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Intercomparison of four different cascade impactors for fine and ultrafine particle sampling in two European locations

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Abstract

Due to the need to better characterise the ultrafine particles fraction and related personal exposure, several impactors have been developed to enable the collection of ultrafine particles (<100 nm). However, to the authors' knowledge there have been no field campaigns to-date intercomparing impactor collection of ultrafine particles. The purpose of this study was two-fold: 1) to assess the performance of a number of conventional and nano-range cascade impactors with regard to the particle mass size distribution under different environmental conditions and aerosol loads and types, and 2) to characterise aerosol size distributions including ultrafine particles using impactors in 2 European locations. The impactors used were: (i) Berner low-pressure impactor (BLPI; 26 nm - 13.5 μm), (ii) nano-Berner low-pressure impactor (nano-BLPI; 11 nm - 1.95 µm) and (iii) Nano-microorifice uniform deposit impactor (nano-Moudi; 10 nm-18 µm), and (iv) Personal cascade impactor Sioutas (PCIS; < 250 nm - 10 µm).

Taking the BLPI as an internal reference, the best agreement regarding mass size distributions was obtained with the nano-BLPI, independently of the aerosol load and aerosol chemical composition. The nano-Moudi showed a good agreement for particle sizes >320 nm, whereas for particle diameters <320 nm this instrument recorded larger mass concentrations in outdoor air than the internal reference. This difference could be due to particle bounce, to the dissociation of semi volatiles in the coarser stages and/or to particle shrinkage during transport through the impactor due to higher temperature inside this impactor. Further research is needed to understand this behaviour.

With regard to the PCIS, their size-resolved mass concentrations were comparable with other impactors for PM_1, PM_2 and PM_{10}, but the cut-off at 250 nm did not seem to be consistent with that of the internal reference.

Keywords: Mass size distribution; Chemical characterization; Ultra-fine particles; Cascade Impactors; Nanoparticles; Ultrafine particles

1 Introduction

Used in numerous areas of air quality research, cascade impactors are established, relatively simple, and robust instruments. They collect airborne aerosols and segregate them into a number of aerodynamic sizes for subsequent determination of mass size distribution, chemical and/or physical properties (Hitzenberger *et al.*, 2004; Seinfeld and Pandis, 2006). The mechanical principle behind size impaction employs the known quantities of Stokes number and slip correction factors to derive particle inertia, therefore ascribing a stopping distance in accordance to particle size (Hinds, 1999). Particulates are collected onto substrates, frequently made of quartz, polytetrafluoroethylene (PTFE; best known as Teflon), polyethylene terephthalate (commonly abbreviated PET, otherwise known as Mylar), polycarbonate or aluminium (Howell *et al.*, 1998; Schaap *et al.*, 2004; Tursic *et al.*, 2008). The choice of substrate is dependent on the type of impactor, sampling conditions and analytical techniques intended to be carried out (Fujitani *et al.*, 2006). A variety of cascade impactor designs have appeared since May (1945) first reported on an initial design to sample coarse aerosols (>2.5

µm). Since then, sampling size fractions for traditionally designed commercially available cascade impactors allowed for particle collection from coarse to fine fractions (<2.5 µm), for example 10 µm - 0.034 µm for the Berner low-pressure impactor (BLPI) (Hering *et al.*, 1978; Berner and Luerzer, 1980; Hillamo and Kauppinen, 1991) and size cuts as small as 0.056 µm for the micro-orifice uniform deposit impactor (Moudi) (Marple *et al.*, 1991).

However, epidemiological studies have evidenced the need to focus on ultrafine particles (UFP; D_p<100 nm), due to their possibly larger impacts on health when compared to coarser particles (Oberdörster, 2000; Oberdorster *et al.*, 2005). Recently, due to the growing need to better characterise the UFP fraction, the second generation of Moudi impactors (Model 122 and Model 125 Nano-Moudi-II™, MSP Corp., Shoreview, MN, USA), both available in the rotating version (122-R and 125-R) and in the non-rotating version (122-NR and 125-NR) and nano-BLPI (not commercially available) were introduced, both adaptions of the original Moudi (Marple *et al.*, 1991) and BLPI impactors (Hering *et al.*, 1978; Berner and Luerzer, 1980; Hillamo and Kauppinen, 1991), modified to enable the collection of UFP down to 11 nm. Also, the need to better understand and characterise personal exposure led to the development of portable, light-weight impactors such as the personal cascade impactor sampler (PCIS; Misra *et al*, 2002).

Due to the physical principle of particle collection associated with all impactors sampling artefacts can occur, including particle bounce, particle blow off, and particle wall loss (Wall *et al.*, 1988; Schwarz *et al.*, 2012). These artefacts vary according to the impactor type (Hillamo and Kauppinen, 1991; Howell *et al.*, 1998; Štefancová *et al.*, 2011) loads, composition of the aerosol sampled (Huang *et al.*, 2004; Sardar *et al.*, 2005; Fujitani *et al.*, 2006; Crilley *et al.*, 2013), and the type of substrate used (Fujitani *et al.*, 2006; Nie *et al.*, 2010). Also, because long sampling time is required for having enough mass of the finest UFP for chemical analysis may produce sampling artefacts of volatilization or absorption.

As well as those previously described, the sampling and accurate sizing of UFP/nanoparticles also present challenges. There is a need to produce a fast flowing jet of air onto an impactor plate, creating the inertia allowing for collection of the smallest size fractions producing a high pressure differential at the lowest cut sizes. This pressure drop changes the vapour pressure in the bulk which can then enhance volatilisation (Hering and Cass, 1999). Attempts to address this issue were successfully carried out by decreasing the pressure drop over a reduced number of stages (Marple *et al.*, 1991; Štefancová *et al.*, 2011). Moreover, the low mass of UFP requires a greater collection concentration which then increases the possibility of mass overloading on the larger fractions. The commercially available Nano-Moudi-II™seeks to reduce jet velocity, pressure drop, particle bounce, re-entrainment and evaporative loss by incorporating micro-orifice nozzles (up to 2000 as small as 50 µm in diameter in the 10 L/min Model 125 and up to 6 000 of 50 µm diameter in the 30 L min^{-1} Model 122). The rotating Nano-Moudi-II™versions (Model 122-R and 125-R) have internal embedded stepper motors for the rotation of the sampling stages, thereby spreading the sample over the filter to reduce build-up (Marple *et al.*, 2014). However, as will be described below, this spreading of the sample may lead to new uncertainties and complications.

