Deliverable D12.2: Documentation of tower-based particle flux measurements

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Within ACTRIS-2, measurements of tower-based particle fluxes are made at six sites above different land surface types. Building on previous long-term and campaign-based measurements at Hyytiälä (Finland), Cabauw (the Netherlands) and Auchencorth Moss (UK), ACTRIS-2 supports long-term routine particle flux measurements. In addition, measurements in Košetice (Czech Rebublic), AGORA (Spain) and Pallas (FMI, Finland) were planned to be started during ACTRIS-2. This aim has been successfully met and tower-based particle flux measurements using the eddy covariance (EC) technique are running at all six sites. All measurement setups include a 3D ultrasonic anemometer to provide all three wind components and sonic temperature, and a condensation particle counter (CPC) giving the number concentrations of aerosol particles. This deliverable will describe the instrumentation and measurement setups, which are also summarised in Table1, and give some preliminary results.

Table 1. Instrumentation and measurement setup used at the six different sites: AGORA (Spain), Auchencorth Moss (AMo, UK), Cabauw (the Netherlands), Hyytiälä (Finland), Košetice (Czech Rebublic) and Pallas (Finland). Table does not include supporting meteorological measurements.

	AGORA	AMo	Cabauw	Hyytiälä	Košetice	Pallas
Instruments						
Anemometer	81000 RM-Young	Gill Windmaster	Gill Windmaster	Metek USA-1	Gill Windmaster	Metek USA-1
Analyser (CPC)	TSI 3776	Brechtel MCPC	TSI 3775	TSI 3010	TSI 3775	TSI 3776
Analyser flow rate (measurement volume)	0.05 lpm	0.6 cc s^{-1}	0.3 lpm	1 lpm	0.3 lpm	0.05 lpm
50% cut off size[#]	2.5 nm	7 nm	4 nm	10 nm	4 nm	2.5 nm
Instrument response of CPC	<0.8 s	180 ms	0.65s	0.85 s	0.65 s	<0.8 s
Measurement setup						
Logging rate	10 Hz	10 Hz	10 Hz	1 s	1 s	10 Hz
Sampling line length	10 m	~2.5 m	2.1 m	4.5 m	6 m	~5 m
Sampling line inner diameter	4 mm	3.2 mm	4 mm	4 mm	4 mm	3.2 mm
Flow rate	15 lpm	~ 10 lpm	0.3 lpm	13 lpm	16.7 lpm	1.5 lpm
Measurement height	50 m	3 m	68 m	35 m	80 m	23 m
Sensor separation	15 cm	15 cm	15 cm	20 cm	50 cm	~15 cm

[#] The actual cut-off size of the measurement is further affected by the losses in the sampling line.

AGORA

In Spain, the tower-based particle flux measurements include a measurement campaign AMAPOLA conducted in an olive orchard between 18-29 April 2016, and continuous measurements made in the city of Granada. The AMAPOLA campaign was developed in an olive orchard, where greenhouse gas fluxes are continuously measured in the frame of ICOS (*Integrated Carbon Observation System*) infrastructure. During the period 18-29 April 2016, continuous particle flux measurements in the ICOS tower were carried out. During the campaign, EC particle measurements were done by combining an ultrafine CPC measuring in the range of 2.5 nm-3µm (TSI3776) and a sonic anemometer (81000 RM-Young). Both instruments were measuring with a sampling frequency of 10 Hz. At 50 m, there was also a remote system station including a Halo Streamline Doppler Lidar and an aerosol lidar operating at 355 nm. The Doppler lidar was operated continuously while the aerosol elastic lidar was operated during daytime at 1 Hz frequency. As result of the campaign, 9 days of simultaneous lidar and in-situ measurements were gathered.

After AMAPOLA campaign, the same setup was moved to perform permanent measurements at Granada city providing information about the particle fluxes between an urban surface and atmosphere. In this case, the EC system was set up in the observation tower of the “Parque de las Ciencias”, that is located 250 m away of the rooftop of the Atmospheric Physics Lab at the Andalusian Institute for Earth System Research IISTA-CEAMA, where remote system instruments are operated (Figure 1). The sonic anemometer and the inlet to the CPC are located on top of the tower, 50 m over the surface. The CPC is

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located 10 m below, in a shelter equipped with a Peltier cooling system. The main aerosol line works at 15 lpm, using an external pump, and the CPC flow rate is 1.5 lpm.

