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Analysis of number size distributions of tropical free tropospheric aerosol particles 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 (FT) were performed from March 2007 until March 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 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 (January–March, $519 \pm 613\,\mathrm{cm}^{-3}$), lowest during the wet season (July–September, $318 \pm 194\,\mathrm{cm}^{-3}$). The more humid FT (relative humidity (RH) range 50–95 %) contained generally higher aerosol particle number concentrations ($573 \pm 768\,\mathrm{cm}^{-3}$ during dry season, $320 \pm 195\,\mathrm{cm}^{-3}$ during wet season) than the dry FT (RH < 50 %, $454 \pm 332\,\mathrm{cm}^{-3}$ during dry season, $275 \pm 172\,\mathrm{cm}^{-3}$ 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.

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1 Introduction

Aerosol particles are omnipresent in our environment, and 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 lifetime and precipitation (Seinfeld and Pandis, 1998; Twomey, 1974). Due to the high variability of the natural as well as the anthropogenic aerosol in the 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 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; Sellegri et al., 2010). 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 GUAN (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, 2B, 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-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., 2005). Besides The Amazon region, a summary of airborne observations over the Pacific Ocean as well as results from the INDOEX experiment (de Reus et al., 2001) show a persistent "horse-shoe" aerosol number density profile in the tropical troposphere with minima and lowest variability in the middle FT (Clarke and Kapustin, 2002). In summary, the body of existing aerosol measurements in the tropical FT has been confined to time-limited field experiments. To our knowledge, long-term observations of aerosols from the tropical FT 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 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 Hadley 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 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 Trade Wind Inversion (TWI). The TWI height is decreasing with distance from the ITCZ.

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 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

2.1 Observation site

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 4765 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), 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 Bolívar (4998 m a.s.l.), the highest Venezuelan mountain. The city of Mérida 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 3200 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.

2.2 Instrumentation

The measurements analyzed in this work cover the period from 6 March 2007 until 31 Mai 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 humidity sensor (Rotronic AG, Bassendorf, Switzerland). 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^{-1} 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 custom-designed Differential Mobility Particle Sizer (DMPS) system. Using a Condensation Particle Counter (model 3010, TSI Inc., Shoreview, MN, USA) it measured across a size spectrum of 10 nm to 470 nm from 5 March 2007 until 12 March 2009. After that date, the size range was extended up to 710 nm. The number size distribution measurements had a time resolution of 15 min. Calibration of the DMPS system was performed by comparing it to a reference DMPS system in the labratory before shipping to Venzuela. During the instrument's operation, flow checks were routinely performed at every service visit. As a general rule, the CPC's sample flow never experienced a change of more than 5%, and the sheath air flow no more than 2% compared to the previous check.

Meteorological parameters, including temperature, pressure, RH, wind speed and wind direction as well as direct and diffuse solar radiation, are continuously detected by an automatic weather station at a time resolution of one minute. Moreover, two webcams were installed that produce images of the southern and northern slopes of the Pico Espejo every 15 min. In-situ ozone concentrations are also measured using an (UV) Photometry Ozone Analyzer (model O341M, Environnement S.A., Poissy, France) with UV irradiation at 254 nm and a detection limit of 0.4 ppb. An analysis of these ozone measurements was published by Calderón et al. (2008).

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 (6 March 2007–31 May 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. Therefore, under cloudy conditions the aerosol examined consists of the interstitial aerosol plus an undefined number of cloud drop residuals. In the absence of clouds, in contrast, the cut-off diameter of the inlet lies well above the range of the DMPS system.

Thus, 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. The authors are aware that a classification of the data set by RH alone is not a perfect procedure. In particular, we took into account that clouds occurring at the station may be patchy. Therefore, different RH ranges with lower threshold values down to 90% for cloud cases were tested. Since the results did not change significantly a threshold value of 95% was appointed as the best choice to remove the majority of clouds. Moreover, the focus of this work is upon long-term trends and a large amount of data is averaged. Thus, an eventual influence of wrongly classified data is assumed to be rather small. 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 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 1 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 data set 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, three lognormal modes, i.e. the nucleation, Aitken and accumulation modes, were fitted by a user-interactive program (Birmili et al., 2001; implemented in LabVIEW 6.1, National Instruments, Austin, USA) using a least-squares algorithm. For a maximum comparability of the fit results, fixed modal spread parameters σ were used, representing the width of each mode (cf. Table 4 below). For a consistent description of the data we decided to always fit three modes, even though some size distributions possessed a rather bimodal character. In such cases, a missing nucleation mode was characterized by a very low number concentration.

