) found this type of particles to be enhanced for inner polar vortex aerosol for the Arctic in 2003 by showing that up to 80% of the detected particles there were of meteoric origin. The number concentration of particles with diameters between 6 and 14 nm ( $N_{6-14}$ ) obviously is available by subtracting  $N_{14}$  from  $N_6$ . These particles are so small that they are assumed to be recently formed in the atmosphere during so-called “nucleation events”.

The reading frequency of count results at the COPAS detectors is 1 Hz. From these 1 Hz data typically 15 s averages are calculated and displayed as running means in the figures below. The counting statistics is such that 1.) at the altitude dependent flow rates through COPAS and 2.) at ambient concentrations as low as 20 particles per cubic centimetre, the sensor detects around 100 counts per second. Only for the heated channel, when particle concentrations as small as  $2\text{ cm}^{-3}$  occur, the generated count rate attains low values of roughly 30 per 15 s. In such (rare) circumstances fluctuations in the data may be due to counting statistics. At the cruise speed of the M-55 “Geophysica” near  $200\text{ m s}^{-1}$  these 15 s flight-time averages correspond to 3 km horizontal

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resolution and for the typical ascent rate of  $10 \text{ m s}^{-1}$  vertical resolution of 150 m results for the vertical profiles. Usually the instrument's control of flow rates and temperature settings attains sufficient stability for correct particle measurements at altitudes higher than 5 km. At lower altitudes the manoeuvring of the M-55 "Geophysica", as for example demanded by Air Traffic Control, often is too fast for the controls to adapt. The measurements gathered under such conditions needed to be removed from the data base. Details on the operation of COPAS and a full experimental characterisation of the individual channels are provided by Weigel et al. (2009). These include descriptions of the calibration procedures, as well as assessments of the pre-heating device efficiencies, of the inlet systems, and the losses within the sampling system. For this characterisation extensive laboratory measurements had been carried out and the resulting correction factors (e.g., for inlet losses) are utilised for the data reduction here also. In addition, aircraft data from inter-comparisons of the different channels during flight are given in Weigel et al. (2009) and consequently the COPAS instrument can be regarded as well characterised by laboratory and aircraft borne experiments.

Also on board of the M-55 "Geophysica" a Multiwavelength Aerosol Scatterometer (MAS; Cairo et al., 2004; Buontempo et al., 2006) was operated, which is a backscatter sonde for in-situ measurements of optical air and aerosol parameters. The MAS measures volume backscatter ratio and depolarization ratio of the air in the vicinity of the aircraft at 532 nm and 1064 nm with a time resolution of 5 s (i.e., roughly 1 km resolution along the flight path). For the backscatter ratio the measurement precision is 5% and the accuracy 0.05. Its data can be used for the detection of cloud and aerosol particle ensembles and based on the polarization measurements, shape, and hence phase, of the optically active particles (i.e., those with size diameters larger than approximately  $0.2 \mu\text{m}$ ) can be discerned.

Measurements of particle concentrations (here  $N_7$  and  $N_5$ ) also were performed on board of the DLR Falcon-20 within the 9 to 12 km altitude band deploying a set of on board CPCs (Weinzierl et al., 2009; Minikin et al., 2003). Other data for this paper originate from balloon-borne in-situ measurements by means of a condensation particle

counter during the SCOUT-AMMA campaign (2006) from Niamey, Niger, Africa. The instrument is described in Rosen and Hofmann (1977). Particle data from the NASA operated DC-8 and the WB-57F high altitude research aircraft measured between 1999 and 2007 at various tropical locations are presented below in conjunction with the M-55 “Geophysica” data. These DC-8 and WB-57F measurements were performed with the CPC (of type NMASS) from the University of Denver (Wilson et al., 1983; Brock et al., 2000).

The ambient temperature, was measured on the M-55 “Geophysica” using a Thermo Dynamic Complex (TDC) probe with an accuracy of 0.5K (Shur et al., 2007), while other relevant parameters as position and true air speed have been adopted from the onboard navigational system UCSE (Unit for Connection with the Scientific Equipment; Sokolov and Lepuchov, 1998).

## 2.2 Field campaign data basis

The measurements presented here originate from local flights of the M-55 “Geophysica” and the German DLR Falcon-20 research aircraft during the following campaigns: TROCCINOX (from Araçatuba, Brazil; Schumann, 2005), SCOUT-O3 (based in Darwin, Australia; Vaughan et al., 2008; Brunner et al., 2009), and SCOUT-AMMA (from Ouagadougou, Burkina Faso; Cairo et al., 2009). Emphasis is also placed on data from the transfer flights from Oberpfaffenhofen, Germany, and Verona, Italy, to the various campaign locations. The flight routes of these transfers are shown together with the intermediate stops in Fig. 1. Table 1 provides the relevant details of the campaigns and transfers. From the websites specified in Table 1 further detailed information concerning the participating research aircraft, instrumentation, and flight patterns can be obtained. In the time frame of the M-55 “Geophysica” transfer flights to Brazil and to Australia the DLR Falcon-20 also travelled at lower altitudes to these campaign locations performing measurements underway. From Fig. 1 it becomes apparent that within the time interval from January 2005 until August 2006 in-situ measurements were conducted from both aircraft in the UT/LS region between Europe on one side and South

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America, Australia, and West Africa on the other side. Thus the background aerosol number concentrations in the altitude band between 9 km and 12 km (DLR Falcon-20) and between 12 km and 17 km (M-55 “Geophysica”) can be documented for the sub-micron particle size range in the UT/LS over a significant part of the globe in times of relative volcanic quiescence. Besides the aerosol measurements a variety of data for other atmospheric variables has been collected en route also.

### 3 Results and discussion

#### 3.1 UT/LS background concentrations for submicron particles

The latitudinal distribution of the measured particle number concentrations  $N_6$ ,  $N_{10}$  from the transfer flights is shown in the upper panels of Fig. 2 for the M-55 “Geophysica” covering altitudes between 12 km ( $\Theta \approx 376$  K) and 18.7 km ( $\Theta \approx 460$  K). The upper left panel contains the measurements obtained during the transfer flights from Europe to Brazil and Australia, while the upper right panel displays those of the flights from Italy to Burkina Faso (for dates and details see Table 1). In addition the results of the  $N_5$  measurements on board of the DLR Falcon-20 are given in the lower panels of Fig. 2 from the transfer flights to and from Brazil and Australia. The potential temperature range covered here extends from 315 K to 355 K for the altitude range between 9 km and 12 km. To provide an overview in Fig. 3 the medians from all measurements derived from 5 degree latitude bands of the M-55 “Geophysica” and DLR Falcon-20 data are summarised where additional DLR Falcon-20 measurements (not shown in Fig. 2) from the transfers to West Africa are visible.

Although the cut off diameters (in terms of 50% detection level) of the various condensation particle counters were set to different values (i.e., 5, 6, 7, and 10 nm for  $N_5$ ,  $N_6$ ,  $N_7$ , and  $N_{10}$ ) the data represent the aged aerosol fraction because either no nucleation events occurred during the flights or nucleation event data were removed. (Usually nucleation events come with particle number densities  $N_6 > 1.5 \cdot N_{14}$ , which

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indicates the presence of many particles larger than 6 but smaller than 14 nm.)

