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Aerosol particle properties in the tropical free troposphere observed at Pico Espejo (4765 m a.s.l.), Venezuela

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Abstract

The first long-term measurements of aerosol number and size distributions in South-American tropical free troposphere were performed from March 2007 until Mai 2009. The measurements took place at the high altitude Atmospheric Research Station Alexander von Humboldt. The station is located on top of the Sierra Nevada mountain ridge at 4765 m a.s.l. nearby the city of Mérida, Venezuela. Aerosol size distribution and number concentration data was obtained with a custom-built Differential Mobility Particle Sizer (DMPS system) and a Condensational Particle Counter (CPC). The analysis of the annual and diurnal variability of the tropical free troposphere (FT) aerosol focused mainly on possible links to the atmospheric general circulation in the tropics. Considerable annual and diurnal cycles of the particle number concentration were observed. Highest total particle number concentrations were measured during the dry season (519±613 cm^{-3}), lowest during the wet season (318±194 cm^{-3}). The more humid FT contained generally higher aerosol particle number concentrations (573±768 cm^{-3} during dry season, 320±195 cm^{-3} during wet season) than the dry FT (454±332 cm^{-3} during dry season, 275±172 cm^{-3} during wet season), indicating the importance of convection for aerosol distributions in the tropical FT. The diurnal cycle in the variability of the particle number concentration was mainly driven by local orography.

1 Introduction

Aerosol particles are omnipresent in our environment. They exert significant effects on the Earth's climate. First, they absorb and scatter incoming solar radiation (direct radiative forcing; e.g. Haywood and Boucher, 2000). Second, they act as cloud condensation nuclei (CCN), influencing the cloud microphysical and optical properties as well as their life-time and precipitation (Seinfeld and Pandis, 1998; Twomey, 1974). Due to the high variability of the natural as well as the anthropogenic aerosol in the

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troposphere, the corresponding radiative effects vary highly in time and space. The latest report from IPCC has shown that atmospheric aerosols represent the largest uncertainty in understanding and giving future predictions of the Earth's climate (IPCC Fourth Assessment Report (AR4), 2007).

Most atmospheric aerosol properties – mainly number and mass concentration but, for instance, also mean diameter, chemical composition and morphology – vary greatly in both, time and space. Meanwhile, the existing in-situ aerosol observation system is patchy with respect to global coverage. The WMO's GAW (Global Atmosphere Watch) network currently encompasses 26 remote observation sites on all continents (Dlugokencky et al., 2010). At most of these remote sites, aerosols are characterized continuously, although particle number and size distributions only at few sites (e.g., Kivekäs et al., 2009). During recent years, observation networks for specialized in-situ aerosol observations have been established, for example the Nordic Network in Scandinavia (Dal Maso et al., 2007), the European Supersites for Atmospheric Aerosol Research (EUSAAR; http://eusaar.net), and the German Ultrafine Aerosol Network (Birmili et al., 2009a). Evidently, the surface-based in-situ measurements have concentrated on the more populated areas of the globe (including continental background and urban sites), while measurements in remote marine environment and in the free troposphere are especially scarce.

In the tropics, the lack of observational data is remarkable. During the last three decades, several field experiments took place in this region. These experiments often combined intensive ground and airborne measurements, but long-term aerosol observations are virtually missing. The NASA Global Tropospheric Experiment (GTE) (Hoell Jr., 2002) included a number of airborne field campaigns performed in tropical regions: The Atmospheric Boundary Layer Experiments (ABLE – 1, – 2A and – 2B, and – 3A and – 3B) are field studies dedicated to study ecosystems which are known to exert major influences on global chemistry and, in some cases, are undergoing profound changes. In June 1984, the ABLE – 1 mission studied the chemistry and transport processes over the tropical Atlantic Ocean and the rain forest of French Guyana. The ABLE –

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2 mission focused on the chemistry and transport over the Amazon rain forest during the dry season of 1985 (ABLE – 2A) (Harris et al., 1988) and during the wet season of 1987 (ABLE – 2B) (Harris et al., 1990). The TRACE A Experiment, which is also part of GTE, was performed during September and October 1992 (Pereira et al., 1996). It focused on determining the relative importance of natural versus anthropogenic emission processes on the formation of seasonal enhancements in tropospheric ozone over a large region of the South Atlantic Ocean between the coasts of Brazil and southern Africa. The Large Scale Biosphere-Atmosphere Experiment in Amazonia (LBA) international research initiative resulted in several field experiments in Amazonia and first long-term measurements of aerosol chemical composition (Artaxo et al., 1998) and observations of aerosol microphysical properties on a seasonal time scale (Artaxo et al., 2002; Guyon et al., 2003). Particle number size distributions and hygroscopic growth factors were also studied in the Amazonian rain forest boundary layer on a seasonal basis (Zhou et al., 2002; Rissler et al., 2006). In 1998, the LBA-CLAIRE-98 experiment examined the chemical composition and photochemical processes of the tropical boundary layer (Quesada et al., 2001). During the same experiment, airborne aerosol measurements provided information about distribution and aerosol properties in the tropical troposphere over the Amazonian rain forest (Krejci et al., 2003; Krejci et al., 2005). Besides The Amazon region, a summary of airborne observations over the Pacific Ocean as well as result from the INDOEX experiment (de Reus et al., 2001) shows a persistent "horse-shoe" aerosol number density profile in the tropical troposphere with minima and lowest variability in the middle free troposphere (Clarke and Kapustin, 2002). In summary, the body of existing aerosol measurements in the tropical free troposphere has been confined to time-limited field experiments. To our knowledge, long-term observations of aerosols from the tropical free troposphere have been entirely missing.

Long-term in-situ measurements of aerosol properties in the FT can only be performed at high altitude stations. Only measurements at Mauna Loa Observatory (Weber and McMurry, 1996), at Izana (Diaz et al., 2006; Rodriguez et al., 2009) and from

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ABC-PYRAMID station in Himalaya (Bonasoni et al., 2008) deliver long-term information from outer tropics and subtropics. Outside of the tropical regions, there are several well established observatories with long-term aerosol measurements. In the European Alps, long-term aerosol observations have been made at several mountain peaks: Jungfraujoch (3580 m a.s.l.) (Nyeki et al., 1998; Cozic et al., 2008), and Zugspitze (2650 m a.s.l.) (Birmili et al., 2009b). Kivekäs et al. (2009) examined particle number size distributions at Mount Waliguan (3816 m a.s.l.) on the edge of the Tibetan Plateau in inland China. Results from high-mountain observations show a clear seasonality in particle number and mass concentrations, with lower concentrations in the cold season, and higher concentrations in the warm season. Atmospheric convection has been made responsible for this annual cycle.