3 Results and discussion

3.1 Meteorological conditions at Pico Espejo

As any mountain in the tropics, Pico Espejo is influenced by the seasonal migrating ITCZ and orographically forced atmospheric circulation patterns. This results in a high variability in the routinely measured meteorological parameters, as can be seen in Fig. 1. Especially, RH shows a strong cyclic behavior and is thus the most useful parameter for our subsequent aerosol data analysis. This cyclic behavior over the course of the year leads to a division of the year in 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 2.

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

Table 1. 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%	50%≤RH≤95%	RH<50%	RH out of scale
# size distributions	24 278	32 860	23 033	9827	3267

Table 2. 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	575
max 2007–2009	99.8	3.7	750	577
min 2007–2009	10.6	−3.6	45.3	569
mean dry season	61.9	0.7	478	575
max dry season	98.9	3.3	705	576
min dry season	10.6	−1.8	69.1	573
mean wet season	91.2	−0.3	366	576
max wet season	99.2	−3.6	658	577
min wet season	46.7	2.2	93.7	569

influenced by orographic upslope flow and orographically driven cloudiness.

3.2 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 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 of the mean value are shown for the different seasons and FT types. Regarding the dry season, the mean number concentration of $573 \pm 9\,\mathrm{cm}^{-3}$ for the humid FT aerosol is slightly higher compared to the one of the dry FT aerosol with $454 \pm 4\,\mathrm{cm}^{-3}$ (daily mean values of total aerosol number concentration ± standard deviation of the mean value). With transition to the wet season, particle concentrations for both FT types decrease in average. During the wet season concentrations of $320 \pm 3\,\mathrm{cm}^{-3}$ and $275 \pm 5\,\mathrm{cm}^{-3}$ are observed for the humid and dry FT, respectively.

The basic cause of the variability of the total aerosol number concentration lies in the general circulation. During the wet season (Northern Hemisphere summer), the ITCZ is located almost directly over the measurement station and deep convection is the prevailing weather feature. Consequently, precipitation rates are very high. The removal of fine mode aerosols ($< 1\mu$m) from the troposphere is usually largely due to the incorporation of aerosol particles into cloud droplets

Table 3. Total particle number concentrations at Pico Espejo for different seasons and air mass types. The table indicates mean values and one standard deviation of the mean in cm^{-3}. Total data coverage: 32 860 size distributions, March 2007–March 2009.

season	dry	dry-to-wet	wet	wet-to-dry
humid FT	573 ± 9	489 ± 4	320 ± 3	399 ± 6
dry FT	454 ± 4	390 ± 9	275 ± 5	448 ± 11

followed by precipitation, as well as to scavenging below clouds during rain (Guyon et al., 2003). Moreover, sources of new aerosol particles are weak since the wet season is also the so called non-burning season (Guyon et al., 2003). This results in lower particle concentrations during these months. During the dry season in contrast, the ITCZ is placed further south of the station and therefore the areas of deep convection are located further south of the station as well. Precipitation rates are lower and thus the influence of wet deposition decreases. Agricultural activities in form of deforestation and grassland fires occur more frequently producing a large quantity of emitted particles (Guyon et al., 2003). 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).

The generally higher particle number concentration for the humid FT aerosol compared to the dry FT aerosol is mainly due to the different natures of these two layers of the FT. The