Cascade impactors have been deployed in a diverse array of measurement campaigns utilising their versatility, characterising size-fractionated chemical composition of urban aerosols (Sardar *et al.*, 2005; Schwarz *et al.*, 2012), particle volatility (Hering and Cass, 1999; Huang *et al.*, 2004), vapour-particle phase partitioning (Delgado-Saborit *et al.*, 2014), influence of relative humidity (Štefancová *et al.*, 2010), indoor - outdoor relationship (Smolík *et al.*, 2008), archive contamination (Mašková *et al.*, 2015), metals in particles collected near a busy road (Lin *et al.*, 2005; Karanasiou *et al.*, 2007; Ondráček *et al.*, 2011), size-segregated emission particles in a coal-fired power station (Tursic *et al.*, 2008), whilst extensive theoretical investigations and experimental characterization of cascade impactors tended to focus on the performance of one type of cascade impactor (Biswas and Flagan, 1984; Wang and John, 1988; Štefancová *et al.*, 2011; Jiménez and Ballester, 2011; Marple *et al.*, 2014). Howell *et al.* (1998) carried out an intercomparison of 'traditional' BLPI and Moudi impactors during a field campaign. Field campaigns usually provide a greater variation of conditions than controlled laboratory based conditions, offering a more robust analysis of comparable instrumentation. Another notable intercomparison study was conducted by Pennanen *et al.* (2007) who tested a modified 4-stage Harvard high-volume cascade impactor against a reference 10-stage BLPI in 6 different European locations over different seasons. The authors note the implicit effects on individual impactors of meteorology and aerosol composition. Other studies have run two or more impactors in tandem measuring simultaneously indoors and outdoors (Smolík *et al.*, 2008; Mašková *et al.*, 2015), to cover extended particle size distributions (Geller *et al.*, 2002), or characterise artefacts caused by particle volatility (Huang *et al.*, 2004; Schaap *et al.*, 2004) or changes in size distribution due to different relative humidity (Štefancová *et al.*, 2010).

To the authors' knowledge there has been no field campaign to-date intercomparing impactor collection efficiency of UFP. As a result, this paper seeks to address this by assessing the performance of a number of conventional and nano-range impactors, namely Berner low-pressure impactor (BLPI, 25/0.018/2, Hauke, Austria), nano-Berner low-pressure impactor (nano-BLPI, 10/0,01, Hauke, Austria), nano-microorifice uniform deposit impactor (Nano-Moudi-II™, MSP Corp., Shoreview, MN, USA Model 125R; U.S. Patent # 6,431,014B1) and Personal cascade impactor Sioutas (SioutasTM PCIS, SKC Inc.; Misra *et al*, 2002), by means of two intercomparison exercises, one in Prague, during winter 2015, and other in Barcelona during summer 2015. The aim of the campaigns was to test the instruments' performance under different environmental conditions and aerosol loads and types. Our work reports on the impactor performances not only with regard to different particle size distributions but also aerosol composition and meteorology.

2 Methodology

2.1 Sampling sites and sampling set-up

2.1.1 Prague

The field intercomparison initially took place in outdoor air (6th-23rd February 2015) and it was subsequently moved indoors (23rd February 2015 - 2nd March 2015) in Prague, Suchdol at the Institute of Chemical Process Fundamentals (ICPF), Academy of Sciences of the Czech Republic (ASCR) compound (50°7'36.47"N, 14°23'5.51"E, 277 m.a.s.l). Suchdol is a residential area in north-western Prague, about 6 km from the city centre. It is recognized as a suburban background site with residential houses and a university campus interspersed between plenty of green spaces. The traffic flow is moderate along one major 2-lane road (average traffic of 10000-15000 vehicles day^{-1}) with regular bus services. Due to its location on a plateau above the river Vltava there are not many contributory roads alongside (Figure S1). Detailed information of the area where the impactors were located were previously provided by Smolík *et al.* (2008) and Hussein *et al.* (2006).

Outdoor sampling consisted of 3 weekend sampling periods (6 - 9th, 13 - 16th and 20th - 23rd February 2015), and 2 week-day samplings, (10 - 12th and 17 - 20st).

In addition, indoor samples were also collected during 2 week-day samplings (23rd - 25th and 25th - 27th February 2015) and a final 3-day weekend sampling period (27th February 2015 -

2nd March 2015). This resulted in a total of 5 valid outdoor samples (three weekend and two week-day) and two valid indoor samples (one weekend and one week-day). For both outdoor and indoor sampling, the weekend runs started on the preceding Friday between 11:00h-13:00h local time and finished at 9:00h local time on the following Monday. The week-day samplings started between 11h00-14h00 and terminated at 9h00. The sample duration in Prague was defined based on the experience from previous research (Smolík *et al.*, 2008; Štefancová *et al.*, 2011). Based on ambient PM concentrations it was considered that samples should be collected over no more than 72 hours, to avoid substrate overload.

2.1.2 Barcelona

The Barcelona intercomparison was conducted exclusively outdoors at an air quality monitoring station at IDAEA-CSIC located in an urban background site in the southwest of Barcelona (41°23′14″ N, 02°06′56″E,, 78 m.a.s.l) from 18th May to 3rd July 2015 (Figure S2). The sampling site, described in detail by Reche *et al.* (2015), is influenced by vehicular emissions from one of the city's main traffic avenues (Diagonal avenue), located at approximately 200 m from the site and with a mean traffic density of 90 000 vehicles day L min^{-1} (Amato *et al.*, 2015). Even though the site is officially classified as urban background, it is located in a city with very high road traffic and influenced by the emissions of one of the largest arterial roads of the city.