The prominent feature of the atmospheric circulation in the tropics is the Headley cell. It is governed by the seasonally migrating Inter-Tropical Convergence Zone (ITCZ), which forms the upward moving branch of the Hadley circulation. The ITCZ is approximately an east-west zone along which the northeast and southeast trade winds converge (Garstang and Fitzjarrald, 1999). Intensive convection associated with the ITCZ is an important transport pathway for aerosols and gases from the boundary layer to the upper troposphere and a source of new aerosol particles on a global scale (Heintzenberg et al., 2003; Hermann et al., 2003; Krejci et al., 2003; Minikin et al., 2003). The lifted air then moves northward or southward experiencing radiative cooling, which leads to a sinking motion in the poleward limps of the Hadley cell at about 30° N and S (Grotjahn, 2002). As the air subsides down into the boundary layer again it is transported back towards the ITCZ by the northeast and southeast trade winds. In addition to the Hadley cell circulation there is a second, so called low-level circulation in actively convective regions of the tropics. This closed regional circulation is caused by shallow convection (Folkins et al., 2007). Shallow convective clouds form an intermediate cloud type (besides boundary layer convection and deep convection) with cloud-top altitudes of 5–6 km and are thus limited by the height of the TWI. The TWI height is decreasing with distance from the ITCZ.

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Thanks to the stable general circulation pattern and vertical transport driven exclusively by convection and subsidence, the tropical troposphere can be divided into several distinct and semi-persistent layers of different characteristics (Garstang and Fitzjarrald, 1999). The lowest layer is called the mixed layer and is followed by the cloud convective layer (CCL). In the CCL the shallow convective fair-weather cumuli clouds are present, allowing transport of air from the mixed layer upwards to the top of the Trade Wind Inversion (TWI). Between the TWI and the tropopause the free troposphere (FT) is situated. Shallow convective clouds usually do not have enough buoyancy to penetrate trough the TWI into the FT. Therefore, the FT is characterized by being almost cloud-free, especially in the subsiding branch of the Hadley cell. Moreover, the only small influence of friction and the low intensity of sinks due to wet and dry deposition in the FT (Garstang and Fitzjarrald, 1999), it offers a good pathway for long-distance transport of aerosols.

As mentioned above, to our knowledge there is no study available on the long-term characteristics of free tropospheric aerosols in the humid tropics. This work aims to partially close this knowledge gap, by examining particle number density and size distributions recorded at a tropical high altitude research station for a period of two years. The main focus of this study is on the seasonal variation of aerosol properties and links to the general circulation in the tropics.

2 Field experiment and meteorological situation

2.1 Observation site and instrumentation

Aerosol measurements were carried out at the Pico Espejo Atmospheric Research Station Alexander von Humboldt. The station at Pico Espejo is located at 8°31′ N and 71°3′ W at an altitude of 4.765 m a.s.l. By the end of 2001, the measurement station was reconstructed for scientific use by the Universidad de los Andes (ULA) in collaboration with the University of Karlsruhe and the Research Centre Karlsruhe (FZK),

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Germany. The aerosol instrumentation was installed at the station by the Department of Applied Environmental Science (ITM), Stockholm University in March 2007. The station is located on top of the mountain ridge nearby the city of Mérida, Venezuela, a couple of kilometers away from Pico Bolivar (4998 m a.s.l.), the highest Venezuelan mountain. The city of Merida is located at an altitude of approximately 1630 m a.s.l. on a plateau in a small valley formed by the Chama and Albarregas rivers between the two mountain chains of Sierra Nevada and Sierra de la Culata. From the center of Mérida, a cable car (Teleférico de Mérida) leads in four sections directly to the Pico Espejo, overcoming an altitude of almost 3.200 m over a length of about 12.5 km. There are no heavy industrial, chemical or oil enterprises in Mérida and the economy of the region is mainly based on agriculture and tourism. The closest large industrial region is approximately 200 km north of the station near the lake Maracaibo, where the centre of the Venezuelan oil industry is located.

The measurements analyzed in this work cover period from 6 March 2007 until Mai 31, 2009. The aerosol was sampled through a custom-made 6 m long vertical inlet with controlled temperature. This leads to sampling at dry conditions with a relative humidity (RH) varying between 10 and 20%. The RH was measured by a Rotronic humidity sensor. The sample line was made of a 1/4″ stainless steel pipe. The temperature of the inlet heater was around 15 °C. The total sample flow varied between 3 and 4 l/min as a result from the variable sample flow of the particle soot absorption photometer, which was also installed at the site.

The aerosol instrumentation contains a Condensation Particle Counter (CPC, TSI model 3010), which measures the total number concentration of particles larger than 10 nm in diameter. To obtain more detailed information about the size distribution of the Aitken and accumulation mode aerosol, a custom-designed Differential Mobility Particle Sizer (DMPS) system is used. This system yields a size spectrum every 15 min. It classified particles into 25 bins between 20 nm and 470 nm from 5 March 2007 until 12 March 2009. Together with the total CPC this yields a size distribution from 10 to 470 nm. After 12 March 2009 the size range was extended to 710 nm in diameter.

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Meteorological parameters such as temperature, pressure, RH, wind speed and wind direction as well as direct and diffuse solar radiation are routinely measured using an automatic weather station. The time resolution of these parameters is one minute.

2.2 Meteorological conditions at Pico Espejo

From the routinely measured meteorological parameters, RH turned to be out the most useful for our subsequent aerosol data analysis (see Fig. 1). There is a high variability over the course of the year showing the annual cycle of wet and dry seasons. The dry season peaks during the period from January to March and the wet season from July to September. Periods between the wet and dry seasons are called wet-to-dry and dry-to-wet transition seasons. During Northern Hemisphere summer months, which represent the wet season, the location of the ITCZ causes organized mesoscale and synoptic-scale systems to produce a large amount of rainfall over a relatively wide area including the region of Venezuela and Pico Espejo. During the dry season, the ITCZ is located further to the south causing a decrease in convective activity and thus a decrease in precipitation. This decrease in the occurrence of deep convection also leads to a higher amount of subsiding air from aloft. These subsiding air masses experience adiabatic warming, which causes further reduction of relative humidity. Annual and seasonal mean values as well as maxima and minima for these parameters are given in Table 1.