Figure 1. Experimental set up for the eddy covariance and remotes sensing setups at the urban area of Granada.

The data analysis of the EC data is in progress, but the diurnal behaviours of preliminary calculated aerosol particle fluxes are given in Figure 2 as separated to workdays and weekends. During the spring and summer months, the urban surface acts as a clear source for aerosols with the rush hours clearly visible in the particle concentrations. Following other studies from urban areas (Martin et al. 2009, Järvi et al. 2009), the two-peak behaviour related to rush hours is not visible from the particle fluxes themselves. The highest particle emissions are seen in late afternoon reaching $3.5*10^9$ particles m^{-2} s^{-1} on workdays and $3*10^9$ particles m^{-2} s^{-1} on weekends. When compared to other European city centres with intense traffic, the particle emissions are at the highest end. The workday particle fluxes remain below $1*10^9$ particles m^{-2} s^{-1} in the city centres of Helsinki and Edinburgh (Kurppa et al. 2015, Dorsey et al. 2002), below $1.5*10^9$ particles m^{-2} s^{-1} in the city of Lecce and Stockholm (Contini et al. 2012, Mårtensson et al. 2006), and below $2*10^9$ particles m^{-2} s^{-1} in Manchester (Martin et al. 2009). However, the analysers and the cut-off sizes vary and therefore direct comparisons are difficult.

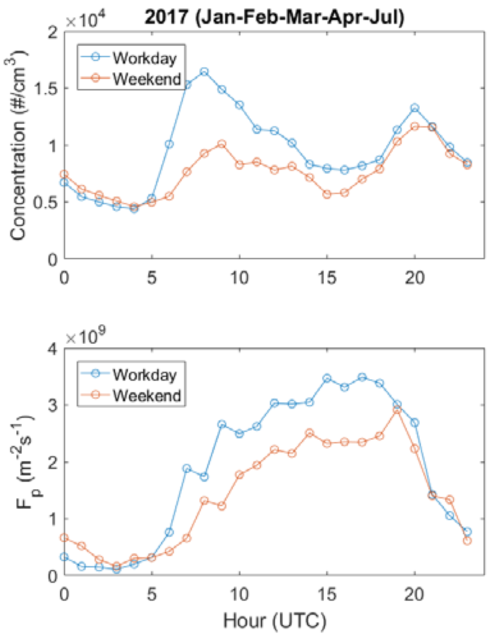

Figure 2. Median values for workdays and weekends of aerosol number concentration (upper panel) and aerosol number fluxes (lower panel) in January-April and July 2017 computed using EDDYPRO.

Auchencorth Moss (UK, NERC)

ACTRIS-2 particle flux measurements started at Auchencorth Moss in June 2016, using an older TSI CPC model (TSI3762) with a large (3 lpm) flow rate and sub-second response time, which was used at this site before (Nemitz et al. 2002). Unfortunately, this instrument failed about 2 weeks into the measurements and a repair was no longer supported by the manufacturer. It took until December 2016 to find institutes-internal funds, for the capital purchase procedures and delivery of a replacement instrument. Because at Auchencorth Moss the measurement height (and thus the eddy size) are small, we opted for the fastest

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CPC commercially available, a Mixing Condensation Particle Counter (MCPC 1720; Brechtel Instruments) with a stated response time of 180 ms and a 50% detection diameter of 7 nm. As far as we are aware it is the first time this MCPC is being used for EC flux measurements. These instruments have been operated at Auchencorth Moss continuously since January 5, 2017, with data being recorded initially at 1 Hz and since 25 March, 2017 at 10 Hz.

During the analysis, data from the MCPC are combined with high frequency measurements made by a Gill R3 sonic anemometer mounted on top of an open lattice tower at a height of 3 m above ground. Air for sampling by the MCPC is drawn down a short length (~2.5 m) of 316 grade stainless steel tubing (1/4" OD, ID 3.2 mm) into the CPC measurement enclosure which is mounted midway up the same lattice tower. A cross-covariance function is used on each 30-min averaging period to estimate the time-lag that exists between the measurement of the vertical wind velocity and corresponding particle number concentration measurement. The data are then re-processed following the recommendations of Langford et al. (2015), using a prescribed time-lag, which changes each day to reflect the median time-lag identified by the cross-covariance function between the hours of 10:00 and 16:00 each day. The identified time-lag was typically around 1.2 s.