Outdoor sampling in Barcelona consisted of 4 valid week-day sampling runs, each run accounting for 96h (4 days duration sampling time). The runs started every Monday between 10:00h-12:00h local time and finished on Fridays around 14:00h-16:00h local time. The sample duration in Barcelona was set longer than in Prague since the averages of particle mass collected during a sampling less than 4 days would not be sufficient for further chemical analysis. Indoor intercomparisons were not carried out due to the absence of an appropriate location for indoor air sampling.

2.2 Instrument set-up and experimental specifications

In the present study, the mass size distribution of the aerosol was measured by different types of cascade impactors:

- A Berner low-pressure impactor (BLPI, 25/0.018/2, Hauke, Austria; (Berner et al., 1979; Preining and Berner, 1979) which collects particles onto PET foils (Mylar 13 µm thick) (flow rate 24.8 L min^{-1}). The impactors separated particle mass into 10 size fractions. The cut diameters of the stages were 0.026, 0.056, 0.1, 0.16, 0.25, 0.43, 0.86, 1.73, 3.425, and 6.61 µm (Štefancová *et al.*, 2011). The impactors were equipped with inlets with the cut-point calculated as 14 µm.

- A modified BLPI (denominated as nano-BLPI, 10/0.01, Hauke, Austria) collecting particles on PET foils (Mylar 13 µm thick) (flow rate 17.2 L min^{-1}) from 0.01 µm to 1.95 µm in 8 size stages. The aerodynamic cut diameters of stages 1 to 8 were 0.011, 0.024, 0.039, 0.062, 0.095, 0.24, 0.49, 1.0 µm, and the inlet cut-point was calculated as 1.95 µm. Given that the nano-BLPI is a custom made instrument, the design parameters of each of its impaction stages are shown in Table S1 in the supporting information.
- A nano-microorifice uniform deposit area impactor (Nano-Moudi-II™, MSP Corp., Shoreview, MN, USA Model 125R; U.S. Patent # 6,431,014B1) equipped with PTFE filters (with diameters of 47 mm) was used to collect size-resolved aerosol samples. This impactor effectively separated the particulate matter into 13 stages with nominal cut diameters of 0.010, 0.018, 0.03, 0.06, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6, 10 µm and the inlet cut-point as 18 µm when operated at an inlet flow rate of 10 L min^{-1}.
- Three personal cascade impactor samplers (SioutasTM PCIS, SKC Inc; Misra *et al*, 2002) operating with a flow rate of 9 L min^{-1} at a pressure drop of 11 inches of H_2O (2.7 kPa). Particles can be separated in the following aerodynamic particle diameter ranges: <0.25; 0.25 to 0.5; 0.5 to 1.0; 1.0 to 2.5; and >2.5 µm. The collection substrates were 37 mm PTFE filters (Pall) or quartz fibre filters (Pall) for the < 0.25 µm stage and 25 mm PTFE filters (Pall) for the 0.25-2.5 µm and >2.5µm stages. Two of the PCIS deployed in Prague separated particle mass in all of the 5 size fractions while another unit collected particles only at 3 of the stages (< 0.25 µm; 0.25-2.5 µm and >2.5 µm). In order to facilitate interpretation of the data, a lower cut diameter of 30 nm was assumed for the last filter stage of particles < 0.25 µm (quasi-UFP).

All the cascade impactors were loaded with uncoated substrates to avoid possible interferences in future chemical analysis (mainly, determination of organics), so the particle bounce that might occur during dry collection has to be considered excepting for the case of BLPI which foils were coated with a thin layer of vacuum grease (Apiezon L, Apiezon products, M&I Materials Ltd, Manchester, England) to ensure adherence of deposited particles and reduce the artefact of bounce.

For the Prague winter intercomparison, the abovementioned six different impactors were deployed simultaneously in both outdoor and indoor sampling periods. The cascade impactors and their inlets were positioned outside above the roof of ICPF building, 285 m.a.s.l. The nano-Moudi, in order to protect its electrical components, was kept inside an air-conditioned cabin with a temperature continually lower than 20˚C and a metal pipe (about 300 cm long) was extended through the roof of the building. With regard to indoor sampling, the impactors were placed inside Laboratory of Aerosol Chemistry and Physics experimental hall on the 2nd floor where office and other experimental activities take place. In both campaigns (indoor and outdoor), the pump exhausts were extended far of the sampling spots in order to avoid sampling artefacts.

For the Barcelona summer intercomparison, the same cascade impactors were deployed (except for the PCIS) at the urban background monitoring site located in IDAEA-CSIC (78 m.a.s.l; South West part of the city) within the University Campus and they were positioned under a plastic shelter to protect them from rain while allowing free ventilation. All the impactor pumps were placed 5 m distance from the impactors whilst long tubes (10 m) were connected to the exhausts to avoid contamination of the samples.

The error in the sampling flow rate and sampled volume in both campaigns was < 5%. Thus, it is assumed that flow rates did not affect the particle size cut-offs. The uncertainty in the particle mass concentration determination was < 15% except in some cases for the smallest stages of nano-BLPI and nano-Moudi impactor which reached mass value deviations > 20 % (standard deviation).

The specifications of the campaigns and the impactors deployed in the intercomparison study are summarized in Table 1. The BLPI was used as internal reference for the size distribution in this study as it was calibrated with the method described by Hillamo and Kauppinen (1991) for the fine stages and by Štefancová *et al.* (2011) for coarse stages. For the intercomparison, the modal pattern of aerosol mass size distribution was divided into four size groups: (i) PM_{10} (D_p<10 μm), (ii) PM_2 (D_p<2 μm), (iii) PM_1 (D_p<1 μm) and (iv) $PM_{0.25}$ (D_p<0.25 μm) particles. Approximate lower cut points for those selected size fractions are shown in Table S2 in the supplementary information.