From the data at potential temperature levels above 367 K, and slightly above the tropopause, an increase in particle number densities from values near  $20\text{ cm}^{-3}$  at higher latitudes to values around  $100\text{ cm}^{-3}$  can be seen in Figs. 2 and 3 for the transfer routes. The increase towards the tropics originates from the fact that the M-55 “Geophysica” was mostly flying at constant altitudes which implies that in the northern latitudes stratospheric air was sampled and at lower latitudes air masses closer to the tropopause were encountered. Although there is a difference between the lines representing the M-55 “Geophysica” data in Fig. 3, the values and their latitudinal changes are remarkably close. The number concentrations of 10 to 50 particles per  $\text{cm}^3$  from the M-55 “Geophysica” flight across the Saharan desert in Fig. 3 fit well into the range of concentrations found at the same latitudes during the other flights. Thus for the times between January 2005 and July 2006 the measured number densities of small particles were fairly homogeneous along the different latitude bands. At lower altitudes within the 315 K to 355 K levels the DLR Falcon-20 data show values between 100 and 500 particles per  $\text{cm}^3$ . Since this is beneath the tropopause it is not surprising that the number densities are significantly higher than above and that the variability – as seen in the lower panels of Fig. 2 – is much larger.

### 3.2 Vertical profiles from transfer and local flights between January 2005 and August 2006 for the latitude band from $55^\circ\text{ N}$ to $20^\circ\text{ S}$

A subset of the  $n_6$ ,  $n_{10}$ ,  $n_{14}$ ,  $n_{10nv}$  vertical profiles obtained from the transfers to and from Darwin and local flights from Darwin and Ouagadougou with 15 s averages is shown in Figs. 4 and 5. A composite plot with medians and percentiles of the vertical profile particle mixing ratio data of the local flights from Darwin (Australia), Araçatuba (Brazil), and Ouagadougou (Burkina Faso) is given in Fig. 6 (left panel). In all figures potential temperature  $\Theta$  in Kelvin has been chosen as vertical coordinate and particle mixing ratios  $n$  are displayed rather than number concentrations  $N$ .

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From Fig. 4 it can be seen that the vertical profiles are roughly separated into two parts: 1.) At altitudes below 380 K there is a “region of large tropospheric variability” in the particle concentrations ranging from  $\approx 100$  particles per  $\text{cm}^3$  to  $\approx 5000 \text{ cm}^{-3}$ . 2.) For altitudes above 380 K the measured values exhibit a steady decrease with rising altitude having much less scatter (“region of stratospheric compactness”). The narrowness of particle mixing ratio in this region is indicated by the small percentile ranges in Fig. 6. The flight trajectory data suggest that in the right panel of Fig. 4 the highest values encountered on 16 November 2005 over Darwin may have been caused by the M-55 “Geophysica” crossing its own contrail (Weigel et al., 2009).

The summary of data shown in the left panel of Fig. 6 encompasses the vertical profiles from Brazil, Burkina Faso, and Australia with the campaign dates given in Table 1 together with a profile from mid-latitudes (purple line) and a reference profile (black line) from Brock et al. (1995). The profile from Brock et al. (1995) contains data from northern and southern hemispheric tropical locations between 1987 and 1994, where the Pinatubo period has been removed. Obviously, between roughly 340 K and 440 K all tropical profiles exhibit remarkable closeness over the 19 years and over the different locations around the globe these data sets cover. At the same time the significant difference between tropical and mid-latitude profiles is evident. Apparently – somewhat in analogy to the global stratospheric Junge layer higher above – a kind of layer of small particles spans the tropical band with maximum mixing ratios between 340 and 360 K. Thus the interpretation given by Brock et al. (1995), of the tropical upper troposphere as a source of stratospheric particles seems to be valid for the global tropical latitude belt and holding over time. Furthermore, when ignoring the long time gaps between the samplings and ignoring large perturbations from the 1991 Mt. Pinatubo eruption in the tropics, the data in Fig. 6 suggest that the characteristics of this particle source remain rather constant with time and background conditions over almost two decades. At altitudes between 440 K and 500 K some differences of significance in the tropical profiles are observed. The origin of these differences remains unclear except for the observations during SCOUT-AMMA in Burkina Faso, where a small volcanic eruption

seems to be the most likely cause (see Sect. 3.4.).

The similarity of the tropical vertical profiles makes it possible to formulate a parameterisation of submicron particle abundance for the use in microphysical, chemical, or global chemistry and transport models. The parameterisation  $n(\theta)$  in Eq. (1) represents the tropical  $n_{10}$  particle background mixing ratios in number of particles per milligram of air as measured with the COPAS systems during the 2005/2006 tropical campaigns excluding the SCOUT-AMMA data (volcanic perturbation) from above 420 K and holds for altitudes between 360 K and 480 K:

$$n(\theta) = c + A \cdot \exp(-\tau \cdot \theta) \quad (1)$$

$$c = 138.04 \text{ mg}^{-1}, \\ A = 8.4038 \cdot 10^{12} \text{ mg}^{-1}, \text{ and} \\ \tau = 0.060044 \text{ K}^{-1}.$$

A parameterisation according to Eq. (1) with different coefficients can be similarly derived from the Brock et al. (1995) data this way this, way representing the time period from 1987 until 1994 and valid between 360 K and 500 K:

$$c = 67.605 \text{ mg}^{-1}, \\ A = 7.2198 \cdot 10^{11} \text{ mg}^{-1}, \text{ and} \\ \tau = 0.053193 \text{ K}^{-1}.$$

The two parameterisations and the comparison to the original profile from Brock et al. (1995) are shown in the right panel of Fig. 6. Apparently the two parameterisations are very close to each other for potential temperatures below 420 K. Above they are in agreement within a factor of 2 and this difference is reflected in the data of Fig. 6. The increase observed at higher altitudes with respect to the data from Brock et al. (1995) is of unknown origin. However the difference in number mixing ratios is small enough that for most practical purposes both sets of parameters can be used in conjunction with Eq. (1).

To place the measurements from the time frame between January 2005 and August 2006 of the SCOUT-O3, TROCCINOX, and SCOUT-AMMA campaigns (see

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Table 1) into a larger context of tropical measurements, Fig. 7 compares the data from Burkina Faso, Brazil and Northern Australia of the 2005/2006 M-55 “Geophysica” deployments with campaigns where the two NASA operated research aircraft WB-57F and DC-8 were utilized. The data include measurements from ACCENT (1999), SOLVE (1999), CrystalFace (2002), SOLVE2 (2003), the AURA Validation Experiment (2004, 2005, 2006) and TC4 (2007). The latitudinal range covered by the data extends from south of 23° N to north of 23° S (with the major part north of 5° S). The M-55 “Geophysica” data fit well into the range of values given by the DC-8 and WB-57F measurements which again demonstrates the uniformity of the tropical particle layer between the continents. Only above West Africa between 370 K and 390 K the mixing ratios seem to be slightly higher than over the other locations. However, the West African profiles during SCOUT-AMMA also significantly deviate towards higher particle number mixing ratios above 430 K. The nature of this deviation is discussed in Sect. 3.4.