Besides the seasonal cycle, there exists also a profound diurnal cycle at Pico Espejo. During night to midday, the station is often in the FT with the boundary layer well below the station. Increasing surface heating and convection during the day result in an intensification of orographic flow. Consequently, from around midday the station is influenced by orographic upslope flow and orographically driven cloudiness.

2.3 Data preparation and classification

The measured particle mobility distributions were inverted into particle number size distributions by a multiple charge inversion routine. Subsequently, the entire data set

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(6 March 2007–31 Mai 2009) was checked for possible errors, i.e. instrumental faults, the effects of service checks at the station, or errors resulting from the size distribution inversion. Erroneous data, which accounted for 12.9% of the entire data set, were excluded from further analysis.

The design of the aerosol sampling inlet required additional attention with respect to the data treatment. The aerosol inlet at Pico Espejo was initially designed with sampling free tropospheric aerosols in mind, i.e. under cloud free conditions. During the course of almost every day, however, and especially during the wet season, orographic flow along the mountain slope causes humid boundary layer air to rise upslope. This results in the presence of clouds at the station. The aerosol inlet at Pico Espejo has an undefined cut-off diameter during in-cloud measurements, i.e. it does not operate as a whole-air-inlet. As a result, an undetermined fraction of cloud droplets can be impacted at or near the entrance of the inlet and thus not reach the instrumentation. Thus, under cloudy conditions the aerosol examined consists of the interstitial aerosol plus an undefined number of cloud drop residuals.

To avoid any bias related to the presence of clouds, it was necessary to split the aerosol data into two groups: Data measured under cloudy conditions and data measured under cloud free conditions. The RH measured at the station was used as a proxy for this division. It was assumed that the station was in clouds when RH values exceeded 95%. Data measured with RH below 95% represents the cloud free observations. Data measured when the RH sensor was not working were excluded from the analysis.

Based on observations made by Ryoo et al. (2009), who analyzed probability density functions of tropical tropospheric humidity, the data from the FT was further divided into two groups: humid and dry FT data. Observations revealed that there are two main physical processes controlling the tropical tropospheric RH distribution leading to two different RH regimes of the FT. Convection and vertical mixing play the key role in regulating humidity near tropical convective regions. In such convective regions, the air at and above 200 hPa is composed mainly of very moist air parcels which have just

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detrained from convection, whereas around 400 hPa there is a combination of moist air from recent detrainment and very dry air that has subsided from 200 hPa. Thus, midlevel air (at about 300–500 hPa) is a mixture of recently detrained moist air, representing the humid FT and very dry air that has subsided from higher altitudes, which represents the dry FT (Ryoo et al., 2009). The division into these two groups was again done using RH. Data measured when RH was between 50% and 95% belong to the "humid FT group". In contrast, all data measured while RH was less than 50% belong to the "dry FT group". Table 2 shows the complete division of the data together with the respective relative humidity ranges and the numbers of size distribution scans for each group.

From the complete dataset of 60 405 size distribution scans, 40.2% of the data belong to the "cloud group" and had to be excluded from further analysis. Furthermore, 54.4% of the data belong to the "cloud free group" and for 5.4% of the data no RH data was available. Regarding the "cloud free data", 70.1% and 29.9% of these measurements belong to the humid FT and the dry FT, respectively.

To analyze the annual cycle of the aerosol data, monthly mean values were calculated. To make the data more accessible to other scientists, multiple lognormal modes were fitted applying a least-squares algorithm on the monthly means of the number size distributions. An interactive LabVIEW program (Version 6.1, National Instruments, Austin, USA) was used for fitting three lognormal particle modes representing the nucleation, Aitken and accumulation modes. For better comparison of these fits, fixed sigma-values (representing the width of the different modes, Table 4) were used.

3 Results

3.1 Annual cycle in total aerosol number concentration

As can be seen in Fig. 2, the total aerosol particle number concentration for the humid and the dry FT aerosol possess a strong variability over the course of the year. Higher

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particle concentrations occur during the dry seasons (January until March) and the lowest particle concentrations were measured during the wet seasons (July until September). In Table 3, the average of the total particle number concentration as well as the standard deviation are shown for the different seasons and FT types. Regarding the dry season, the mean number concentration of 573±768 cm^{-3} for the humid FT aerosol is slightly higher compared to the one of the dry FT aerosol with 454±332 cm^{-3}(daily mean values of total aerosol number concentration ± standard deviation). With transition to the wet season, particle concentrations for both FT types decrease in average. During the wet season concentrations of 320±195 cm^{-3} and 275±172 cm^{-3} are observed for the humid and dry FT, respectively. This persistent feature of generally lower particle concentrations for the dry FT compared to the humid FT can be seen in Fig. 2.

Figure 3 displays the particle number concentration in form of Box-and-Whisker plots. Again, the higher concentrations during the dry season compared to the wet season become obvious, as well as higher values for the humid FT compared to the dry FT. Furthermore, the variability in the aerosol concentration for the dry season 2007 is the largest over the whole period of time for both aerosol groups. Measurements started at the beginning of March 2007 during the later part of the dry season when biomass burning is usually more active than during onset of the dry season. This is very likely the main reason why data from the dry season 2007 shows a larger variability and a higher aerosol number concentration in general. Similar reasons cause unusually high values in the dry-to-wet transition season 2009. Moreover, during the dry seasons the total number concentration for the humid FT aerosol shows more variability compared to the one of the dry FT.