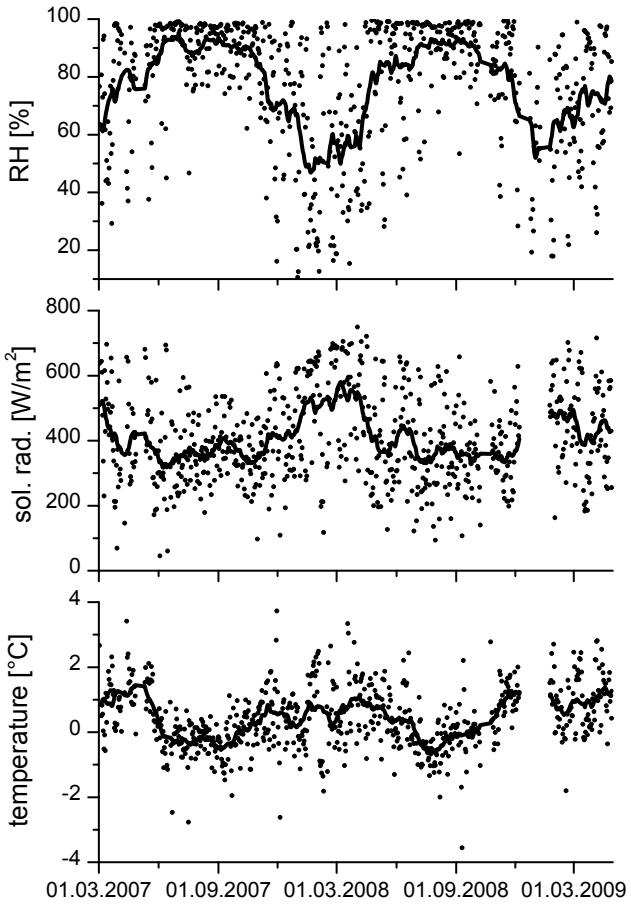

Fig. 1. Multi-annual time history of relative humidity, solar radiation and temperature at Pico Espejo. Dots represent daily averages, lines 30-day moving averages. Solar radiation was averaged for the illuminated part of the day only.

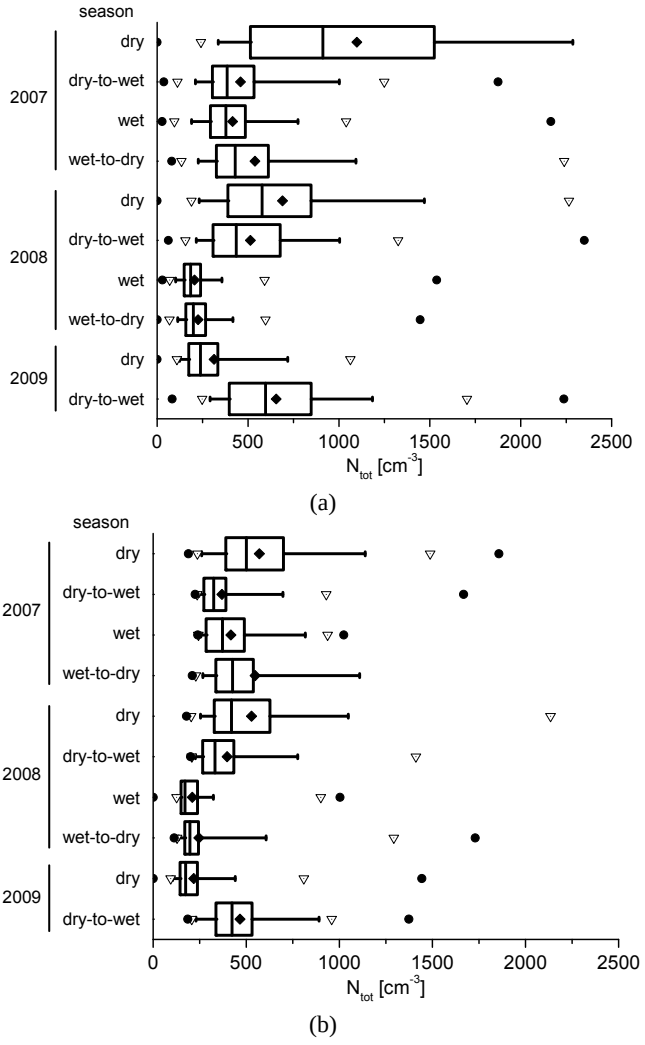

Fig. 2. Box-and-whisker plots of total particle number concentration (10–470 nm) at Pico Espejo: **(a)** humid free troposphere ($50\,\% \leq \mathrm{RH} \leq 95\,\%$), **(b)** dry free troposphere ($\mathrm{RH} < 50\,\%$). In the plots, boxes indicate the 25th, 50th and 75th percentiles. Diamonds indicate arithmetic mean, downward triangles the 5th and 95th percentiles, and circles minimum and maximum values.

humid FT with higher RH values was very likely more recently influenced by convective transport from the boundary layer, which is also linked to the transport of large amounts of water vapor. Deep convection in the tropics has been associated with the formation of new particles (Clarke et al., 1999). From this point of view it is plausible that RH is correlated with particle number concentration. Moreover, boundary layer air masses contain a relative high amount of particulate matter, compared to air masses from the upper troposphere, since most of the aerosol sources are located at or directly above the earth's surface. The dry FT in contrast indicates that the air masses were most likely not affected by convection recently. Furthermore, it is very likely that these air masses spend a longer period of time in the FT, which leads to higher dry deposition rates. When the air masses slowly subside from higher altitudes due to the circulation of the Hadley cell, they experience adiabatic compression and thus warm up. Therefore, RH decreases and reduces the chance for convection. New particle formation due to convection is thus very unlikely for the dry FT aerosol data.