Table 1. Impactors deployed in Prague and Barcelona and their specifications.

Impactor type	**BLPI**	**nano-BLPI**	**nano-Moudi**	**PCIS (5 stages)** [c]	**PCIS (3 stages)** [d]
Number of samplings in Prague	5x outdoor (3x weekend-days + 2x week-days) 2 x indoor (1xweekend-days + 1x week-days)	5x outdoor (3x weekend-days + 2x week-days) 2 x indoor (1xweekend-days + 1x week-days)	5x outdoor (3x weekend-days + 2x week-days) 2 x indoor (1xweekend-days + 1x week-days)	5x outdoor (3x weekend-days + 2x week-days) 2 x indoor (1xweekend-days + 1x week-days)	5x outdoor (3x weekend-days + 2x week-days) 2 x indoor (1xweekend-days + 1x week-days)
Number of samplings in in Barcelona	4 x outdoor (4 x week-days)	4 x outdoor (4 x week-days)	4 x outdoor (4 x week-days)	N/A	N/A
Flow rate (L min^{-1})[a]	24.8	17.2	10	9	9
Sampling substrates	PET foils (MYLAR) 13 µm thick	PET foils (MYLAR) 13 µm thick	PTFE 47 mm	37 mm PTFE filters (Pall) < 0.25 µm stage and 25 mm PTFE filters (Pall) for the 0.25-2.5 µm and 2.5-10 µm stages	37 mm quartz-fibre filters (Pall) < 0.25 µm stage and 25 mm PTFE filters (Pall) for the 0.25-2.5 µm and >2.5 µm stages
Nº Stages	10	8	13	5	3
Lower cut sizes (µm) [b]	0.026	0.011	0.01	0.03	0.03
	0.056	0.024	0.018	0.25	0.25
	0.10	0.039	0.032	0.50	2.50
	0.16	0.062	0.056	1.00	
	0.25	0.095	0.10	2.50	
	0.43	0.24	0.18		
	0.86	0.49	0.32		
	1.73	1.0	0.56		
	3.42		1.00		
	6.61		1.80		
			3.20		
			5.60		
			10		
Inlet cut-point (µm)	14	1.95	18	10	>2.5

[a] Volumetric flow rate at 20°C and ambient pressure

[b] All sizes are aerodynamic equivalent diameters

[c] Two units deployed; A cyclone was installed ahead which cut PM$_{10}$

[d] One single unit deployed

N/A – Not available

2.3 Sample conservation and gravimetric analysis

Particle mass concentrations on impactor substrates were gravimetrically determined by pre- and post-weighing the Mylar foils and filters (PTFE and quartz fiber) with a Sartorius M5P-000V001 electronic microbalance in Prague and a Mettler MT5 electronic microbalance in Barcelona, both with a ±1 μg sensitivity. Blank samples (1 per sample) were collected per each impactor type in both intercomparison (Prague and Barcelona) for each of the sampling periods. The deviation of mass values due to varying conditions was corrected with the help of the corresponding blanks.

All samples were equilibrated for a period of 24 hours before weighing in a temperature and relative humidity controlled room. The electrostatic charges of the filters were removed using an U-shaped electrostatic neutralizer (Haug, type PRX U) in Prague and a zerostat anti-static instrument (Z108812-1EA, Sigma-Aldrich Co. LLC.) in Barcelona. Each sample was weighed three times with an accuracy of mass determination of ± 2 μg. After weighing, the sampled foils and filters were stored in the freezer at -18 °C.

2.4 Ion chromatography analysis

Ion chromatography analysis were only carried out for the Prague samples and for the BLPI, nano-BLPI and nano-Moudi impactors with the aim to support the interpretation of the particle mass size distributions data. The PCIS filters were not analysed due to the differences observed for the finest size fraction with the other impactors, as will be discussed below.

The whole nano-Moudi impactor samples were extracted in 7 ml of ultrapure water. In case of the Berner impactors, approximately 1/3 of each foil with samples from each stage was cut out and number of aerosol spots on cut piece was calculated. The ratio between cut and total number of spots at each impactor stage was used to recalculate results to overall ion amount on each stage. All samples were then extracted with 7 ml of ultrapure water, sonicated for 30 min in ultrasonic bath and shaken for 1 hour using a shaker. The extracts were then analyzed using a Dionex 5000 system both for cations (Na^+, NH_4^+, K^+, Ca^{2+} and Mg^{2+}) and anions (SO_4^{2-}, NO_3^-, Cl^-) in parallel. An IonPac AS11-HC 2 x 250 mm column was used for anions using hydroxide eluent, IonPac CS18 2 x 250 mm for cations using methane sulfonic acid solution as an eluent. Both anion and cation set-up were equipped with electrochemical suppressors. External calibration was done using NIST traceable calibration solutions.

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3 Results

3.1 Meteorological data and mean aerosol concentrations in outdoor air

Table 2 displays the meteorological data (ambient temperature, relative humidity, ambient pressure and wind speed), the mean and standard deviations (±σ) of aerosol concentrations for Prague and Barcelona and season during sampling with BLPI.

Table 2. Meteorological data and mean daily aerosol concentrations in outdoor air in Prague from 6th to 23rd February 2015 and in in Barcelona from 18th May to 3rd July 2015.

Sampling site	**Temperature (ºC)**		**Relative humidity (RH, %)**		**Barometric pressure recalculated to sea level (mbar)**	**Wind Speed (km h^{-1})**	**Mean PM$_{14}$* (µg m^{-3})**
	Min	Max	Min	Max			
Prague (winter)	-3.4±2.6	3.9±3.3	51±15.4	92±2.1	1023±9.4	12.5±6.6	34.6 ± 15.8
Barcelona (summer)	18±3.3	26±3.3	39±9.9	85±7.1	1018±3.1	12±2.6	15.2 ± 2.1

During the winter campaign in outdoor air from 6th to 23rd February 2015 in Prague, the daily maximum average temperature was 3.9±3.3 ºC and the minimum average temperature was -3.4±2.6 ºC. The relative humidity varied in the range of 51-92% from day to day.