3.2 Annual cycle in particle number size distributions

Aerosol size distributions have been divided the same way as aerosol densities. Similarly they also show a distinct seasonal pattern. During the beginning of the dry season (January to February) the mean number size distribution for the humid FT aerosol

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reveals a well defined Aitken and accumulation mode (Fig. 4). The particle number concentration in the accumulation mode is higher than the one in the Aitken mode. The dry FT aerosol in contrast shows only a significant Aitken mode, which has a higher particle concentration (323 cm^{-3}) than the humid FT aerosol (261 cm^{-3}). Dry FT accumulation mode particle concentrations (87 cm^{-3}) are lower than for the humid FT aerosol (155 cm^{-3}). The same can be seen in the right panel of Fig. 5, which displays particle number concentrations for the different size modes and FT types derived from a least-squares mode fit program. The left panel of Fig. 5 shows the monthly mean diameters for the size classes and FT types derived from the same program. At the beginning of the dry season and onward to the biomass burning season (March to April) the nucleation, Aitken and accumulation modes experience an increase in modal size, with exception of the dry FT nucleation mode. They all reach their maximum at the end of the biomass burning season. The only exception forms the accumulation mode of the dry FT aerosol, which shows further maximum diameter values during the wet and wet-to-dry transition season (Fig. 5a, data points in brackets). These high values are consequences of a lower data capture of dry FT aerosol data during these wet months resulting in a lower input for monthly averaging.

The mean particle number size distribution for the biomass burning season shows a clear maximum in the accumulation mode of the humid FT aerosol. Particle concentrations in this mode are significantly higher (487 cm^{-3}) than during the first two months of the year (155 cm^{-3}). The dry FT aerosol also experiences an increase in the particle concentration of the accumulation mode. This can be seen in the shift of the modal maximum particle concentrations from about 30 nm towards larger particle diameters of about 80 nm during the course of the dry season (Fig. 4, see also Fig. 5).

With regard to the wet season (July to September), the relation between the humid FT Aitken and accumulation mode particle number concentration changes. The particle number concentration in the Aitken mode increases up to 275 cm^{-3} whereas the concentration in the accumulation mode decreases to 51 cm^{-3}. A similar behavior can be seen in the particle number size distribution for the dry FT aerosol.

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Thus, the monthly mean particle number size distributions reveal a strong variability over the course of the year with significant differences for both, the humid and the dry FT aerosol. Remarkable is a relatively persistent Aitken mode throughout the whole year. The most striking differences occur in the accumulation mode when comparing the beginning of the dry season, the adjacent biomass burning season and the wet season. Monthly mean modal parameters for both FT types derived from a mode fitting program are depicted in Table 4. They again confirm the existence of a seasonal cycle with highest particle concentrations during the biomass burning season and lowest during the wet season, especially with regard to the accumulation mode. Moreover, the differences between the two FT aerosol types become obvious (especially the generally higher concentrations of Aitken and accumulation mode particles for the humid FT).

The data shows a remarkable relationship between the particle number concentration of the accumulation mode and the humidity of the ambient air. Fig. 6 shows the correlation between the specific humidity and the accumulation mode aerosol number concentration. The specific humidity is a measure, which describes the amount of water vapor in g contained in 1 kg of humid air. Its benefit compared to the RH is that it takes into account the role of the air temperature, which controls the saturation vapor pressure in ambient air. Obviously, the dry FT aerosol goes always together with lower specific humidity values compared to the wet FT aerosol. Furthermore, the concentration of accumulation mode particles is highest during the biomass burning season, second highest during the dry season and lowest during the wet season.

3.3 Diurnal cycle of the particle number size distribution

Besides the annual cycle of the particle number concentration and size distribution, the diurnal evolution also shows a profound repeating pattern of the measured aerosol properties. To analyze the diurnal cycle for the period of the dry and the wet season, mean particle number size distributions were calculated and displayed in the form of contour plots (see Fig. 7a–d). Since there is a strong correlation between the daily development of the humidity of the ambient air and the aerosol particle size and number

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concentration, a calculated mean specific humidity curve is depicted as well. It shows that the aerosol measured at the Pico Espejo station is strongly influenced by radiatively and orographically forced convection.

3.3.1 Diurnal cycle during the dry season

In the diurnal cycle for the humid FT aerosol during the dry season (Fig. 7a), the specific humidity has its minimum during the morning hours. At the same time, the aerosol number concentration is relatively low in all three modes. With increasing solar insulation the boundary layer air masses experience a lifting and rise upslope to the station. This convective activity results in an increase in the specific humidity around midday. At the same time, the particle number concentration of all three modes increases, whereas the mean mode diameters of the nucleation and Aitken modes experience a decrease. The strongest influence on the particle number concentration is observed in the accumulation mode (Fig. 7a, lower panel). However, the mean diameter of the accumulation mode shows a slightly different behavior. When the specific humidity increases during the afternoon and finally reaches its maximum values, the mean diameter of the accumulation mode experiences an increase as well. With beginning of the evening hours the strength of the solar insulation weakens and the convective activities die. Therefore, the specific humidity and the particle concentration, especially the one of the accumulation mode particles, experience a decrease. The mean modal diameters in contrast increase since the particles experience aging through condensation of gaseous compounds and coagulation of smaller particles.

The dry FT aerosol shows a similar diurnal cycle during the dry season (Fig. 7b). Again, the aerosol number concentration is lowest for all modes when the specific humidity is at its minimum. The maximum specific humidity values go again together with minima in the mean mode diameters of the nucleation and Aitken mode. The mean diameter of the accumulation mode, in contrast, remains rather constant und thus unaffected by the specific humidity maximum. Increasing specific humidity during the midday hours from about 1.3 g/kg to approximately 3 g/kg represents a convective

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lifting of boundary layer air masses upslope the mountain, which is weaker compared to the one observed for the humid FT. Consequently, particle number concentrations increase in all modes. The strongest influence is seen in the Aitken mode. In the nucleation mode, the increase in particle concentration is even higher than observed for the humid FT aerosol. The increase in the concentration of accumulation mode particles is lower compared to the humid FT aerosol.

3.3.2 Diurnal cycle during the wet season

The diurnal cycles of the humid and dry FT aerosol measured during the wet season reveal a remarkable lower particle concentration in all three modes, compared to the dry season. The typical diurnal evolution of the specific humidity can again be seen. This evolution is again strongly linked to the properties of the observed aerosol caused by a typical diurnal cycle in convection and atmospheric circulation along the mountain range.

In the diurnal pattern of the humid FT aerosol (Fig. 7c), a higher specific humidity correlates with a higher particle concentration in all three modes, as already observed for the dry season. Thereby, the concentration of the Aitken mode particles increases the most. Accumulation mode particles do not experience such a strong increase in concentration, since they are most affected by wet deposition and therefore most efficiently removed from the atmosphere. Still, a remarkable correlation between the accumulation mode number concentration and the specific humidity was found (see Fig. 7c, lower panel). Furthermore, the mean mode diameters change with altering specific humidity. The diameters of the nucleation and Aitken mode experience a reduction while the specific humidity reaches its maximum values. The mean diameter of the accumulation mode, in contrast, increases as already observed for the humid FT aerosol of the dry season.