Furthermore, the Box-and-Whisker plots reveal that 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. Furthermore, this results in less amount of data for the dry season of 2007 compared to the other years. These facts are very likely the main reasons 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 because these air masses were most likely recently influenced by convective transport from the boundary layer while the dry FT is rather unaffected by these processes, as already mentioned earlier.

3.3 Annual cycle in particle number size distributions

Aerosol size distributions have been divided the same way as aerosol number concentrations. Due to the different sensitivity of the aerosol sizes to the particle sinks and sources 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 reveals a well defined Aitken and accumulation mode (see Fig. 3). The particle number concentration in the accumulation mode is higher than the one in the Aitken mode. This dominating accumulation mode is the result of an enhanced occurrence of biomass burning during the dry seasons, especially in the area of the Amazon Basin but also in the forests, grasslands and agricultural areas of Venezuela itself. During this time of the year deforestation and pasture cleaning for economic reasons forms a steady source of especially accumulation mode particles with diameters around 100 nm or greater (Martin et al., 2008; Guyon et al., 2003). 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 ($155\,cm^{-3}$). There are no known sources of accumulation mode particles in the FT apart from convective transport from the boundary layer and in-cloud processing of aerosols. These two processes are more common for the humid than for the dry FT. The same behavior can be seen in the right panel of Fig. 4, which displays particle number concentrations for the accumulation mode of the dry and humid FT aerosol derived from a least-squares mode fit program. The left panel of Fig. 4 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. These high values are also reproduced when analysing the data from 2007, 2008 and 2009 separately and occur always at May, July and October. The cause for this phenomenon has not been identified.

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 ($155\,cm^{-3}$) since burning activities are more pronounced. 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 70 nm during the course of the dry season (Fig. 3, see also Fig. 4).

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. As mentioned already with regard to the total aerosol number concentration, this is caused by the shifting of the ITCZ further to the north and the resulting higher precipitation rates. Biomass burning activities stop. In-cloud and below-cloud scavenging are removal processes which affect mainly particles in the accumulation mode size. Thus, during the wet season, the accumulation mode experiences the strongest decrease due to wet deposition and weaker sources on the surface. Aitken mode particles in contrast are to small to be activated in clouds or collected by falling rain drops. Consequently, they remain rather unaffected.

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 constant Aitken mode throughout the whole year. In tropical regions convective clouds form an effective source of new particles (Clarke et al., 1999; 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 in the tropics throughout the year. At the same time, they are rather unaffected by wet deposition processes, as mentioned above. Therefore, a well developed Aitken mode is one of the most pronounced features of the tropical and sub-tropical FT, often resulting in a monomodal, nearly self-preserving aerosol size distribution (de Reus et al., 2001; Krejci et al., 2003). Nucleation mode particles show a similar behavior as Aitken mode particles when analyzing the seasonal variability of their number concentration. But their number concentrations are significantly smaller. 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