As expected, higher temperatures during summer were monitored in Barcelona from 18th May to 3rd July 2015 (minimum of 18±3 ºC and maximum of 26±3 ºC). However, slightly lower RH (minimum of 39±10 % and maximum of 85±7%), similar pressure (1018±3 mbar) and wind speed (12±3 km h^{-1}) values were recorded. The results imply that aqueous particles may have been collected on an impaction stage different from the stage where they ought to be collected due to the flow-induced relative humidity changes during the day (Fang *et al.*, 1991; Štefancová *et al.*, 2010). Aqueous particles can shrink due to evaporation caused by pressure drop through the impactor and/or grow due to condensation caused by aerodynamic cooling. Also, a distortion of the size distribution due to bounce-off should not be neglected for Barcelona given that foils were not greased prior to sampling.

In Prague, the mean PM$_{14}$ mass concentration measured outdoors (with BLPI) was 34.6 ± 15.8 µg m^{-3} whilst in Barcelona (with BLPI) it was 15.2 ± 2.1 µg m^{-3} (Table 2), in agreement with previous results from 2008 winter campaign in ICPF (Schwarz *et al*. 2012; PM$_{14}$=34 µg m^{-3}) and of the same order of magnitude as PM$_{10}$ from a 2014 summer campaign in the monitoring station at IDAEA-CSIC (PM$_{10}$=19.6 µg m^{-3}). The reason of higher averages of particle mass concentrations in winter in Prague than in summer in Barcelona are due to higher emissions (mainly due to coal and biomass burning used for residential heating) and meteorological conditions such as the lower mixing heights of the boundary layer or even temperature inversions occurring in Prague (Schwarz *et al.*, 2012).

3.2 Average particle mass concentrations per stage for the different impactors

To estimate the cumulative mass concentration for the different size ranges in each of the impactors, the integrated curve of the measured particle mass size distributions was determined by Eq. 1:

$$M_i = M_{i-1} + \int_{D_{pi-1}}^{D_{pi}} \frac{dM}{dlogD_p} \times dlogD_p \qquad \text{Eq. (1)}$$

Where, M is the estimated mass concentration for each impactor stage i, D_{pi-1} and D_{pi} are respectively the lower and upper cut-off diameters of the impactor stage i

The cumulative curves of the particle mass size distributions from Prague (indoor and outdoor) and Barcelona are shown in Figures 1 and 2, respectively.

Results show that the nano-BLPI behaved similarly to the internal reference considered for this work (BLPI), especially for particles larger than 250 nm. Outdoors and indoors, the nano-Moudi was in agreement with the BLPI for particles larger than 320 nm (independent of the aerosol load and type). However, for particles below 320 nm, the particle mass concentration of the nano-Moudi tended to be higher than for the BLPI, especially during winter in Prague. In indoor air, the nano-Moudi cumulative curve of the mass size distributions was closer to the curve obtained for the BLPI impactor.

While in Prague, the nano-Moudi mass size distributions for particles >1 µm were lower than the rest of the impactors, in Barcelona, this trend was not so evident (Figure 1 and 2). This different behaviour could be ascribed to a number of causes: (a) in outdoor air the effect of particle bounce and/or the shrinkage of semi volatile compounds may have caused a shift in particle mass towards the lower sizes of the nano-Moudi, especially in winter in Prague; and/or (b) indoors, the mechanism of the nano-Moudi of spreading the sample (rotating plates), with the increase in temperature, both in indoor air and inside the nano-Moudi shell,

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could favour particle dissociation/evaporation from the PTFE filters and thus result in lower mass loads across the lower size ranges, and thus the nano-Moudi curve would appear to be closer to the internal reference BLPI. This effect would not be so prominent in outdoor air, given that the instrument does not reach such high temperatures. Nie *et al.* (2010) also attributed the loss of volatile compounds to the increase of the temperature inside the MOUDI. However, nitrate concentrations were low in indoor air (see sections below), and therefore the volatilization of this species would have had a low impact on particle mass (leaving only the organic fraction to account for this). Further research is necessary to clarify the different behaviours observed.

The size-fractionated average mass concentrations ($PM_{0.25}$, PM_1, PM_2 and PM_{10}) collected by each impactor along with standard errors deviation (±σ) in the respective size fractions, using data from a total of 5 experiments outdoors and 2 indoors in Prague, and a total of 4 valid samples outdoors in Barcelona are summarised in Figure 3. Approximate cut points for the selected size fractions are shown in Table S2 in the supporting information. However, it is important to take into account that some differences in the results could be partially attributed to the differences in the real cut points for the selected size fractions.

The average PM_{14} mass concentrations and corresponding standard deviation obtained using the internal reference (BLPI) in Prague outdoors were 34.6 ± 15.8 µg m^{-3}. In Barcelona, the PM_{14} mass concentrations and standard deviation in summer were 15.2 ± 2.1 µg m^{-3}. Comparison of independent data from Grimm (corrected with high volume sampling) and the impactors with PM_1 and PM_{10} size cuts, was carried out for the outdoor campaign in Barcelona (4 samples). A slope of 0.98 and a R^2 of 0.7 was obtained for the PM_{14} for BLPI with PM_{10} from an online laser spectrometer (corrected with regard to reference instrumentation) whereas for PM_1, a slope of 0.7 and a better fit of the data was obtained (R^2=0.9). Similarly to BLPI, the nano-BLPI shows a slope of 0.7 and a R^2 of 1 for the cut point PM_1. The mass differences detected for PM_1 suggest that impactors sampling artefacts such as particle blow off, particle wall losses and/or particle bounce occurred.