### 3.3 UT/LS vertical profiles of submicron particle number densities from mid-latitudes between 1999 and 2009

To assess the variability of the particle number concentrations at mid latitudes the  $N_{10}$  data from COPAS flights on the M-55 “Geophysica” are shown in Fig. 8 as function of pressure altitude and potential temperature for various European locations as well as for a large number of balloon flights from Laramie, Wyoming, USA (see Deshler et al., 2003). The geographic latitudes as well as years of the measurements are indicated as colour code. The measured profiles are remarkably close together above 15 km. This indicates that in the mid latitude stratosphere under background conditions the submicron particle population is under a stable equilibrium regime starting roughly 5 years after the 1991 Mt. Pinatubo eruption. The time between 1999 to 2009 is characterised by an absence of major volcanic eruptions and the resulting extremely small background particle number concentrations were significantly below the levels of the period between the 1982 and 1991 eruptions of El Chichon and Mt. Pinatubo (Borrmann et

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al., 1993; Borrmann et al., 2000; Brock et al., 1993). At the same time between 1999 and 2009 a significant increase of commercial air traffic occurred as the growth rates of revenue passenger miles consistently were near 5 % per year according to IATA data. It had been conjectured that rising commercial air traffic may lead to increases in stratospheric aerosol properties (Hoffman, 1990). Judging from the – admittedly sparse – data sets in Fig. 8 most likely there are no striking increases in total particle number densities despite strong changes in air traffic. If the air traffic influences larger particles in the stratospheric background aerosol these influences may be more subtle.

### 3.4 Volcanic influence on the UT/LS aerosol over West Africa

The particle number mixing ratios in the vertical profiles from West Africa (2006) in Figs. 5, 6 and 7 show significant deviations from the other tropical profiles for altitudes above 430 K. These deviations were not observed in the vertical profiles over Verona, Italy, and Marrakech, Morocco, during the intermediate landings of the transfer flights, when the M-55 “Geophysica” travelled to Burkina Faso (31 July 2006 and 1 August 2006) and back (16 and 17 August 2006, see Fig. 1 and Table 1). Thus this deviation has to be considered as an atmospheric feature, albeit geographically limited to the tropical locations. An intercomparison of the COPAS data measured from the M-55 “Geophysica” over Ouagadougou, Burkina Faso in August 2006, and the balloon borne CPC measurements of the University of Wyoming group over Niamey, Niger, ( $2^{\circ}10'8''$  E longitude,  $13^{\circ}29'11''$  N latitude) on 31 July 2006 is shown in Fig. 9. Considering the different nature of the two platforms (i.e., fast flying aircraft vs. slowly ascending balloon) and the difference in location (Ouagadougou vs. Niamey with 428 km separation) the agreement between the two different instruments has to be regarded as remarkable, at least for the theta levels between 360 and 460 K. Also it follows from these two figures that the increase of particle number mixing ratios above 430 K is detected by the instruments on both platforms and consequently cannot be considered as artefact of the measurements. In the balloon data the rate of increase in mixing ratios with altitude is stronger than for COPAS at theta levels above 450 K.

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Furthermore the balloon data show that the enhanced particle number densities are reaching up to  $\approx 550$  K, forming a broad layer. Taking into account the absence of such an enhancement or layer in the mid-latitude and subtropical profiles over Verona, Italy, and Marrakesh, Morocco, this stratospheric increase seems to be confined to the tropical regions. The most likely cause of these increases is the eruption of the tropical stratovolcano Soufrière Hills on Montserrat Island in the West Indies (16.72 N latitude 62.18 W longitude, 915 m a.s.l.), which occurred on 20 May 2006 (Carn et al., 2007) and roughly two months before performance of the M-55 “Geophysica” and the balloon measurements in West Africa. The amount of sulphur in form of  $\text{SO}_2$  injected into the stratosphere was estimated to be 0.1 Tg(S) based on satellite observations (Carn et al., 2007).

In Fig. 5 the left curve (red points and square symbols) shows the measurements of non-volatile particle mixing ratios from the heated COPAS channel  $n_{10, nv}$ . Above 420 K the absolute number of non-volatile particles is smaller than below and decreases with altitude although the total particle number increases. Thus the ratio of non-volatile to total particle mixing ratios decreases strongly with height. Also, the MAS backscatter sonde reported enhanced backscatter ratio with no depolarization in this altitude region with peak values of 1.3 for the backscatter at 19.3 km. Profiles measured by the balloon borne versions of MAS, which were deployed in Niamey and launched on different balloon missions within the timeframe of the M-55 “Geophysica” flights in West Africa, also regularly detected such increases in backscatter. This supports the interpretation of the aerosols being of volcanic origin because the fraction of volatile (i.e., sulphuric acid like) particles becomes larger with altitude (see also Sect. 3.5 and Fig. 10) and the non-depolarization of the aerosol indicates the presence of spherical (i.e., liquid phase) particles. Although the Soufrière Hills eruption provided the largest single sulphur input into the stratosphere between late 1991 and 2006, its amount was small compared to the 20 Mt of  $\text{SO}_2$  from Mt. Pinatubo (Chapter 3 in WMO, 1999). Prata et al. (2007) showed by means of cloud top temperatures measured from satellite platforms that the overshooting of the eruption plume core from Soufrière Hills reached the stratosphere.

The evolution of the cloud was tracked by satellite (e.g., CALIPSO) measurements of  $\text{SO}_2$  (e.g., <http://earthobservatory.nasa.gov>) also. According to these sources the location of the eruption cloud started to develop patchiness and inhomogeneity three days after the eruption and the air masses containing volcanic  $\text{SO}_2$  moved westward towards Africa confined mostly within the tropical belt. The inhomogeneities in the volcanic aerosol distribution are reflected in the particle mixing ratio data of the vertical profile in Fig. 9. Over Niamey, Niger, above 475 K, values near 1000 particles per milligram of air were encountered on 31 July 2006, while the air masses sampled over Burkina Faso 4–13 days later were different and contained mixing ratios only half as high. Between 375 K and 450 K the instruments show identical values for both locations. From the vertical profile two months after the eruption it can be concluded, that the ageing of the volcanic aerosol had not yet proceeded to the point where sedimentation of the particles from the layer above 18 km enhances the number densities found in the TTL. Similarly, after the Pinatubo eruption it took more than six months (in mid latitudes) until the aerosol at the tropopause had significantly increased (Borrmann et al., 1997; Keim et al., 1996).