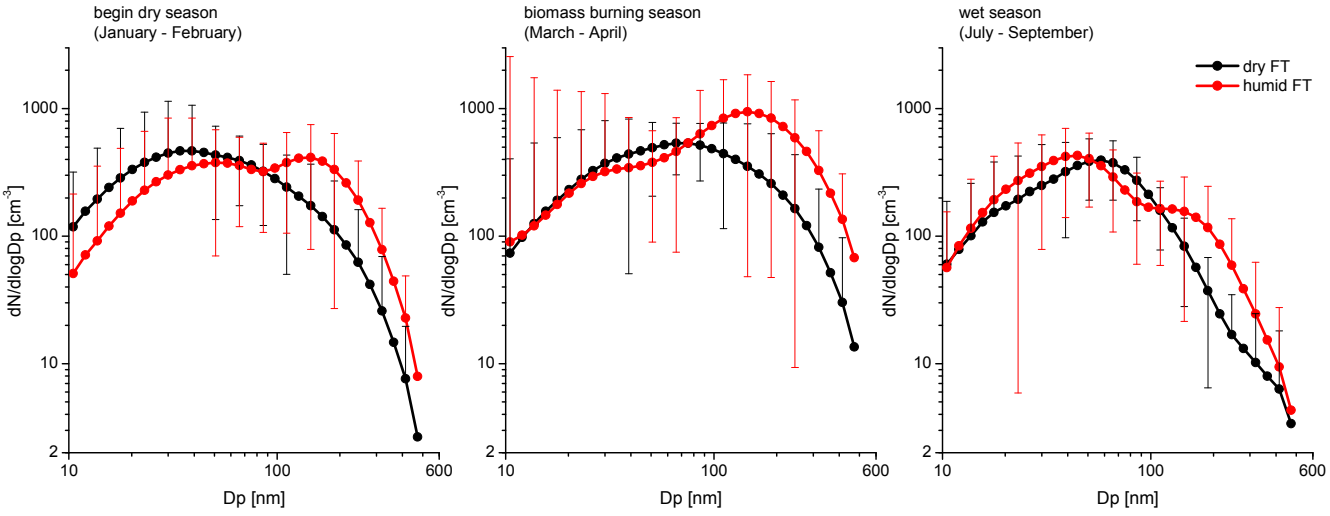

Fig. 3. Mean particle number size distributions covering the period March 2007–March 2009. Whiskers indicate one standard deviation.

Table 4. Geometric mean diameter (nm) and number concentration (p./cm^3) of the three size distribution modes, as derived from multiple lognormal fits. Data coverage: March 2007–March 2009.

	Accumulation mode (σ=1.600)				Aitken mode (σ=1.583)				Nucleation mode (σ=1.562)			
	humid FT		dry FT		humid FT		dry FT		humid FT		dry FT	
month	D_p	N	D_p	N	D_p	N	D_p	N	D_p	N	D_p	N
1	138	295	127	69	40	268	56	189	18	75	23	171
2	139	164	139	75	44	137	50	186	16	26	22	143
3	160	455	153	123	84	178	66	226	27	165	28	170
4	182	350	166	124	75	160	70	241	28	106	20	75
5	171	151	195	66	50	195	68	186	16	45	22	57
6	130	116	132	40	41	248	54	243	18	89	20	84
7	143	90	218	8	46	212	62	200	19	107	23	85
8	131	59	122	24	39	185	49	142	15	37	22	95
9	126	76	139	10	39	213	53	143	13	24	17	47
10	145	69	207	18	48	161	58	216	16	62	20	37
11	133	100	133	29	41	263	47	322	16	54	22	178
12	128	134	132	31	41	275	52	197	16	107	21	171

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. Figure 5 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. The dry FT aerosol goes always together with lower specific humidity values compared to the wet FT aerosol, as supposed to due to the classification via RH. The concentration of accumulation mode particles is highest during the biomass burning season, second highest during the beginning of the dry season and lowest during the wet season. Thus, with regard to the seasonal variability the concentration of these particles is higher, the lower the specific humidity is, resulting in an inversely proportional relationship between these two quantities.