Example results are shown in Figure 3. Panel A shows the average diurnal cycle on a monthly basis between March and September, 2017. Figure 3B shows the relationship between the measured deposition velocities and the friction velocity, u_*, which is a measure of turbulence. The open circles show the average deposition velocities, binned by friction velocity. There are two remarkable features to these data. Firstly, the observed deposition velocities are very large relative to previous values reported for semi-natural vegetation and are approximately an order of magnitude larger than the fluxes reported by Nemitz et al., (2002) at the same site. This may relate to the faster response time of the new instrument and improved setup which may limit the high frequency losses, but warrants further investigation to ensure there are no irregularities in the data processing procedure. The second observation is that the relationship between deposition velocity and u_* changes throughout the growing season potentially reflecting the change in canopy height and morphology.

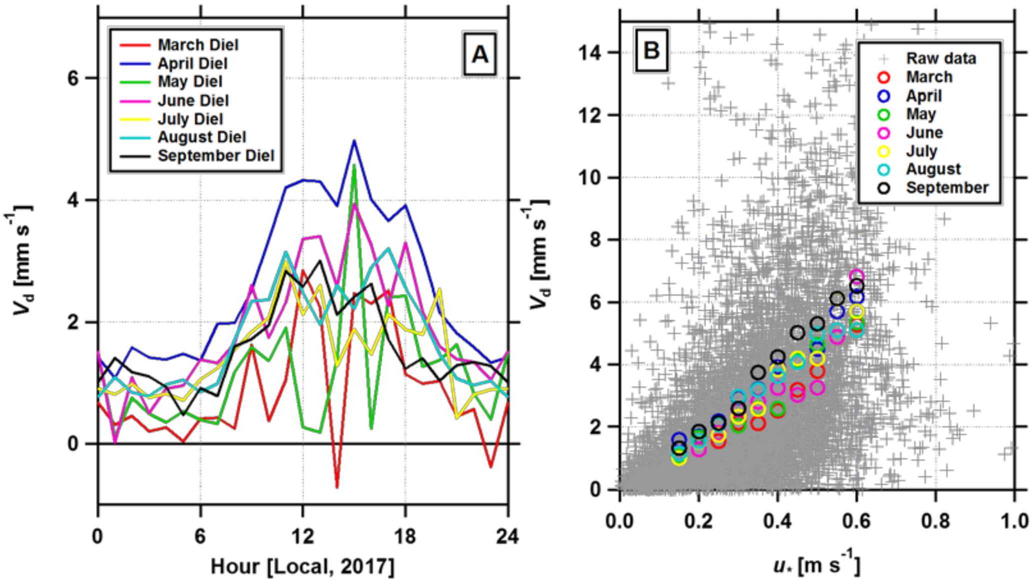

Figure 3. Monthly average diurnal cycles of particle number deposition velocities (V_d) measured above the Auchencorth Moss measurement site between March and September 2017 (Panel A). Panel B show the average deposition velocities binned by friction velocity (u_*).

Cabauw (The Netherlands, KNMI)

The CESAR (*Cabauw Experimental Site for Atmospheric Research*, www.cesar-observatory.nl) Observatory is located in the western part of the Netherlands (51.971° N, 4.927° E) in a polder 0.7 m below average

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sea level. The nearby area is dominated by flat agricultural grassland with relatively little industry and households. In contrast, the wider surrounding area at distances 15 – 50 km away, more than 10 million people live and work in one of the most densely populated areas in Europe. The CESAR Observatory includes a 212 m tall tower specifically built for meteorological and air pollution studies. The EC system measuring the turbulent fluxes of aerosol is placed in the tower at 68 m above ground. The system consists of a Gill 3D ultrasonic anemometer (WindMaster Part 1590-PK-020) and a TSI Condensation Particle Counter (CPC 3775). The CPC detects particles larger than 4 nm in diameter. The sample air, drawn 15 cm apart from the anemometer measurement volume, is transported to the CPC with stainless steel inlet tube of 2.1 m length and with a diameter ¼ inch outer diameter. The delay time between entering the tube and final counting was determined experimentally at 10.5 ± 0.5 s by single pulse exposure to aerosol spray. CPC and anemometer where connected to the same PC with a data acquisition frequency fixed at 10 Hz.