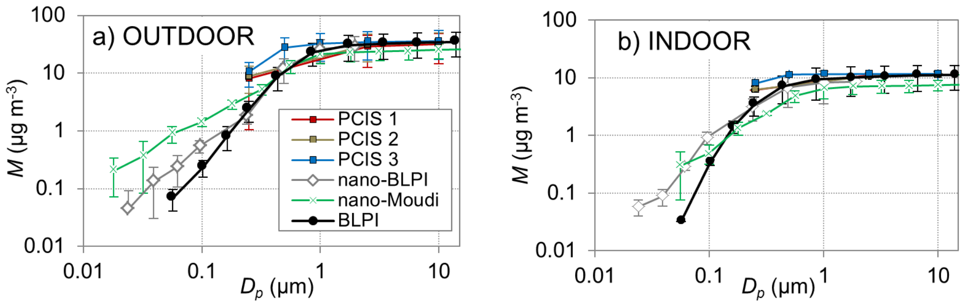

Figure 1. Cumulative mass concentrations measured by the six impactors in Prague: (a) outdoors and (b) indoors. Error bars indicate the standard deviation (±σ).

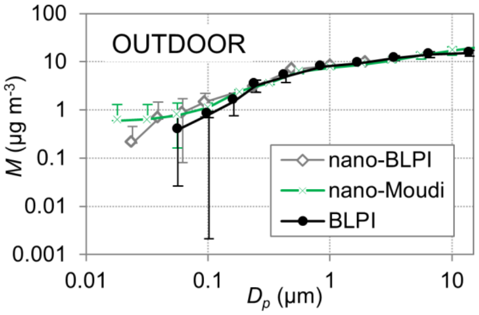

Figure 2. Cumulative mass concentrations measured by the three impactors in Barcelona, outdoors. Error bars indicate the standard deviation (±σ).

As shown in Figure 3, the largest relative difference between the average mass concentrations collected with the three impactors (PCIS, nano-BLPI and nano-Moudi) and the internal reference (BLPI) was calculated for the $PM_{0.25}$ size fraction measured outdoors in Prague by PCIS and nano-Moudi, when concentrations were larger by 354 and 126 %, respectively. The best agreement between the three impactors and the internal reference was obtained in the Barcelona summer campaign.

Intercomparisons between the nano-BLPI impactor and the reference BLPI indicate an overall good agreement with absolute differences in mass concentrations per size fraction being <30%, independent of the aerosol type. A consistent underestimation of the particle mass concentrations for the $PM_{0.25}$ size fractions was obtained with the nano-BLPI for all campaigns and locations (Figure 3). This consistent underestimation was in the order of 5 and 22% outdoors in Barcelona and Prague, respectively, and 10% indoors in Prague, for $PM_{0.25}$. As for PM_1, a slight overestimation of mass concentrations with regard to the BLPI was obtained by the nano-BLPI in both sampling campaigns outdoors. The largest deviation in this size fraction was obtained in Prague outdoors (15%) whereas the smallest difference was obtained in Barcelona (5%). Similar to the $PM_{0.25}$ fraction, the PM_1 and PM_2 concentrations obtained indoors by the nano-BLPI were lower (12 and 15%, respectively) than those of the BLPI.

As for the nano-Moudi, it consistently measured lower PM_1 and PM_2 concentrations in all campaigns (max difference obtained indoors for PM_1 = 31% and PM_2 = 30 %). These differences can be explained by the difference in the cut points given that PM_1 and PM_2 fractions from the BLPI are actually 0.86 μm and 1.7 μm, respectively. For quasi-UFP mass concentrations were significantly higher (126%) in Prague outdoors, whereas the disagreement with the BLPI was reduced in Barcelona outdoors (14%). Finally, in indoor air, concentrations registered by the nano-Moudi were lower (30%) than the BLPI, in Prague.

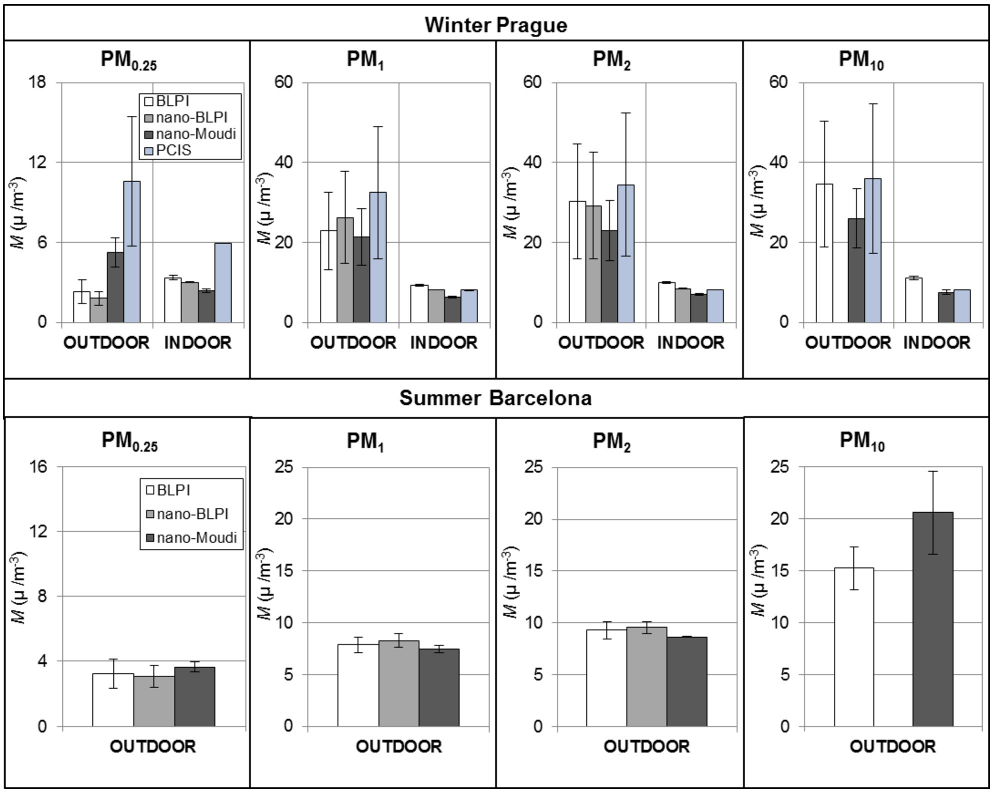

Figure 3. Average mass concentrations collected by the impactors in approximate size ranges: $PM_{0.25}$, PM_1, PM_2 and PM_{10}, during winter Prague campaign (top) and summer