The mean pattern for the diurnal cycle of the dry FT aerosol during the wet season (Fig. 7d) reveals slightly different particle properties. First of all, it is limited by the RH

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values. Thus, there are no aerosol data for the afternoon and evening hours since then RH always exceeds the critical value of 50%. Nevertheless, for the existing data the typical relation between the specific humidity and the particle number concentration becomes obvious. During the morning hours, Aitken mode particle concentrations are higher compared to the humid FT aerosol. Moreover, the particle number concentration in the Aitken mode is higher during times with higher specific humidity. The particle number concentration of the accumulation mode aerosol is again low due to the generally lower concentration of large particles (belonging to the upper Aitken mode size range and the accumulation mode size range) in the upper tropospheric layer and the influence of wet deposition. Around midday, the specific humidity increases and especially the particle number concentration of the nucleation mode shows a strong increase. In the Aitken and accumulation mode, an increase is seen, too. The mean diameters of the nucleation and accumulation mode show a strong variability, whereas the one for the Aitken mode remains rather constant. With increasing specific humidity the mean diameter of the nucleation mode enlarges. For the accumulation mode, a significant increase in the mean diameter is seen when the specific humidity decreases at about 10 UTC. But afterwards, when the specific humidity shows a constant increase, the mean diameter of the accumulation mode increases as well.

4 Discussion

The main reason for the observed variability in the tropical FT aerosol lies within the Headley cell and its connection to the seasonal migrating ITCZ. Migration of the ITCZ results in the formation of dry and wet seasons in the area of the tropics. Consequently a seasonal and different diurnal cycle can be observed in the FT aerosol properties observed at Pico Espejo.

The generally higher particle number concentrations and aerosol size distributions dominated by accumulation mode aerosol measured during the dry seasons (January until March) are due to an enhanced occurrence of biomass burning during the dry

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seasons, especially in the area of the Amazon Basin. Deforestation and pasture cleaning for economic reasons forms a steady source of aerosol particles during this time of the year (Martin et al., 2008). Moreover, burning activities in South America constitute almost 30% of the world's biomass burning activities (Rissler et al., 2006). If these particles become injected to the free troposphere by convective transport, they can be transported over long distances and affect large areas (Andreae et al., 2001). Furthermore, the ITCZ is placed south of the station during these months. Therefore areas of intensive deep convection are located further south of the station and large-scale subsidence has stronger influence in the region of the station compared to the wet season. Precipitation and aerosol wet removal are lower and thus the influence of wet deposition decreases.

During the wet season (Northern Hemisphere summer), the aerosol particle number concentrations are generally lower, compared to the dry season. During this time of the year, the ITCZ is located almost directly over the measurement station and deep convection is the prevailing weather feature. Consequently, precipitation rates are very high. This results in an effective removal of aerosols from the troposphere caused by in-cloud scavenging when the air masses are transported from the boundary layer to the FT. Moreover, sources of new aerosol particles due to biomass burning are weak during the wet season (Guyon et al., 2003). Convective clouds form the only effective particle source. Thus, the accumulation mode particle transport efficiency from the boundary layer to the free troposphere is rather low (compared to the dry season) due to wet removal.

The variability in the measured aerosol number size distributions is a consequence of the different sensitivity of the aerosol sizes to the particle sinks and sources. Regarding the accumulation mode, the variability is most pronounced. This is due to the fact that biomass burning produces mainly accumulation mode particles with diameters around 100 nm or greater (Guyon et al., 2003). Consequently, their number concentration is largest during the dry season. In-cloud and below-cloud scavenging are removal processes which affect mainly particles in the accumulation mode size. Thus, during

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the wet season, the accumulation mode experiences the strongest decrease due to wet deposition and weaker sources on the surface.

Aitken mode particles show a rather constant number concentration throughout the course of the year. In tropical regions convective clouds form an effective source of new particles (Avery et al., 2001; Hermann et al., 2003; Minikin et al., 2003; Krejci et al., 2003). Those very small particles experience a subsequent growth from nucleation to Aitken mode sizes due to condensation and coagulation, while transported slowly downwards from convective cloud outflow with large-scale subsidence. Thus, Aitken mode particles have a relatively continuous source since convective clouds occur throughout the year in tropics. A well developed Aitken mode is one of the most pronounced features of the tropical and sub-tropical free troposphere, often resulting in a monomodal, nearly self-preserving aerosol size distribution (de Reus et al., 2001; Krejci et al., 2003). Moreover, they are rather unaffected by wet deposition since they are too small to become activated in clouds or collected by falling rain drops. Nucleation mode particles show a similar behavior as Aitken mode particles when analyzing the seasonal variability of their number concentration.

Regarding the two types of FT, the humid FT possesses a generally higher aerosol particle number concentration than the dry FT. This is a result of the different natures of these two air mass types. Higher humidity indicates that the humid FT air masses were recently influenced by convective transport from the boundary layer to the FT. Moreover, as already mentioned, convection forms a source for new particles in the area of the tropics and higher RH is therefore correlated with a higher particle concentration. These factors are also responsible for a higher variability of the humid FT aerosol, compared to the dry FT aerosol (see Fig. 3). The dry FT air masses in opposite indicate the influence of more aged and well mixed air which was not recently influenced by convective transport. Thus the occurrence of new particle formation associated with convective clouds is lower. At the same time the amount of particles transported from the boundary layer to the FT is lower as well. Moreover, it is very likely that air masses belonging to the dry FT have spent a longer time at great altitudes in the atmosphere.

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When these air masses slowly subside from higher altitudes, they experience adiabatic compression and thus warm up. Therefore, RH decreases even more and reduces the chance for convection and new particle formation even further.

A closer look at the concentration of the different aerosol size modes reveals that it is especially the accumulation mode, which exhibits a strong difference in its particle number concentration depending on the FT regime. The number concentration of accumulation mode particles from the humid FT is higher throughout the year compared to the one for the dry FT. Reason is that these particles have their sources mainly on the surface. The convective transport, whose intensity varies with proximity of the ITCZ, carries them to the FT and consequently to the measurement station. Moreover, there are no known sources of accumulation mode particles in the FT apart from transport from the boundary layer and in-cloud processing of aerosols. Regarding Aitken and nucleation mode particles, the difference in the number concentration for humid and dry FT is rather insignificant.