From the beginning of June until 7th July 2016, the experimental setup consisted of the two instruments allocated at different heights. While the ultrasonic anemometer was located at 60 m level, the CPC was downstairs at the base of the mast. The inlet system 68 m in length, consists of 4 PM_{10} size selective inlets and a stainless steel pipe that brings the air in high flow to the basement. The residence time from inlet to CPC is about 60 seconds. Starting from 7th July 2016 until 6th February 2017, the experimental setup for this work was arranged by relocating the CPC at the 60 m level high of the CESAR mast Third Set up. From 6th February 2017 until 1st October 2017, particles of a fixed known diameter a selected by a Differential Mobility Analyser. Two sizes where measured 60 nm and 90 nm.

Figure 4 shows the diurnal variation of aerosol particle concentrations and fluxes measured at the tower at 68 meters. Clear human influence is seen in the concentrations with increased values during the morning and evening rush hours. Particle fluxes show distinct diurnal behavior with higher fluxes during the day and lower during the night.

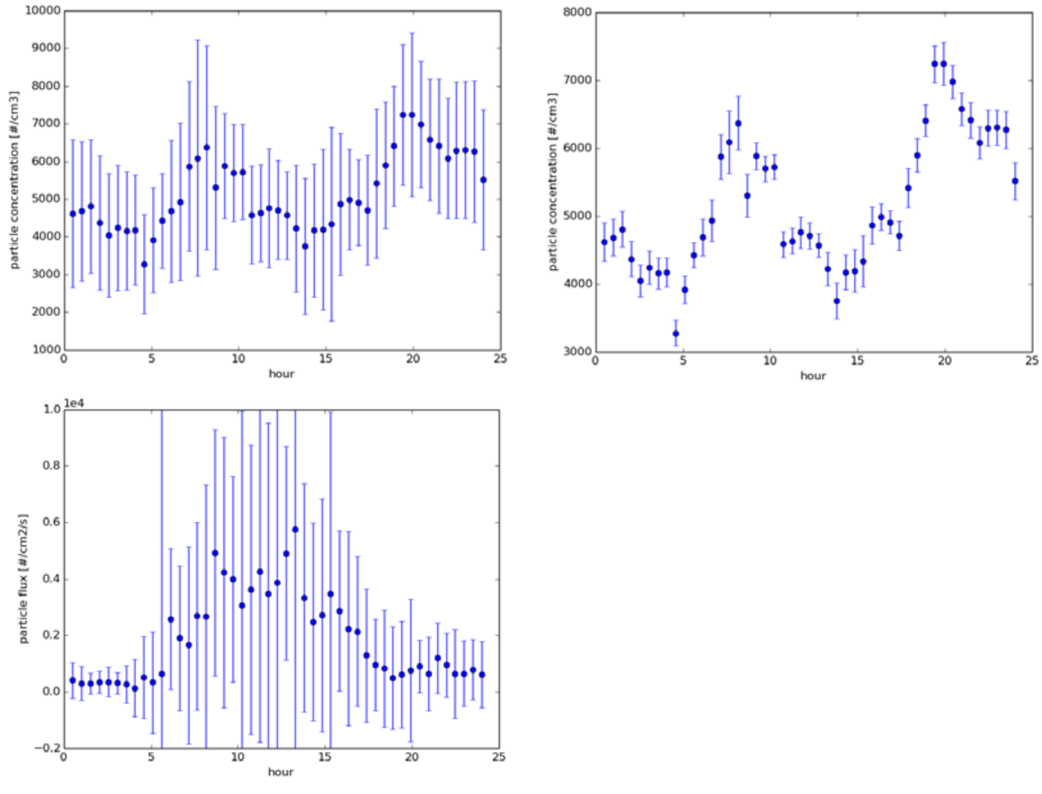

Figure 4. Diurnal results for 7th July 2016 until 6th February 2017. (left) concentration of undifferentiated 'total' CPC with its standard deviation of the distribution. (middle) same as left but now with standard deviation of the mean. (right) corresponding particle flux.