Barcelona Campaign (bottom). Error bars indicate the standard deviation (±σ) of the measured concentrations. The PCIS data correspond to PCIS 3 from Figure 1, where more particle size fractions were collected.

Finally, the portable PCIS were only used in Prague during winter given the differences obtained with regard to the BLPI for the quasi-ultrafine size mode $PM_{0.25}$ (354%). A similar pattern was observed for indoor air, although with a relatively smaller, but still high difference (75%). A possible reason for the discrepancies observed regarding the $PM_{0.25}$ fraction could be ascribed to the different pressure drops across the impactor stages originating from different flow rates (e.g., PCIS 9 L min^{-1} vs. BLPI 24.8 L min^{-1}). The higher pressure drop in the stationary impactors (e.g., BLPI) may increase the probability of volatilisation of semi-volatile species during prolonged sampling, and could contribute to an underestimation of the $PM_{0.25}$ when compared to the PCIS (Sioutas, 2004).

The differences with regard to the coarse fractions were much smaller when compared to the quasi-UFP fractions (<[±42%] and <[±27%] in outdoors and indoors, respectively). In outdoor air, the PCIS showed consistently higher PM_1, PM_2 and PM_{10} concentrations (42, 14 and 4%, respectively). Similar results were reported by Sioutas (2004) where an average ratio PCIS to Moudi (Model 110, MSP Corp, Minneapolis, MN) of 2.02 (± 0.59) and 1.21 (± 0.35) was reported for an aerodynamic size range < 0.25 μm and 2.5-1 μm, respectively. However, in indoor air a consistently underestimation (12, 16 and 21 % for PM_1, PM_2 and PM_{10}), was observed.

In summary, for the aerosols and sampling conditions in this work, the PCIS provided comparable size-resolved mass concentrations for PM_1, PM_2 and PM_{10} while the cut-off at 250 nm did not seem to be consistent with the internal reference BLPI. In order to fully understand these phenomena, a more systematic evaluation might be required. For this reason, data from PCIS will not be discussed in the following sections.

3.3 Aerosol mass size distributions

3.3.1 Particle size distribution in outdoor air

The average particle mass size distributions obtained in the outdoor intercomparison study (Prague and Barcelona) can be found in Figure 4.

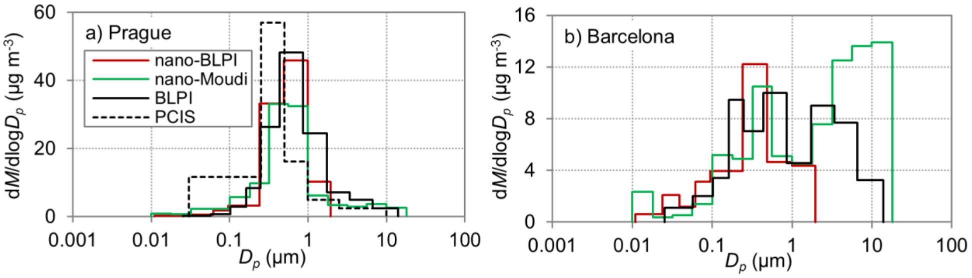

Figure 4. Average mass size distributions obtained outdoors: (a) winter in Prague and (b) summer in Barcelona.

As can be seen, the particle mass size distributions are very different depending on the season and sampling location. During winter in Prague (outdoors), the mass size distributions have a predominantly fine mode, with the coarse mode being almost negligible (by all impactors). The maximum mass concentration obtained in the fine size fraction mode was between 0.4-0.9 µm, whereas in summer in Barcelona, this maximum was shifted towards smaller size fractions between 0.2 and 0.4 µm. In addition to the different aerosol types, this shift to lower sizes might be caused by a lower average relative humidity during sampling in Barcelona that could have caused the particle drying (Tables 2) and therefore, be a reason for particle bounce (Fang *et al.*, 1991; Štefancová *et al.*, 2010).

While in Prague during winter the coarse mode was mostly insignificant, in Barcelona during summer the mass size distributions were clearly bimodal, with larger coarse mode concentrations (Figure 4). The coarse mode obtained may be due to mineral and marine aerosol contributions in the study area (Querol *et al.*, 2008).

The majority of mass concentrations were found in the accumulation mode (PM_1) for both campaigns (7.9 ± 0.7 µg m^{-3} and 22.9 ± 9.8 µg m^{-3} according the internal reference BLPI in summer Barcelona and winter Prague, respectively). With the increase in mass there was an increase in agreement between the impactors, where the closest agreement was observed (between 200-600 nm) (Figure 4).