### 3.5 Non-volatile components in the tropical UT/LS submicron aerosol

From the values for  $N_{10nv}$  and  $N_{10}$  the non-volatile fraction,  $f$ , of particles which do not evaporate (when heated to  $250^\circ\text{C}$ ) with respect to the total number of ambient particles can be calculated. The results from the tropical data are shown as a function of altitude in Fig. 10 for South America (TROCCINOX, top panel), Northern Australia (SCOUT-O3, middle panel), and for Western Africa (SCOUT-AMMA, lower panel). The measurements cover altitudes in potential temperature from 340 K to 500 K. In the upper and bottom panels of Fig. 10 the measurements of the DLR Falcon-20 are included reaching almost down to the boundary layer at 300 K. The two profiles over Australia and Burkina Faso exhibit a broad maximum of non-volatile ratios,  $f$ , between 360 K and 460 K with a maximum at altitudes of 400 K in both cases. Thus this maximum roughly extends from the tropopause (i.e., including the upper part of the TTL) well into the

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lowermost stratosphere. Such a maximum is not visible in the data from Brazil where the ratio,  $f$ , remains fairly constant at values around 20% between 390 K and 490 K and values near 10% below down to 360 K. At the altitude of 360 K, near the tropical cold point tropopause, the lowest fractions,  $f$ , of non-volatile particle mixing ratios are roughly 30% for Northern Australia (with considerable scatter in the data) and 5% over Burkina Faso and Brazil. In the stratosphere above the maximum at 400 K the smallest values are 25% over Australia (interestingly increasing towards 40% higher above) and 5% over West Africa. In this altitude the values over Brazil are near 25% and the minimum of 10% non-volatile fraction is found at 370 K. The highest values of  $f$  are for the profiles over West Africa and Northern Australia between 40% and 60 % with the maxima appearing in a layer between 380 K and 420 K.

For the case of meteoric dust subsiding inside the polar vortex values for  $f$  of 70% to 80% were reported within the same potential temperature band by Curtius et al. (2005). Consequently the dynamical range of values so far observed by COPAS on the M-55 “Geophysica” for the non-volatile fraction,  $f$ , is between 5% and 80%. The question arises, of course, whether the structure seen in the profiles is an atmospheric feature or possibly an instrumental artifact. On one hand the efficiency of the pre-heating device of the heated COPAS  $N_{10nv}$  channel has been experimentally characterised in the laboratory by Weigel et al, (2009), as function of ambient pressures between 70 hPa and 300 hPa. On the other hand the values for  $f$  from the DLR Falcon-20 very well match those from the M-55 “Geophysica” at 350 K (see lower panel of Fig. 10) for Australia and the combined profiles of the two aircraft show a distinctive narrow minimum in  $f$  at 360 K. The corresponding profiles over Brazil do not match similarly well. However, the scatter in the data between 350 K and 390 K is so large that the DLR Falcon-20 data are within its range. For these reasons we believe this apparent decrease of non volatility from 400 K to 360 K in the SCOUT-AMMA data from Fig. 10 most likely is not due to an instrumental artifact.

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The three profiles significantly differ from each other: Over West Africa there is the broadest maximum for  $f$  extending from 370 K to 440 K while over Brazil the ratio of non volatility roughly remains constant at low values (of 20% to 25%) above 330 K. Above altitudes of roughly 400 K the non-volatile fraction,  $f$ , decreases in all profiles, except for Brazil. This may indicate that the higher one gets into the stratosphere the more the volatile sulfuric acid-water solution droplets dominate the aerosol. A significant increase, however, is observed in the profiles over Northern Australia above 460 K. The extremely low levels of  $f$  found at high altitudes over West Africa are most likely a consequence of the volcanic eruption where the oxidation of the gaseous  $\text{SO}_2$  to  $\text{H}_2\text{SO}_4$  primarily produces binary  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  solution droplets. A common feature of the profiles from Burkina Faso and Australia is that the level of non-volatility,  $f$ , becomes smaller with decreasing altitude between 400 K and 360 K. The distinct minimum at 350 K found over West Africa does not occur over Brazil.

It is difficult to find conclusive explanations for the observed vertical profiles of  $f$  without additional measurements. Concerning the difference of the profile from Brazil with respect to the others it may be important that the measurements over Brazil were performed at latitudes near  $22^\circ\text{S}$  which is at the Southern edge and not well inside the tropical belt. Differences between the dry and wet seasons, the position of the ITCZ, and the level of biomass burning activity certainly plays a role for the volatility of the particles present in the sampled air masses. For the profiles over Australia and West Africa possibly below the tropopause the proportion of particles containing significant amounts of water on non-volatile cores is larger than higher above. It should be kept in mind that despite the lower non-volatile *fraction* the *absolute* number of non-volatile particles below the tropopause still is similar or larger than in the lower stratosphere because of the higher *total* ambient particle number density. This can be seen for example from the abscissa in Fig. 5. The increase in  $f$  over Northern Australia for altitudes above 460 K remains unexplained at this point. Obviously, without direct measurements of the chemical composition of these particles (e.g., like in Murphy et al., 1998; Froyd et al., 2009a, b; or Schneider et al., 2005) no simple explanation

for the observed features in the profiles can be given, which is in particular the case for the distinct “question mark shaped” profile over West Africa. If – as concluded by Brock et al. (1995) – the tropical upper troposphere is the source reservoir of aerosol particles for the global stratosphere, then our data suggest that those particles arriving in the tropical stratosphere contain significant amounts of non-volatile components. Following Froyd et al. (2009a) this can be due to the fact that convection rapidly carries air from the boundary layer aloft and that often biomass burning residues are reaching the tropical UT/LS.

#### 4 Summary and conclusions

In this contribution we present in-situ measured data of submicron particle properties from the 9 to 12 km altitude range, the UT/LS, and the TTL over Southern Brazil, West Africa, and Northern Australia. The tropical data are from local flights of the Russian high altitude research aircraft M-55 “Geophysica” and the German DLR Falcon-20 aircraft during the TROCCINOX campaign in Brazil, the SCOUT-O3 campaign in Northern Australia, and the SCOUT-AMMA campaign in West Africa (Burkina Faso; also Niger for the balloon flight). In addition and for comparison, tropical measurements of the NASA WB-57F high altitude research aircraft and the NASA BC-8 are shown from previous campaigns in Hawaii and Costa Rica dating back to 1999.

The transfer flights from Oberpfaffenhofen, Germany, and Verona, Italy, to the various tropical campaign locations offered the opportunity to document the particle number densities in the 9 to 12 km altitude band (from DLR Falcon-20) and above (from M-55 “Geophysica”) in the relatively short time frame between January 2005 and August 2006 during which no major volcanic eruption occurred. Only the local measurements over West Africa in July/August 2006 above altitudes of 430 K in potential temperature turned out to be influenced by the small eruption of Soufrière Hills on Montserrat two months earlier. At potential temperature levels above 367 K the ambient particle

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number densities range from  $20 \text{ cm}^{-3}$  at higher latitudes to values around  $100 \text{ cm}^{-3}$  on these transfer routes and the geographical distribution of the submicron particle number densities was rather homogeneous along the different latitude bands. This means when flying to the different continents no striking differences occur between the various routes of travel.

Concerning the vertical profiles of the particle mixing ratio over West Africa, Brazil and Northern Australia between  $55^\circ \text{ N}$  to  $20^\circ \text{ S}$  there is a remarkable similarity within the potential temperature band from 340 K to 430 K. Also these profiles very closely resemble the (significantly) earlier measurements from 1987 to 1994 (Brock et al., 1995) from Hawaii. Thus the particle abundance remained constant in the TTL altitude band for almost 20 years, as far as one can judge from all of these data sets which are sparse and irregularly spaced in time. The interpretation brought forward by Brock et al. (1995) of this layer being a continuous source of particles for the maintenance of the stratospheric Junge aerosol layer receives this way additional justification. Above 430 K the profiles from 2005 and 2006 differ from the 1987 to 1994 data by showing higher values at all levels up to roughly 500 K. Only for the vertical profiles over West Africa from 2006 we know that the volcanic eruption of Soufrière Hills on Montserrat is the likely cause of that increase. This is corroborated by the volatility measurements and the non-depolarizing nature of the sampled particle populations. For the measurements over Brazil and Northern Australia a reason for the increase with altitude cannot be given, and in particular it is not possible to identify some kind of an underlying trend. However, at all altitudes the data are reasonably compact and a parameterisation for use in models could be extracted.