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Hyytiälä (Finland, UHEL)

The SMEAR II (*Station for Measuring Ecosystem-Atmosphere Relations*) is located in Hyytiälä in Southern Finland. The station is located in a boreal forest with majority of the tree species being pine trees. At SMEAR II, the tower-based aerosol particle flux measurements have been ongoing for several years at the height of 23 m above the ground (Rannik et al. 2009). The measurement location however starts be too close to the forest canopy (mean height 15 m in 2007) and therefore within the framework of ACTRIS-2, a second EC setup was installed at the station in spring 2016. This setup is planned to be long-term and after some years of simultaneous measurements between the old and new sites, the old measurement setup will be removed.

The new EC measurement setup comprises of a Metek USA-1 anemometer and TSI3010 particle analyser located 35 m above the ground level. The air to the CPC is drawn through a 4.5 m long tube with inner diameter of 4 mm. The tube inlet is located 21 cm away from the anemometer centre. The flow rate is 20.1 lpm whereas the CPC flow rate is 1 lpm. The new measurement setup has been performing well and as an example the co-spectra between vertical wind speed and aerosol particle concentration calculated over April-August 2016 is shown in Figure 5. The attenuation caused by the instrument response and measurement tube can be seen as difference between the heat flux and particle flux co-spectra at the inertial subrange. Generally, the two EC setups follow each other closely (Figure 6) but slightly higher downward particle fluxes are measured at the new site that at the old site. This is also seen in the deposition velocities between the two measurement setups. Maximum deposition velocity obtained with the new setup is 2.8 mm s^{-1} and with the old setup 1.8 mm s^{-1}. The deposition of aerosol particles to the surface is of similar magnitude at both Hyytiälä and Auchencorth Moss (See Figure 3) with slightly higher deposition velocities at the latter.

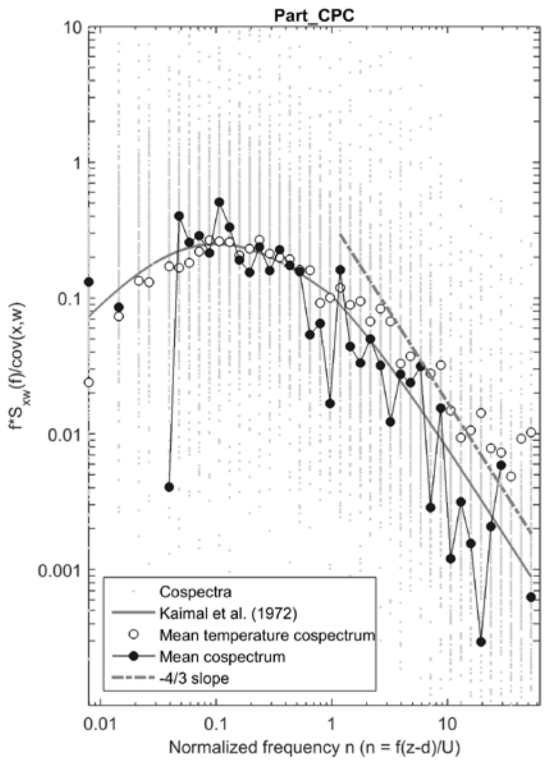

Figure 5. Mean cospectra of vertical wind speed and aerosol particle concentration calculated over April – August 2016. The measurements were made with the new measurement setup at the height of 35 meters.

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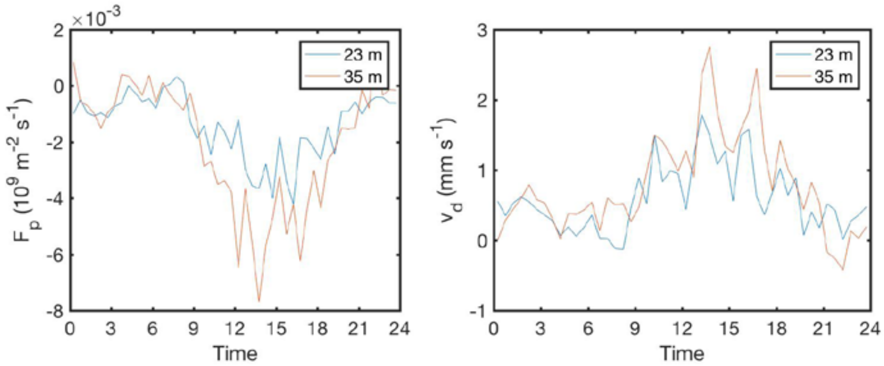

Figure 6. Average diurnal cycle of aerosol particle flux (F_p) and deposition velocity (V_d) measured at the two locations (heights 23 m and 35 m) in Hyytiälä in May-July 2016.