Besides the seasonal cycle there exists also a diurnal cycle in the particle number size distributions of the FT aerosol. The observed variability is strongly linked to the prevailing RH (see Fig. 7a – 7d). This can again be explained by the different sensitivity of the aerosol size classes to the ambient conditions. Therefore, the characteristic features for the dry and wet season, as well as for the dry and humid FT show on the smaller time scale a characteristic diurnal pattern. Due to the orography warming air masses are forced to rise upslope the mountain side during the morning hours. This leads to an increase of the height of the boundary layer and to the development of convection. Around midday the station is thus surrounded by air masses with high RH and often even surrounded by clouds. Convective activity leads again to the formation of new particles causing an increase in the concentration of nucleation and Aitken mode particles. At the same time, the concentration of the accumulation mode aerosol increases as well, as these particles are transported with convection. Towards the evening hours, when solar insulation decreases, the convective activity diminishes and the RH decreases.

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5 Summary and conclusion

The goal of this investigation was to achieve a better understanding of the tropical FT aerosol. As a basis of this study, aerosol particle number size distribution measurements were performed at the Atmospheric Research Station Alexander von Humboldt, located on the mountain Pico Espejo (4.765 m a.s.l.), Venezuela. Our data set covers the period from 6 March 2007 until 31 May 2009, and thus provides the first long-term observations of FT aerosol in the tropics.

Ambient relative humidity was used as a criterion to classify the data set into different groups: cloudy conditions (RH > 95%) and cloud free conditions (RH ≤ 95%). The cloud free data was further divided into humid FT aerosol (95% ≤ RH ≤ 50%) and dry FT aerosol (RH < 50%). Particle number and size distributions in these groups were separately analyzed with regard to annual and diurnal cycles. For all data groups, a cyclic annual behavior was found, which could be linked to the variability of the general circulation.

The highest particle concentrations are observed during the dry season ($519 \pm 613\,cm^{-3}$) and lowest during the wet season ($318 \pm 194\,cm^{-3}$). This can be ascribed to the seasonal migration of the ITCZ, which forms the upwards moving branch of the Headley cell. During the dry season (January–March) the ITCZ is located southwards of the station causing a decrease in deep convection and precipitation. Thus, wet removal rates are low. At the same time, biomass burning activities are high leading to new particle formation, especially in the size range of the accumulation mode. In contrast, during the wet season (July–September), the ITCZ is located almost directly over the measurement station. Consequently, biomass burning is strongly suppressed and efficient wet removal results in a strong decrease of especially accumulation mode particles. Therefore, a cyclic behavior in the particle number size distributions becomes obvious with a relatively persistent Aitken mode but highly variable accumulation mode.

Looking at the humid and dry FT separately, the humid FT contains a higher particle concentration during all the seasons. It is very likely that the humid FT was recently

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influenced by convective transport of air masses from the boundary layer. Since most of the particle sources are located on or directly above the ground, boundary layer air contains more particles than FT air. Moreover, high RH and convection is a source of new particles in the area of the tropics (Avery et al., 2001).

With regard to a diurnal cycle of the Pico Espejo aerosol, the influence of the seasonal variability of the general circulation becomes obvious as well. During the dry season, the daily mean particle concentration is higher in all modes, compared to the wet season. Moreover, a significant correlation between the particle number concentration (especially of accumulation mode particles) and the humidity of the air masses around the station (specific humidity) can be observed. Higher specific humidies go together with a higher concentration of accumulation mode particles, since a high specific humidity indicates convective transport of boundary layer air masses to the measurement station. These higher particle concentrations are always observed from midday and in the afternoon hours, when solar insulation leads to a forced upward motion of boundary layer air and the orography amplifies the convective activity. With decreasing solar insulation towards the end of the day the convection decreases, resulting in lower particle concentrations.

The results obtained from these long-term measurements of tropical FT aerosol could be among others of interest to test the output (particle number concentration and size distribution, diffusion and distribution of emissions) of global aerosol models.

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Table 1. Average meteorological parameters derived from daily means at Pico Espejo from March 2007–March 2009 and for individual seasons (for solar radiation only daytime data used).

	RH [%]	temperature [°C]	solar radiation [W/m^2]	pressure [hPa]
mean 2007–2009	76.8	0.5	408.1	575.2
max 2007–2009	99.8	3.7	749.8	577.2
min 2007–2009	10.6	−3.6	45.3	569.1
mean dry season	61.9	0.7	478.3	574.8
max dry season	98.9	3.3	705.4	576.3
min dry season	10.6	−1.8	69.1	573.2
mean wet season	91.2	−0.3	365.7	575.5
max wet season	99.2	−3.6	657.7	577.2
min wet season	46.7	2.2	93.7	569.1

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Table 2. Classification of the particle number size distributions into five groups according to different levels of relative humidity (RH).

data group	cloud data	cloud free data	humid FT	dry FT	no RH data
RH range	RH>95%	RH≤95%	95%≤RH≤50%	RH<50%	RH out of scale
# size distributions	24 278	32 860	23 033	9827	3267

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Table 3. Average total particle number concentration ± 1 standard deviation in cm^{-3}

	dry	dry to wet	wet	wet to dry
humid FT	572.86±768.06	488.55±261.66	320.42±194.54	398.55±465.28
dry FT	453.63±332.10	389.59±222.47	275.10±171.75	447.86±489.55

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Table 4. Diameter and number concentration derived from LabView mode fit program for 2008.