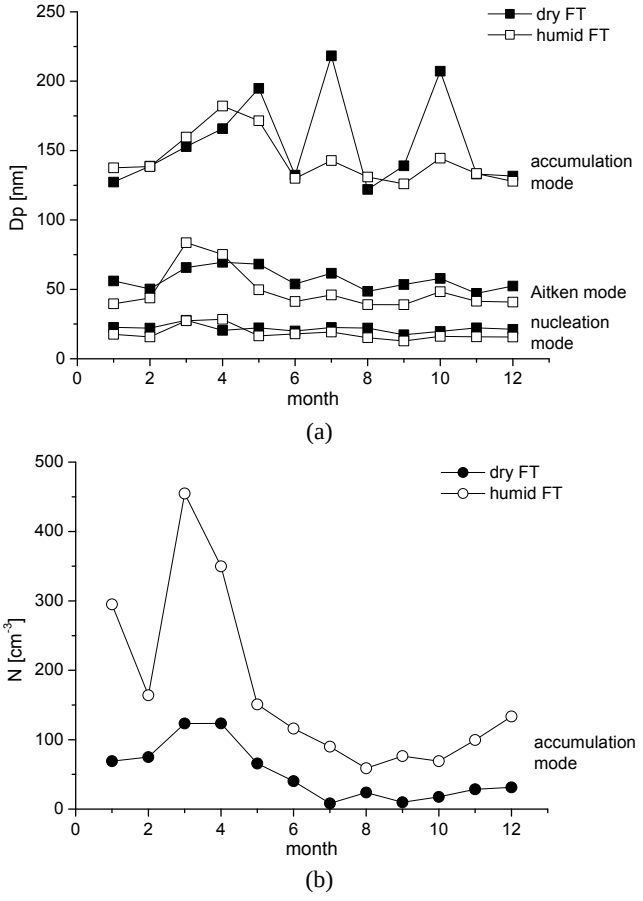

Fig. 4. Annual cycle of **(a)** the geometric mean diameters of three size distribution modes derived from monthly average distributions (data coverage March 2007–March 2009), **(b)** the particle number concentrations for the accumulation mode. The number concentrations for the Aitken and nucleation modes (cf. Table 4) are not displayed because they do not show any significant cyclic behaviour.

3.4 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. 6). Since there is a strong correlation between the daily development of the humidity of the ambient air and the aerosol particle size and number concentration, a calculated mean specific humidity curve is depicted as well as the total mean number concentration of accumulation mode particles. It shows that the aerosol measured at the Pico Espejo station is strongly influenced by radiatively and orographically forced convection.

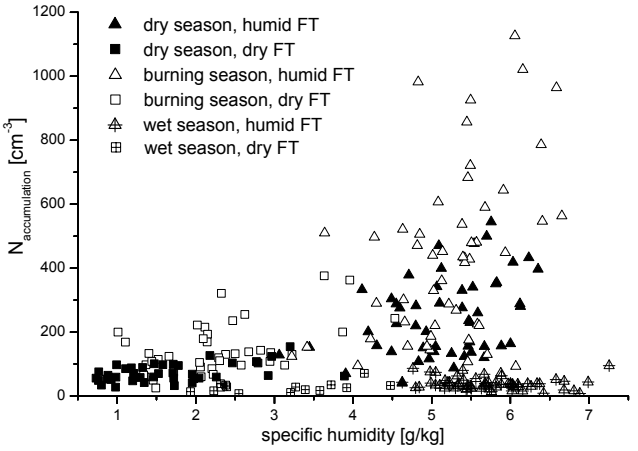

Fig. 5. Relation between specific humidity and the number concentration of accumulation mode particles.

3.4.1 Diurnal cycle during the dry season

In the diurnal cycle for the humid FT aerosol during the dry season (Fig. 6a), 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. The particle number concentration of all three modes increases since particles are carried from the boundary layer to the FT. Moreover, convective cloud outflow creates new particles which can quickly grow to Aitken mode sizes. The strongest influence on the particle number concentration is observed in the accumulation mode (Fig. 6a, bottom graph). However, the mean diameter of the accumulation mode shows no significant changes. The mean mode diameters of the nucleation and Aitken modes in contrast experience a decrease due to dilution of the aged particles by fresh and thus smaller particles. When the specific humidity increases during the afternoon and finally reaches its maximum values, the number concentration of the accumulation mode particles reaches its maximum as well. With beginning of the evening hours the strength of the solar insulation weakens and the convective activities stop. 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. 6b). 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. This is again caused by the