Košetice (Czech Republic, CHMI)

The National Atmospheric Observatory Košetice is situated in the agricultural countryside in the Czech Highlands (N 49°34'24", E 15°04'49", 534 m above sea level). In addition to meteorological parameters, many different air quality ground based parameters are measured at the station (PM_{10}, $PM_{2.5}$, NO_x, NH_x, O_3, SO_x, CO, POPs, VOCs) as well as aerosol characterization (PNSD, light absorption, light scattering, EC/OC). Further measurements are running at the 250 m high atmospheric tall tower. These measurements include vertical gradients of GHGs (CH_4, CO_2, CO, N_2O), O_3 and Hg. Flask sampling of ^{13}C, ^{18}O, H_2, SF_6 and O_2/N_2 are performed at tower platform at 230 m a.s.l.

Additionally, on 26th September 2016 continuous EC measurement in the atmospheric tower with the following setup was started. The setup consists of a Gill ultrasonic anemometer and CPC (TSI3775). The anemometer was placed on a 5 m long shoulder at 80 m height above the ground with sampling frequency 10 Hz. The CPC sampled through a 6 m long sampling tube (inner diameter 4 mm) with frequency 1 Hz and flow rate 0.3 lpm. The CPC sampling inlet was located about 50 cm from the anemometer body and was bent perpendicularly to the ground facing downwards. We used an additional pump (Leckel) set to 16.7 l min^{-1} to shorten the residence time in the sampling train. We stopped the measurement in January 2017 due to anemometer failure.

The continuous measurements started again at the beginning of the Lidar campaign that took place from 15 August till 15 September 2017. A new EC measurement was started on the top of the mast at the height of 250 m on a 1.5 m long shoulder. Similar configuration as the configuration of measurement at 80 m height was set up at the top of the tower. The same model of anemometer (3D Gill ultrasonic) and similar CPC (TSI 3772) with the same sampling frequency were used. CPC was placed into a ventilated box and we used again an auxiliary pump sucking through a critical orifice. The sampling line length was 2 m and distance of the inlet from the anemometer was about 15 cm. After several days of campaign the CPC unfortunately failed and it was not possible to repair it. At the height of 250 m, currently only the 3d anemometer is running.

With the start of the Lidar campaign also an SMPS and APS measurement was set up at the tower platform at 230 m. An SMPS (TSI CPC 3025 and Electrostatic Classifier 3080 with the long DMA 3081) and APS (TSI 3321) were placed into a ventilated box. Aerosol is sampled into the APS through a mini PM10 inlet and about 1.5 m long steel tube with ½ inch i.d. Aerosol for the SMPS is isokinetically subsampled and dried with silica gel diffusion drier. The flow rate for the SMPS is set to 0.3 lpm. Another measurement connected to ACTRIS-2 project is located in the air-conditioned container under the tower. It is a measurement of the smallest aerosol particles, down to 1.2 nm in diameter, using particle size magnifier (Airmodus A11 nCNC-system, consisting of the A10 PSM and the A20 CPC). All obtained data is stored and backed up in the original format.

Figures 8 and 9 show the time series of aerosol particle concentration from the CPC located at 80 meters and from the SMPS at 230 meters, respectively. On both time series, one can see similar diurnal variation measured by both instruments. The data analysis of the EC measurements in currently ongoing and afterwards we can tell more how is the site acting as a sink or source relative to the other ecosystems.