Similar analyses of the vertical profiles at mid latitudes from the instrument test campaigns and the transfer flights to the tropical campaign sites show that the particle number densities as function of altitude remained fairly constant with time between 1996 and 2008 above 15 km. Thus, concerning submicron particle abundance, the background conditions of the Northern hemispheric mid latitude stratosphere are characterised by a relative stability since 1996 until 2008. Or at least one can conclude that

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no major trends of increases or decreases can be discerned from the available small data base.

The measurements of the non-volatile number fraction of the submicron particles performed by COPAS and the thermodenuder system on the DLR Falcon-20 provided useful – albeit limited – additional information on the particle properties in the tropical UT/LS region. For the profiles over West Africa the resulting data show that the proportion of volatile particles increases with altitude. This is in agreement with the notion that at higher altitudes binary sulphuric acid/water solution droplets are the major aerosol constituents resulting from the volcanic eruption. When comparing the fractions of non-volatile particles encountered over West Africa and Northern Australia a broad maximum (with 55% non-volatile particle number density fraction) is found centred at 400 K potential temperature altitude equivalent, i.e., directly at and above the tropical tropopause. Below and above the proportional number of non-volatile particles decreases to values near 25% with altitude up to 460 K in Australia, while the lowest values are 5% at 500 K over West Africa. The profiles from Brazil largely differ from this as there the non-volatile fraction is roughly 20% to 25% and constant with altitude above 330 K. Probably the measurements in Brazil took place so far South that the data are not entirely representative for the tropical UT/LS. If the tropical upper troposphere provides a reservoir of particles for the stratospheric Junge aerosol layer, then these “source”-particles are not only consisting of sulphuric acid and water but are enriched in non-volatile components. A variety of processes may be responsible for the observed vertical profiles of  $f$  including deep convection of boundary layer air and biomass burning residues being uplifted into the tropical UT/LS. Obviously, improved instrumentation and more field campaigns specifically dedicated to the properties of the tropical upper tropospheric and lower stratospheric aerosols and clouds are much needed in order to clarify some of these issues in addition to better geographical coverage especially over Africa and South East Asia.

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**Table 1.** Locations and times of the tropical campaigns TROCCINOX, SCOUT-O3, and SCOUT-AMMA with the Russian M-55 “Geophysica” and the German DLR Falcon-20 research aircraft

	TROCCINOX	SCOUT-O3	SCOUT-AMMA
Time period	January–March 2005	November–Dec. 2005	July–August 2006
Location of local mission flights	Araçatuba Brazil	Darwin Australia	Ouagadougou Burkina Faso
at latitude and longitude	21°8′37″ S. lat. 50°25′34″ W. long.	12°28′0″ S. lat., 130°50′1″ E. long.	12°22′13″ N. lat. 1°1′29″ W. long.
Transfer flights originating from	Oberpfaffenhofen Germany	Oberpfaffenhofen Germany	Verona Italy
at latitude and longitude	48°5′0″ N. lat. 11°17′0″ E. long.	48°5′0″ N. lat. 11°17′0″ E. long.	45°23′45″ N. lat. 10°53′19″ E. long.
Transfer flight time period to campaign site	20 January 2005 until 27 January 2005	4 November 2005 until 12 November 2005	31 July 2006 until 1 August 2006
Return transfer flight period	24 February 2005 until 2 March 2005	9 December 2005 until 17 December 2005	16 August 2006 until 17 August 2006
Campaign website	<a href="http://www.pa.op.dlr.de/troccinox/">http://www.pa.op.dlr.de/troccinox/</a>	<a href="http://www.ozone-sec.ch.cam.ac.uk/scout_o3/field_campaigns/index.html">http://www.ozone-sec.ch.cam.ac.uk/scout_o3/field_campaigns/index.html</a>	<a href="http://www.isac.cnr.it/~utls/m55amma/">http://www.isac.cnr.it/~utls/m55amma/</a>
Reference for campaign overview	Schumann, U., 2005	Vaughan et al., 2008, Brunner et al., 2009	Cairo et al., 2009

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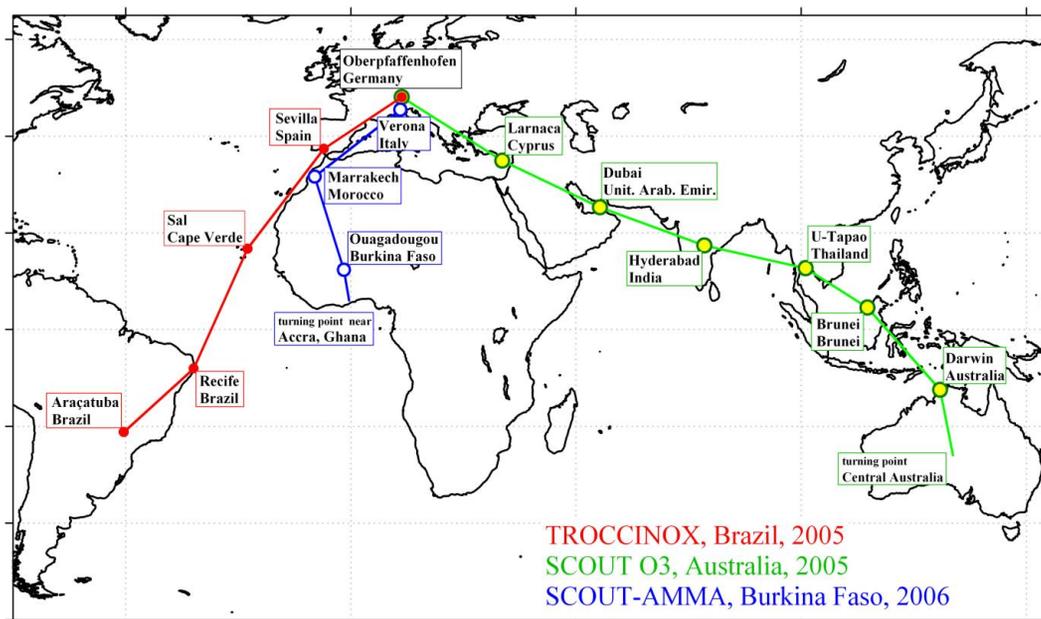
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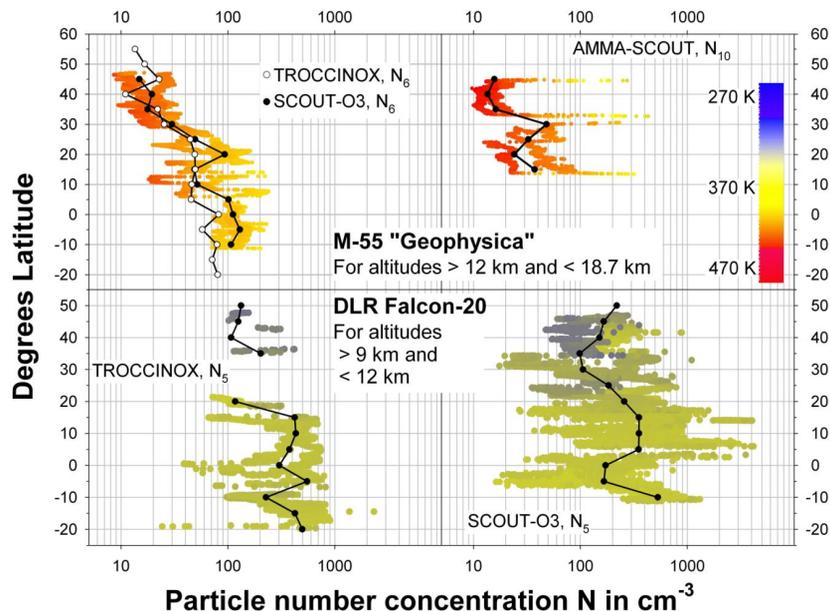