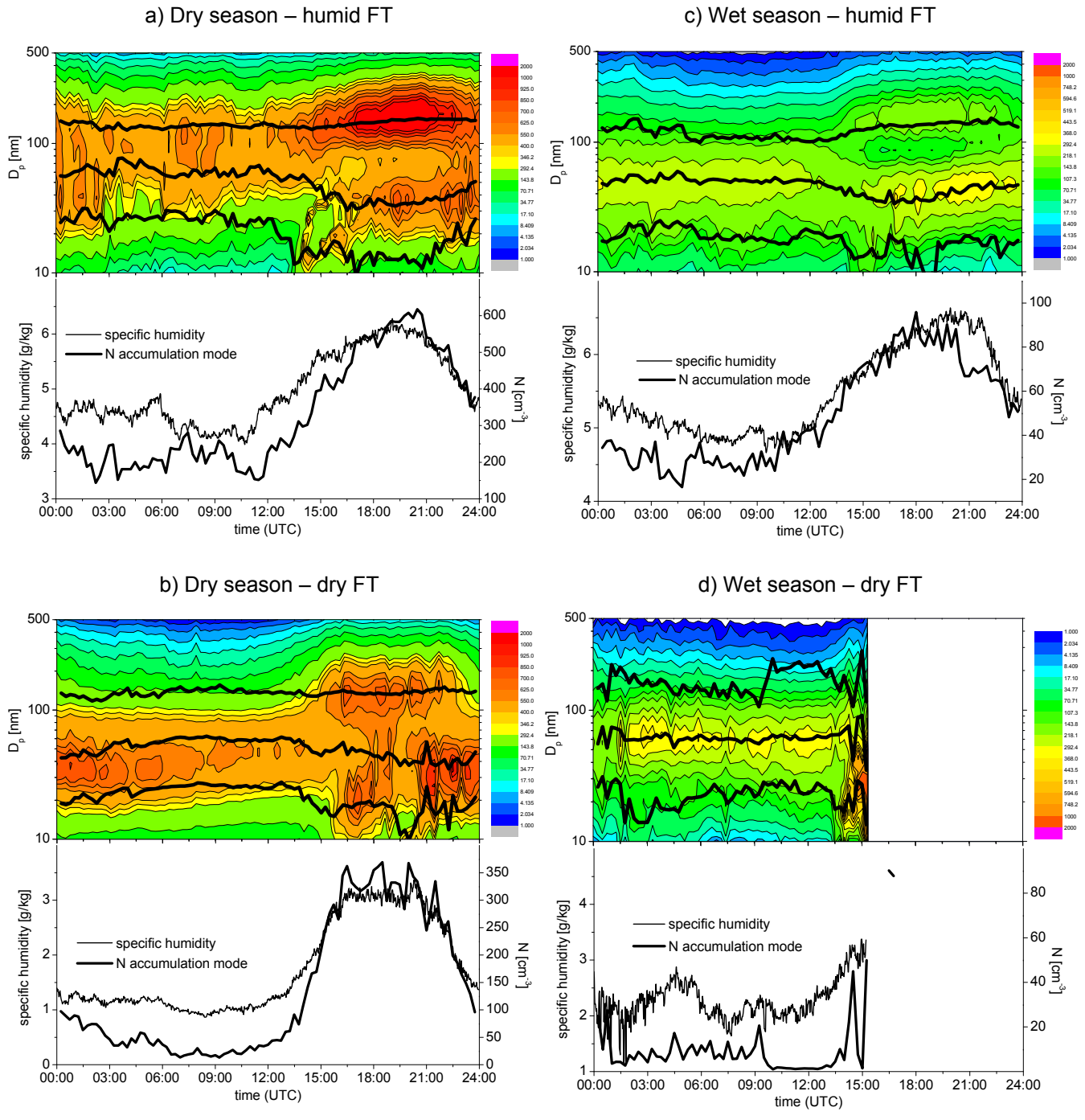

Fig. 6. Mean diurnal cycles of particle number size distributions for 2008. The data is keyed after dry season **(a, b)** and wet season **(c, d)**, as well as humid FT aerosol ($50\,\% \leq \mathrm{RH} \leq 95\,\%$) **(a, c)** and dry FT aerosol ($\mathrm{RH} < 50\,\%$) **(b, d)**. The black lines in each upper graph indicate the corresponding geometric mean diameters of the accumulation, Aitken and nucleation modes. Each bottom graph shows the mean diurnal cycle of accumulation mode number concentration and specific humidity.

transport of fresh particles to the station. The mean diameter of the accumulation mode, in contrast, remains rather constant and thus unaffected by the specific humidity maximum. Increasing specific humidity during the midday hours from about $1.3\,\mathrm{g\,kg^{-1}}$ to approximately $3\,\mathrm{g\,kg^{-1}}$ represents a convective 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. This is due to the fact that the dry FT possesses a generally lower concentration of accumulation mode aerosols and is also not so so strongly affected by convection compared to the humid FT. During the evening hours the mean modal diameter of all modes experience an increase due to aging processes, as already observed for the humid FT.

3.4.2 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. Reason is an enhanced influence of wet deposition processes. 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. 6c), 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. 6c, bottom graph) indicating transport of accumulation particles from the boundary layer to the FT by convection. Furthermore, the mean mode diameters change with altering specific humidity. The diameters of the nucleation and Aitken mode experience a reduction due to dilution by fresh particles while the specific humidity reaches its maximum values. The mean diameter of the accumulation mode, in contrast, increases slightly as already observed for the humid FT aerosol of the dry season. Towards the end of the day, when convection stops the mean mode diameters increase again due to aging processes.