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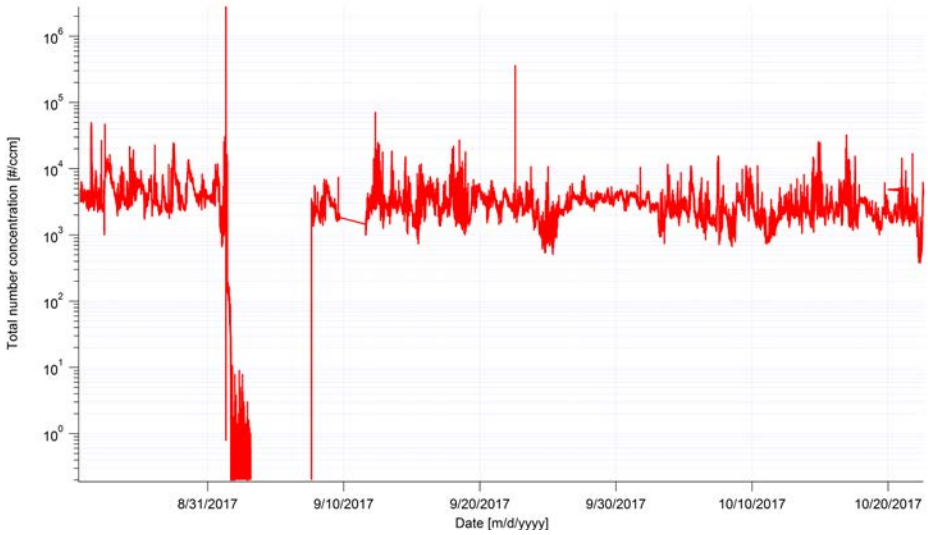

Figure 8. Total number concentration measured by CPC at the height of 80 m on the tall tower.

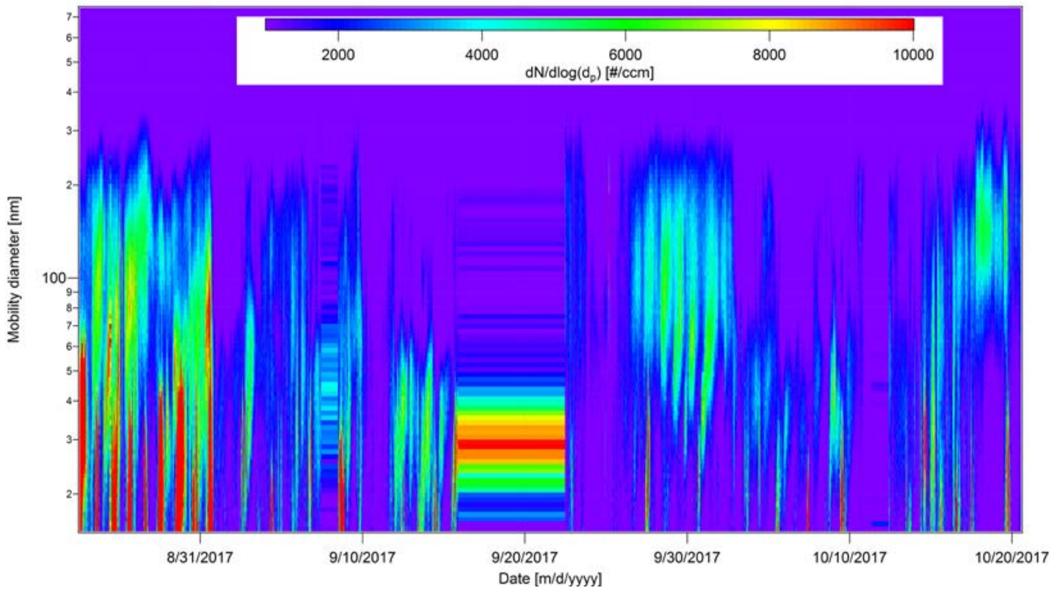

Figure 9. Particle number size distribution spectra measured by SMPS at the height of 230 m on the tall tower.

Pallas (Finland, FMI)

The Pallas research station comprises of versatile infrastructure for monitoring and studying the atmosphere, ecosystems and their interactions. Pallas (67.973°N, 24.116°E) is located 170 km north of the Arctic Circle, partly in the area of Pallas-Yllästunturi National Park. Pallas is a northern node of Pallas-Sodankylä research infrastructure of the Finnish Meteorological Institute, the other node being located in Sodankylä Arctic Research Centre. The main atmospheric measurements are conducted on top of a barren fell called Sammaltunturi (565 m above mean sea level). A supporting ecosystem site, Kenttärova, is situated on a hill (347 m above mean sea level) that is circa 60 m above surrounding planes near to the northern border of the boreal vegetation zone; close to the Sammaltunturi fell. EC measurements of CO_2 has been conducted at the site since 2003.