**Fig. 1.** Routes of the transfer flights for the M-55 “Geophysica” with intermediate landings from January 2005 until August 2006 for the TROCCINOX (Araçatuba, Brazil), SCOUT-O3 (Darwin, Australia), and SCOUT-AMMA (Ouagadougou, Burkina Faso) campaigns.

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**Fig. 2.** Number densities of submicron particles  $N_5$ ,  $N_6$ ,  $N_{10}$  (i.e., with size diameters larger than 5 nm/6 nm/10 nm and smaller than  $\approx 1 \mu\text{m}$ ) as function of geographical latitude and altitude in potential temperature from the M-55 "Geophysica" measurements (upper panels) and  $N_5$  concentrations from the DLR Falcon-20 (lower panels) of the transfers to the indicated campaign sites. Upper left panel: Open symbols are medians of 5 degree latitude bins and include the transfer from Europe to Brazil from 20 to 27 January 2005, as well as from the return flights between 24 February and 2 March 2005. The coloured data points represent 15 s flight time averages of these transfer flights with the colour coding referring to the potential temperature. The black line with filled symbols denotes the 5° medians from the transfer flights to Australia (4 until 12 November 2005) and back (9 until 17 December 2005). The individual 15 s data points are not shown for clarity. The filled dots and open circles represent five degree latitude bins in all panels. Upper right panel: Transfer flight data of  $N_6$  from Europe to Burkina Faso (31 July and 1 August 2006) and return (16 and 17 August 2006) in medians of 5 degree latitude bins and 15 s averages. Lower left panel: Latitudinal profiles of  $N_5$  between Europe to Brazil in the 9 to 12 km altitude band (15 s flight time averages). Lower right panel: Particle concentrations  $N_5$  for the transfer from Europe to Australia.

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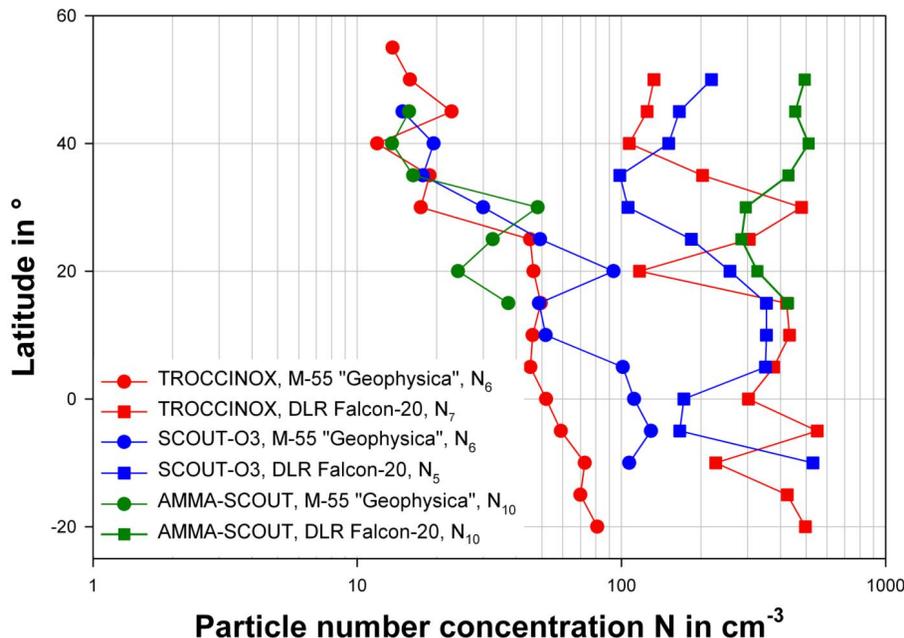
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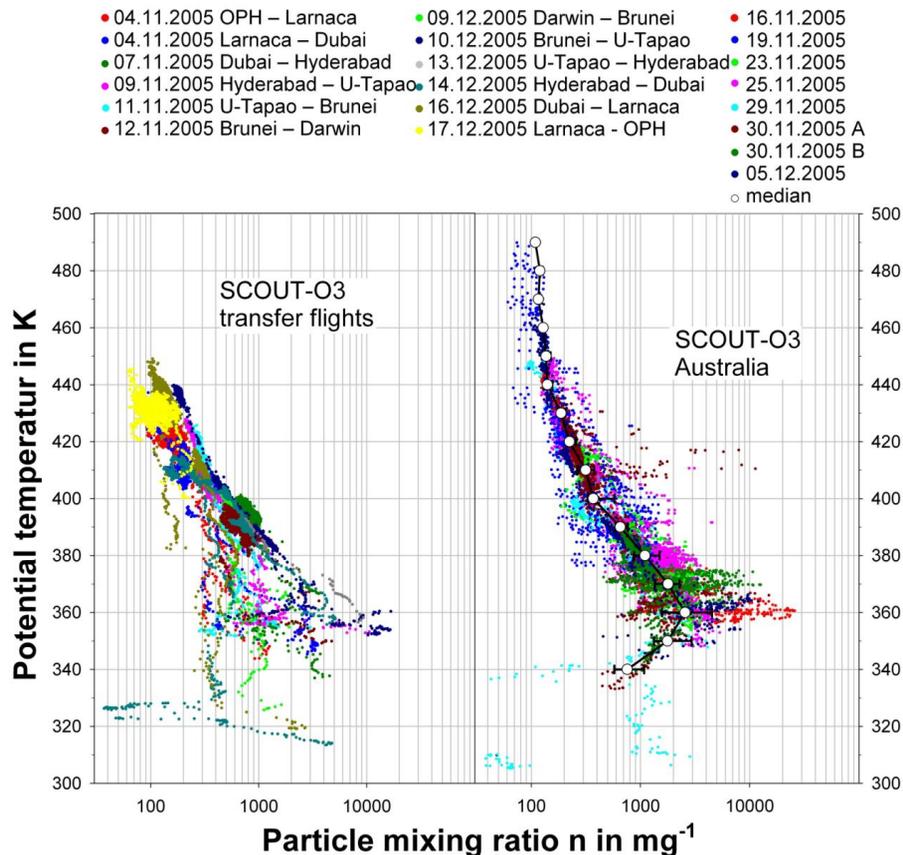


**Fig. 3.** Summary of the submicron particle concentration latitude profile data of Fig. 2 from the transfer flights of the DLR Falcon-20 and the M-55 “Geophysica” between Europe and Brazil, Australia, West Africa in 2005 and 2006. The DLR Falcon-20 data for the transfers to West Africa are not shown in Fig. 2 for clarity.