The mean pattern for the diurnal cycle of the dry FT aerosol during the wet season (Fig. 6d) reveals slightly different particle properties. First of all, it is limited by the RH 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. This is on the one hand caused by convective transport from the boundary layer and on the other hand by convective cloud outflow. In the Aitken and accumulation mode, an increase is seen, too, which is also caused by convective transport and a subsequent growth of newly formed particles. 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 10:00 UTC. But afterwards, when the specific humidity shows a constant increase, the mean diameter of the accumulation mode increases as well.

4 Conclusions

On the basis of continuous particle number size distribution measurements at Pico Espejo (Venezuela) this study has gained new insights into the time-dependent behaviour of the tropical free tropospheric (FT) aerosol.

In a first step, the data set referring to cloudy conditions – defined by RH > 95%, were isolated from the rest of the data due to the design features of the air inlet at the station. (The inlet has an undefined upper cut-off size, thus not fully separating cloud droplets from the interstitial aerosol.) The cloud free data (RH $\leq$ 95%) were further divided into humid FT aerosol (50% $\leq$ RH $\leq$ 95%) and dry FT aerosol (RH < 50%) according to the approach suggested by Ryoo et al. (2009).

For both data groups, a cyclic annual behavior was found, which could be linked to the variability of the general circulation: the total particle number concentrations were the highest during the dry season (January–March, $519 \pm 613\,\mathrm{cm}^{-3}$) and the lowest during the wet season (July–September, $318 \pm 194\,\mathrm{cm}^{-3}$). This can be understood by the seasonal migration of the ITCZ, which forms the upwards moving branch of the Hadley cell: during the dry season the ITCZ is located southwards of the station causing a decrease in deep convection and precipitation. Thus, wet removal rates are low. Moreover, biomass burning activities are particularly intense in the dry season, emitting large numbers of primary particles, especially in the size range of the accumulation mode.

During the wet season, in contrast, the ITCZ is located almost directly over the station area. Due to the associated precipitation, biomass burning is strongly suppressed; moreover, the efficient wet deposition is another cause of low accumulation mode particle concentrations. The overall result

is a strong annual cycle in the particle number size distribution, manifested mainly by the accumulation mode. Meanwhile, the Aitken mode concentrations remained relatively constant throughout the year. The latter points to relatively constant source rates of atmospheric Aitken particles in the region.

Looking at the humid and dry FT separately, the humid FT contains a higher particle concentration during all the seasons. One reason is that humid FT air parcels are very likely to be influenced by convective transport from the boundary layer prior to arrival at Pico Espejo. Boundary layer air is expected to contain significantly more particles than FT air due to the emissions of primary particles at ground level as well as the possibility of secondary particle formation from boundary-layer gaseous precursors. In addition, high RH at high altitudes is an indicator of deep convection, which has been associated with the formation of new particles in tropical latitudes (Clarke et al., 1999).

Besides the annual cycle of particle concentrations, the seasonal variability of the general circulation had also visible implications on the diurnal variation of particle concentrations at Pico Espejo: during the dry season, the daily mean particle concentration was higher in all particle modes compared to the wet season. The same aspect is valid when comparing humid FT aerosols (high particle concentrations) against dry FT aerosols (low particle concentrations). Generally, a significant correlation between the particle number concentration (here, especially accumulation mode particles) and specific humidity was observed on the diurnal scale. While on the annual time scale RH and accumulation number concentration show an inverse proportional relationship, they correlate highly on the diurnal scale. The reason is that convective transport carries both, specific humidity and high particle concentrations from the boundary layer to the station. These high particle concentrations are always observed between midday and late afternoon, i.e. when solar insulation leads to the most intense upward motion of boundary layer air. In a mountainous area as around Pico Espejo, upslope winds additionally amplify the convective activity. Towards the end of the day, convection ceases along with decreasing solar insulation, resulting in lower particle concentrations.

Overall, our study provides the first long-term analysis of tropical free tropospheric aerosols, and therefore bears relevance for other scientists as well. The particle number size distributions could be used to validate the predictions of regional and global aerosol models. It would also be interesting to see whether such transport models would reproduce the observed annual and diurnal cycles. Intercomparisons between transport models and observed data might also be helpful to reduce the uncertainties associated with existing parameterizations of aerosol emissions and processing. The study has also helped to better understand the Hadley circulation and its implication on aerosol dispersion, because particle size distributions bear a fingerprint of the related source and removal processes.

Unfortunately, the aerosol measurements at Pico Espejo are currently not operational because of limited access to the site (no cable car). It is, however, planned to resume the aerosol measurements in the near future and to continue the time series presented in this study.

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