	Accumulation mode (sigma = 1.600)				Aitken mode (sigm = 1.583)				Nucleation mode (sigma = 1.562)			
	Humid FT		Dry FT		Humid FT		Dry FT		Humid FT		Dry FT	
	Dp	*N*	*Dp*	*N*	*Dp*	*N*	*Dp*	*N*	*Dp*	*N*	*Dp*	*N*
1	135	290	127	68	39	256	58	172	18	54	24	181
2	139	165	139	72	44	139	51	179	16	25	23	139
3	162	436	154	135	92	185	66	244	32	140	28	193
4	186	332	168	119	91	147	72	235	35	126	25	66
5	172	151	196	65	50	195	69	184	17	40	24	53
6	130	116	133	39	41	248	54	241	18	89	20	83
7	143	90	208	9	46	212	61	200	19	106	22	84
8	131	59	122	24	39	185	49	142	15	37	22	95
9	126	76	137	10	39	213	53	144	13	23	17	33
10	145	69	207	18	49	155	58	216	19	39	20	37
11	133	100	135	26	42	257	48	286	16	39	24	158
12	128	133	132	30	41	256	53	195	17	99	22	160

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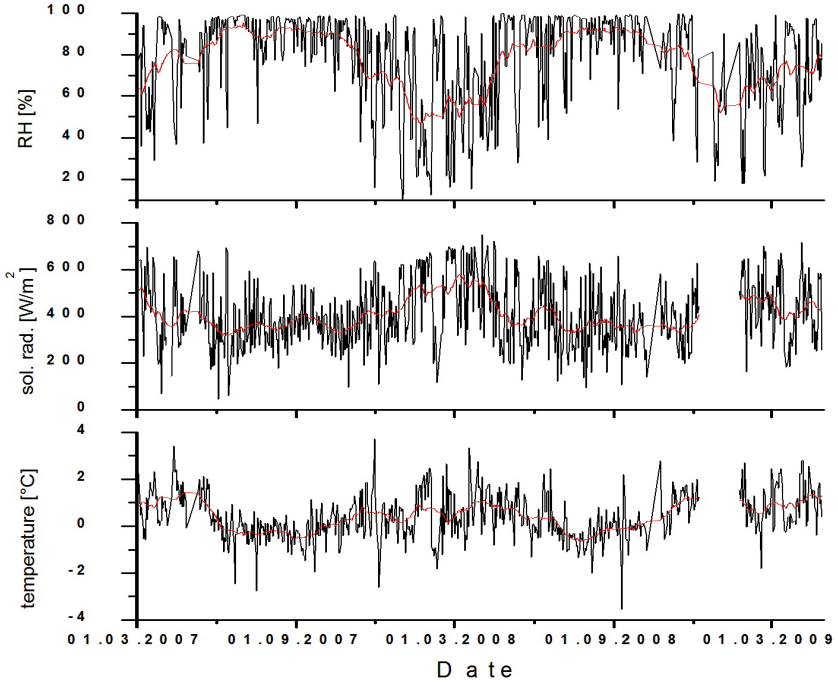

Fig. 1. Multi-annual time history of relative humidity, solar radiation and temperature at Pico Espejo (black – daily average, red – 30 days moving average; for solar radiation only daytime data)

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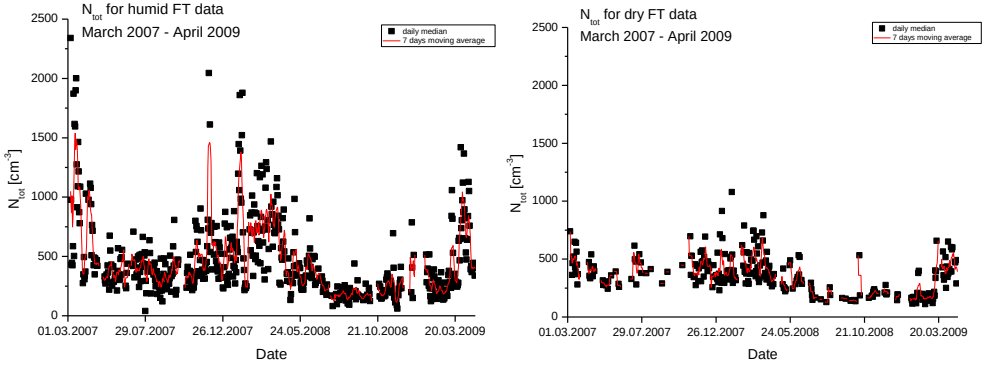

Fig. 2. Total particle number concentration for humid FT data (**a**, left panel) and dry FT data (**b**, right panel).

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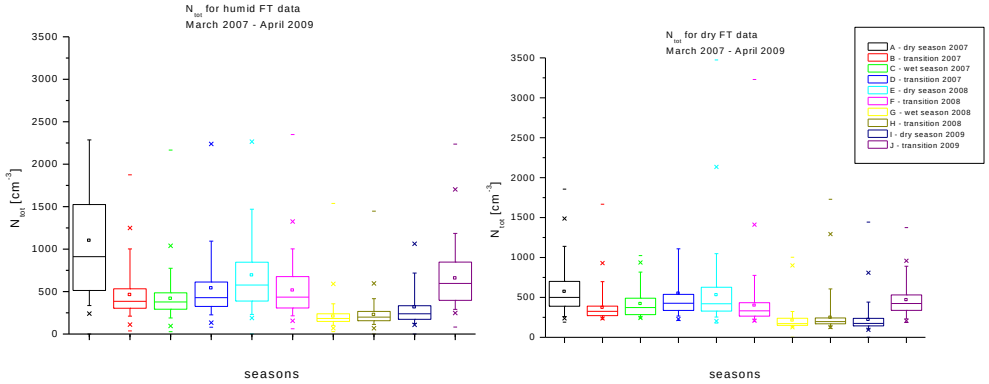

Fig. 3. Box-and-Whisker plots of total particle number concentration for humid FT data (**a**, left panel) and dry FT data (**b**, right panel).

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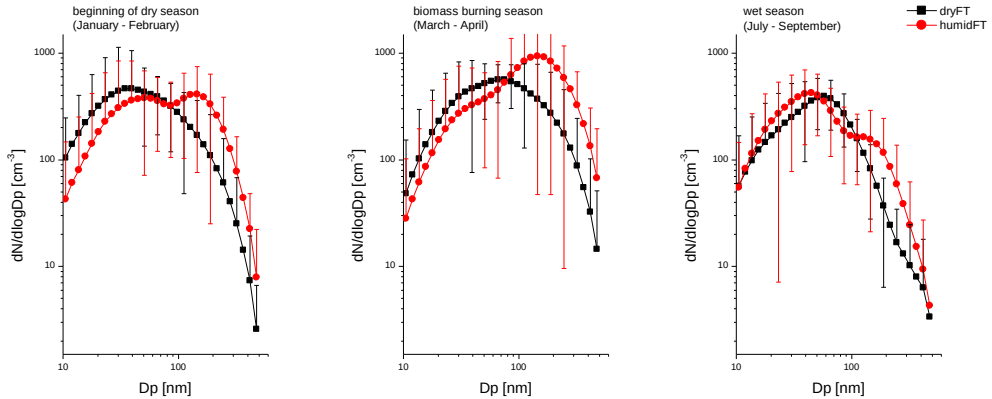

Fig. 4. Mean particle number size distributions (standard deviation only for every second value for better overview).