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**Fig. 4.** Left panel: Vertical profiles of  $n_{10}$  particle mixing ratios as measured by COPAS on the M-55 “Geophysica” during the intermediate descents and ascents from the transfer flights between Oberpfaffenhofen (Germany) and Darwin (Australia). The individual points represent 15 s flight time averages and the dates and locations of the profiles are colour coded as indicated in the legend. Right panel: Vertical profiles of  $n_{10}$  from the M-55 “Geophysica” local flights out of Darwin. Here the colour code refers to the flight dates, the points representing 15 s averages. The open symbols are 10 Kelvin potential temperature bins with the bars denoting the 25% and 75% percentiles.

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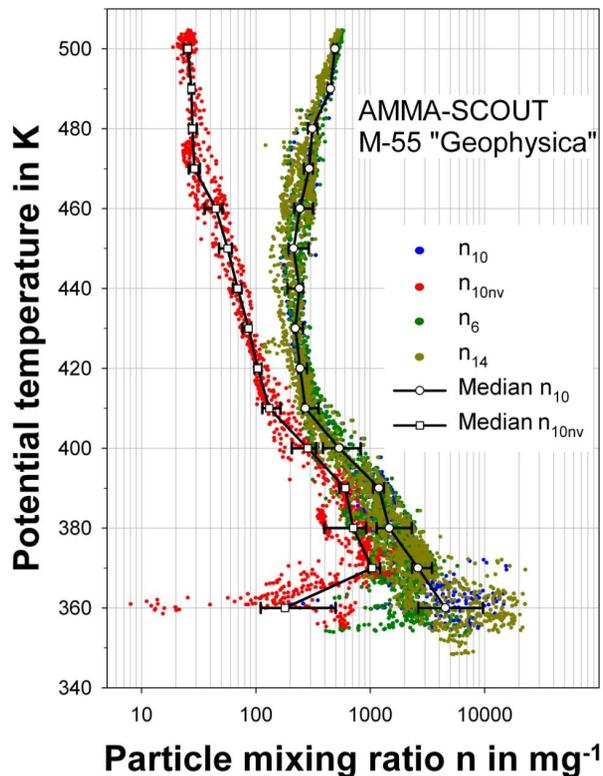
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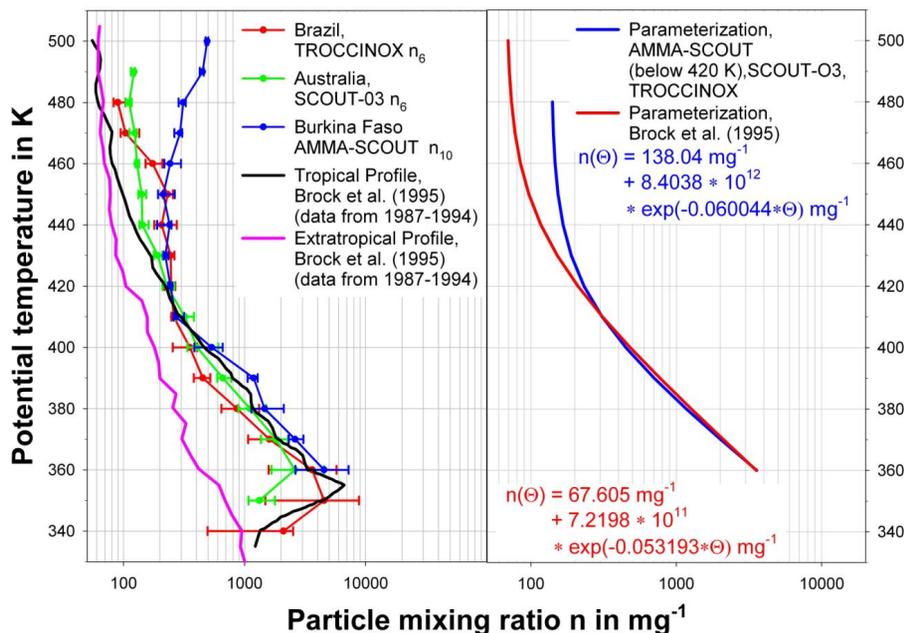


**Fig. 5.** Vertical profiles of the  $n_6$ ,  $n_{10}$ ,  $n_{14}$  and  $n_{10nv}$  ambient particle number mixing ratios from all of the M-55 “Geophysica” local flights out of Ouagadougou (Burkina Faso) in August 2006. The open symbols are medians of 10 K bins with the 25% and 75% percentiles. The left, red curve shows  $n_{10nv}$  with the 10 K binned medians of the heated channel representing the number densities of non-volatile particles.

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**Fig. 6.** Left Panel: Summary of the COPAS local particle mixing ratio vertical profiles ( $n$  vs. potential temperature  $\Theta$ ) as measured from the M-55 “Geophysica” in Araçatuba (Brazil), Darwin (Australia), and Ouagadougou (Burkina Faso). The solid symbols represent 10 K potential temperature bins with 25% and 75% percentiles. The black (purple) line shows the results from Brock et al. (1995) representing tropical (mid latitude) measurements from various campaigns with the NASA ER-2 between 1987 until 1994. Right Panel: Parameterisations derived from the  $n_{10}$  particle mixing ratio data of the tropical vertical profiles.