Continuous tower-based surface particle flux measurements with the EC technique were started at the site in September 2015 and are conducted using an ultrasonic anemometer (Metek USA-1) and a CPC (TSI3776). This setup was added to the existing trace gas flux measurement system using the EC technique

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at height 23 m above the ground level. Low total particle number concentrations and problems with data stream synchronization has proven to be challenging with the present setup. Data stream synchronization after the introduction of the CPC has affected data acquisition of both the CPC and Licor instruments, resulting in deteriorating quality of the trace gas fluxes, in addition to the issues measuring the particle fluxes (Figure 1). This issue can be seen from the lag time of CO_2 before and after the CPC was introduced. The issue is related to the synchronisation only, as both CO_2 measurements and CPC counts themselves compare well with other measurements at the station. Figure 10 also shows how low the CPC counts can be at a clean Arctic site; the poor sampling statistics for low counts can introduce an additional source of uncertainty. The aerosol flow through the CPC 3776 nozzle is only 0.05 L min^{-1}, so changing to another CPC model with greater flow rates could improve the statistics. At present, the *in-situ* flux setup needs both software and hardware updates to provide reliable data.

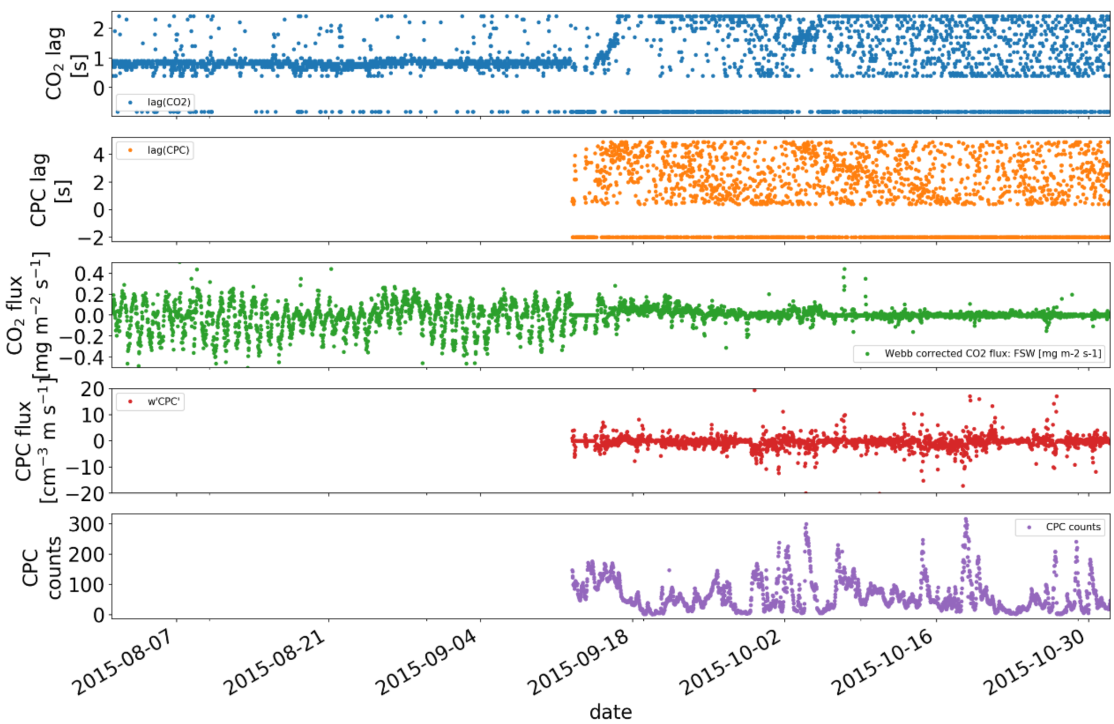

Figure 10: Flux measurements at Kenttärova station (Pallas) before and after introduction of the CPC on 18th September 2015. Upper two panels display calculated lag time for carbon dioxide (CO_2) and CPC measurements, centre panel displays CO_2 flux, lower two panels display CPC flux and CPC counts.

Conclusions

Calculations of final, quality controlled, particle fluxes are currently ongoing from all sites and only after that comparisons of particle fluxes between the different ecosystems can start. Now we can however already say that the highest particle fluxes are measured in Granada and lowest in Pallas following the expected number of sources at the sites. The urban surface acts as a source for particles whereas the vegetation canopies can occasionally act as sinks. Deposition velocities in Hyytiälä, Auchencorth Moss and Cabauw are of the same order of magnitude.

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