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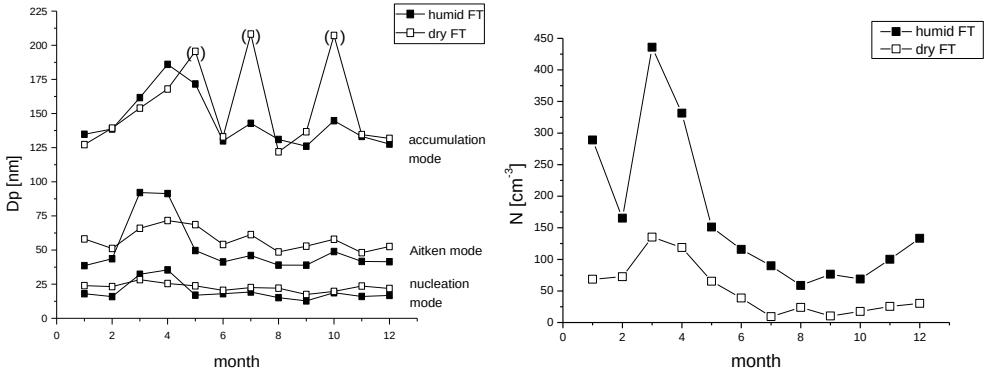

Fig. 5. Diameters and number concentration derived from a least-squares mode fit program for 2008 (number concentration only for accumulation mode particles, Aitken and nucleation mode do not show such significant results and are thus not displayed here); Data points in brackets in Fig. 5a derived under conditions of low data capture (number of size distributions less than 100 for input for averaging, compared to 1000 or more).

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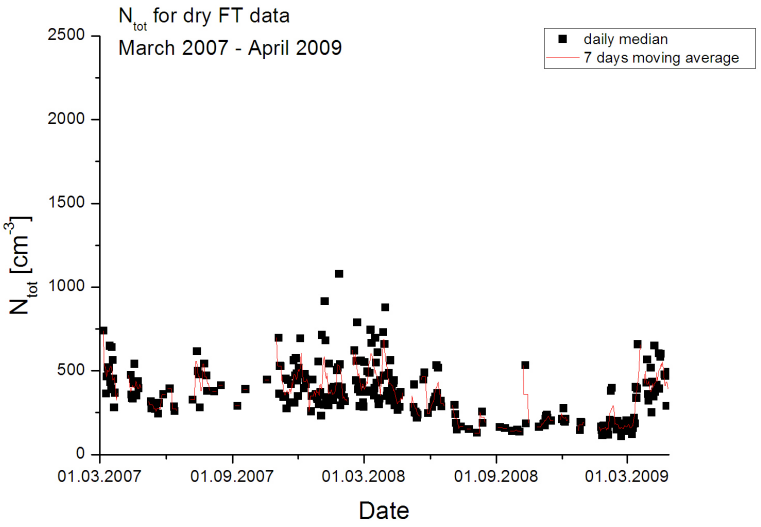

Fig. 6. Relation between specific humidity and number concentration of accumulation mode particles.

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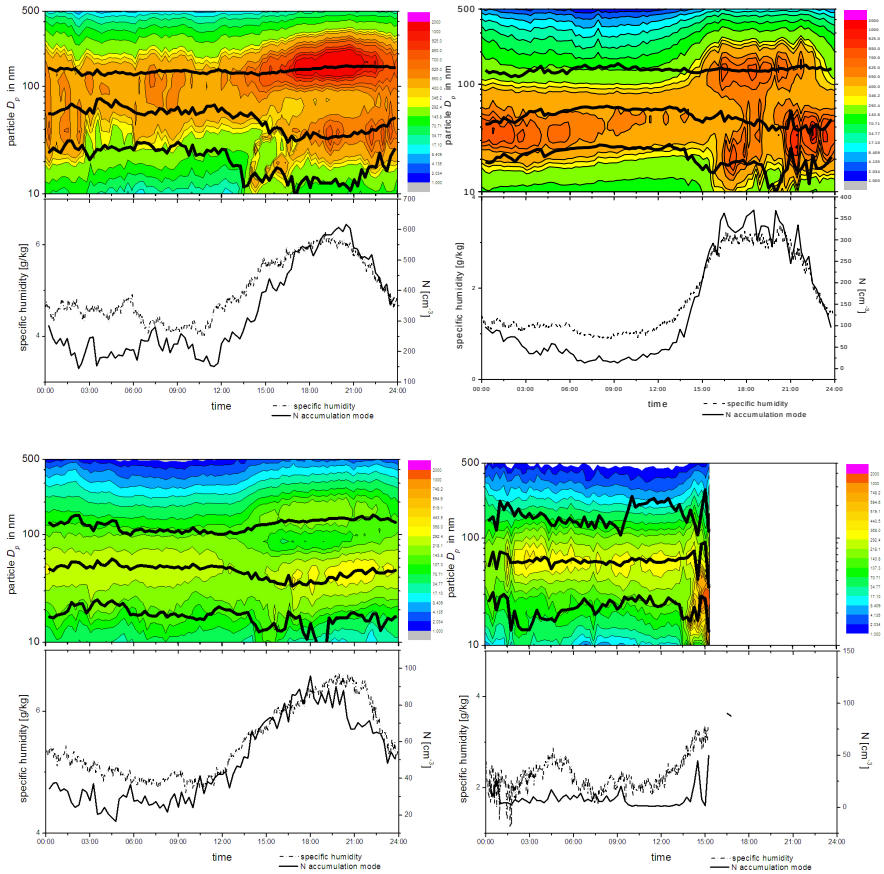

Fig. 7. Contour plots of the daily mean humid and dry FT aerosol during the dry and wet season of 2008, respectively (upper panel, thick lines indicating geometric mean diameters of the accumulation, Aitken and nucleation modes). Corresponding daily mean number concentration of accumulation mode particles and specific humidity (lower panel). **(a)**: Humid FT aerosol, dry season, **(b)**: Dry FT aerosol, dry season, **(c)**: Humid FT aerosol, wet season, **(d)**: Dry FT aerosol, wet season.

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