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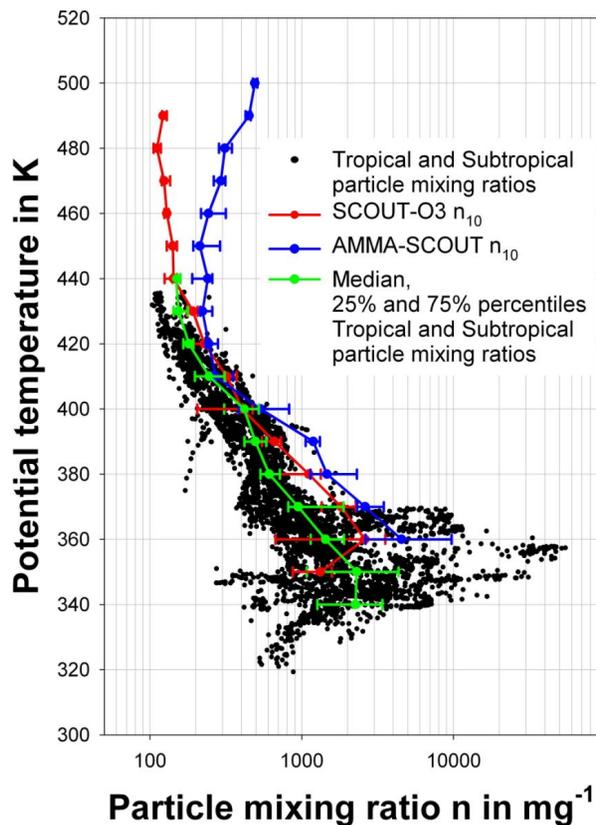
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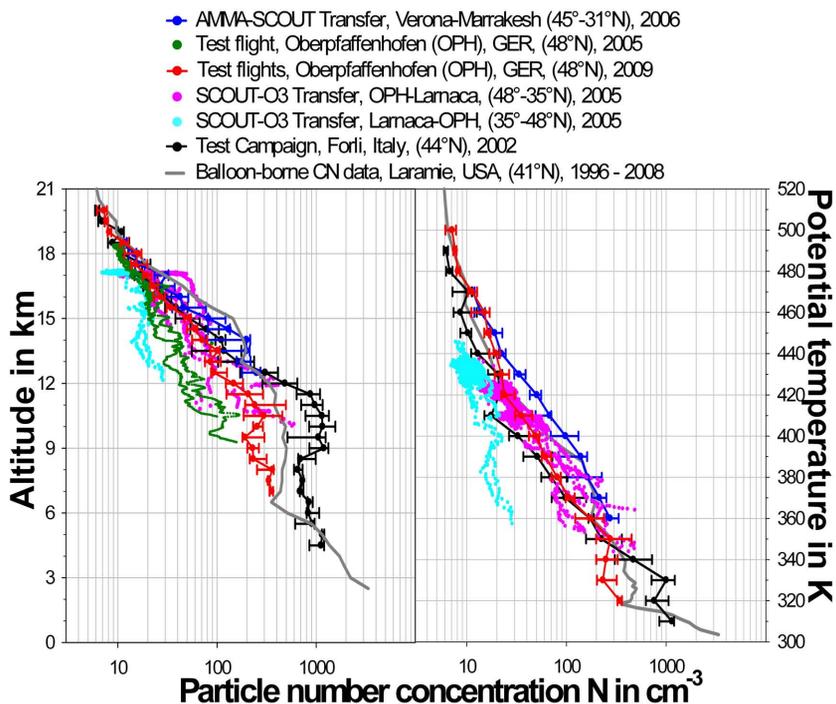


**Fig. 7.** Overview over available tropical and subtropical in-situ aircraft measurements of submicron particle mixing ratios. The black points are from NASA DC-8 and NASA WB-57F data covering the period from 1999 until 2007 (Source: University of Denver group) from the Caribbean, the Gulf of Mexico, and Hawaii obtained between 1996 and 2006. The coloured lines show the M-55 “Geophysica” data from Northern Australia (red), Burkina Faso (blue) and Brazil (green).

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**Fig. 8.** Mid latitude vertical profile data of  $N_{10}$  particle number densities for COPAS measurements from 2002 to 2009. (OPH denotes Oberpfaffenhofen, Germany.) A profile with an average of the balloon borne measurements from the University of Wyoming between 1996 until 2008 is shown for comparison (grey line). The blue, red, and green symbols are medians of 10 K altitude bins with the 25% and 75% percentiles.

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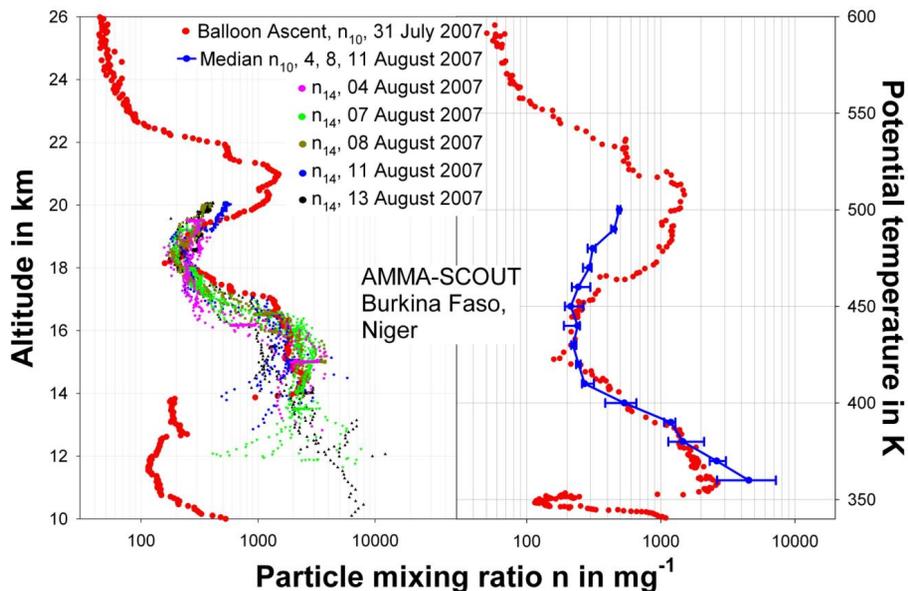
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**Fig. 9.** Intercomparison of the individual  $n_{10}$  particle mixing ratio measurements of the balloon launch on 31 July 2006 from Niamey, Niger, with the  $n_{14}$  COPAS data of all local flights from Ouagadougou, Burkina Faso. The COPAS data points in the left panel represent 15 s averages while the balloon data (red points) are averaged over 5 s. In the right panel the same data are displayed in dependency of potential temperature with the blue points designating the mean over 10 K altitude bins while the bars denote the 25% and 75% percentiles.

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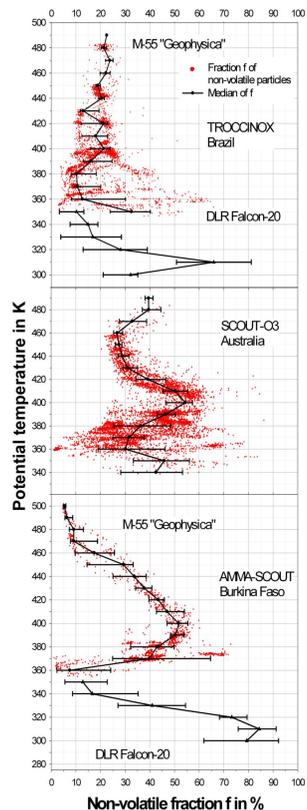
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Interactive Discussion



## Aerosols in the tropical and subtropical UT/LS

S. Borrmann et al.



**Fig. 10.** Ratio  $f$  of non-volatile particle number concentration  $N_{10NV}$  to total ambient particle number density  $N_{10}$  as function of altitude. The data points are 15 s flight time averages. Also the medians of 10 K altitude bins are indicated together with the 25% and 75% percentiles. The upper panel are data of the local flights from Araçatuba (Brazil) as well as from tropical and subtropical transfer flights between Sal (Capo Verde) and Recife (Brazil, 2005) while the middle panel shows the measurements from all local flights in Darwin (Australia, 2005). The lower panel contains the 2006 COPAS data from West Africa (red points and upper line with symbols and bars for potential temperatures above 360 K). Similarly DLR Falcon-20 thermodenuder measurements of  $N_7$  are included in the upper and lower panels for potential temperature levels below 350 K.

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