Figure 4 reveals that the nano-Moudi recorded higher particle mass concentrations in the ultrafine range (<100 nm) than the reference BLPI during winter in Prague (5 samples in total outdoors). Although differences were smaller, the same is true for the Barcelona summer campaign (4-week sampling, Figure 4). As previously mentioned, to protect the electrical components of the nano-Moudi during winter campaign in Prague outdoors, it was kept inside a climate controlled cabin with a temperature continually lower than 20˚C. At these temperatures dissociation of ammonium nitrate can still occur at a slow rate (Smolík *et al.*, 2008). In addition, during the sampling, an increase of temperature inside the nano-Moudi shell was detected due to the internal mechanism of spreading the sample (rotating plates) which generates heat. It is therefore likely that the internal temperature in the nano-Moudi was higher than that of the cabin and thus led to particle volatilisation (Štefancová *et al.*, 2010). The lower nitrate and chloride concentrations in the accumulation mode on the nano-Moudi filters (see below) would support this interpretation. It is also known that a 5ºC difference between the PTFE filter (of the type used in the nano-Moudi) and sampling temperature may accelerate the dissociation of ammonium nitrate on PTFE filters up to 20% (Hering and Cass, 1999). The BLPI and nano-BLPI have no internal warming mechanisms and were located outdoors in Prague and Barcelona, so it is expected that lower volatilisation would occur in these scenarios. However, drying of particles before they are deposited on a substrate may happen also in the BLPI and nano-BLPI due to higher pressure drops (at equivalent sizes) in comparison with the nano-Moudi. This would increase the driving force for evaporation at those stages, which would encourage particle shrinkage. All these previous facts (temperature, RH, high surface area) appear to enhance the evaporation of semi-volatiles (and dissociation of ammonium nitrate) and therefore particle shrinkage during transport through the nano-Moudi. Also, the residence time of particles inside the nano-Moudi low pressure stages is longer due to the lower volumetric flow rate in this instrument. All of this could thus explain the mass size distributions from the nano-Moudi being skewed towards smaller particle fractions during the Barcelona and Prague campaigns (Figure 4). It should be stated that the rotation of the impaction plates and the nozzle plates of the nano-Moudi was specifically designed to achieve a uniform deposit on the collection substrates and therefore, eradicate the particle bounce-off artefact (Marple *et al.*, 2014) that may otherwise occur. Particle bounce-off would only be expected when collecting particles in dry conditions such as in Barcelona (< 50% RH) (Table 2) or indoors. Finally, the overall internal volumes in the low pressure stages seem similar in all of the impactors tested; however, this would need experimental confirmation.

3.3.2 Particle size distribution in indoor air

In Prague, indoor concentrations were lower than outdoors mainly due to a change in weather conditions resulting in cleaner air masses during sampling periods (Figure 4 and Figure 5). Reduced penetration efficiency and faster settling times probably explain the lower indoor coarse mode mass obtained (Figure 5; Hussein *et al*, 2007). Once again, the nano-BLPI measured similar mass concentrations to the reference BLPI (Figure 3) while the nano-Moudi recorded notably lower mass from fine to coarse modes. In addition, the nano-Moudi size distribution showed a slight shift towards larger particle sizes (Figure 5). The difference between the BLPIs and the nano-Moudi could suggest that the latter underestimated mass during this campaign for all particle cut sizes. Initially this would appear to reduce the possibility of volatility losses being responsible for this difference, as ammonium nitrate dissociates readily indoors thereby causing equal losses to all impactors (Lunden *et al.*, 2003). However, because of the way the sample is spread across the substrate in the nano-Moudi, as described above, the ammonium nitrate collected would be more prone to volatilization than that collected on the other impactors. Therefore it could be considered that the mechanism of the nano-Moudi of spreading the sample (rotating plates), with the increase in temperatures, both indoors and inside the nano-Moudi shell, could enhance dissociation/evaporation from the nano-Moudi PTFE substrates. This conclusion can be supported by Figures 6 and 7, which show significantly lower mass concentrations of major species of ammonium nitrate with the nano-Moudi, in comparison with the BLPI.

A number of sources of uncertainty in this interpretation should be taken into account:

a) Increased uncertainty in the mass determination due to lower mass concentrations and shorter sampling times

b) No blank correction available for nano-Moudi IC data

c) No uncertainty calculations for mass determinations available for nano-Moudi, possibly resulting in negative mass concentrations in the lower stages

d) Only 2 valid samples available for indoor air (for all impactors)

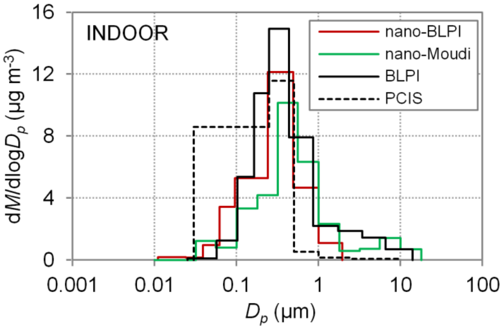

Figure 5. Average mass size distributions in Prague during winter in indoor air.

3.3.3 Size distribution of inorganic ions

Figures 6 and 7 show the particle mass size distributions of major (SO_4^{2-}, NO_3^- and NH_4^+) and minor (Cl^-, Na^+, K^+, Mg^{2+} and Ca^{2+}) aerosol constituents for the winter campaign in Prague in outdoor and indoor air, respectively. In the winter in Prague, the mass size distributions of components have a predominantly fine mode (< 1 µm), with the coarse mode being almost negligible in winter in Prague (by all impactors) but highly significant in Barcelona during summer, such as the case for BLPI.

While the fine mode was dominant for the particle mass concentration and all the predominant aerosol constituents (SO_4^{2-}, NO_3^- and NH_4^+) for both indoor and outdoor air during winter in Prague, the average mass size distributions for minor species (Cl^-, Na^+, K^+, Mg^{2+} and Ca^{2+}), were clearly multimodal (Figures 6 and 7). Similar mass size distributions of these species were obtained by the nano-BLPI and the reference BLPI both outdoors and indoors in Prague. However marked differences in the mass size distributions of these species were observed with the nano-Moudi impactor. In outdoor air there is a clear decrease of NO_3^- and NH_4^+ concentrations measured with the nano-Moudi, confirming the interpretations provided in the previous sections. This is also visible, even if less pronounced, indoors. In addition, outdoors in Prague, the mass size distributions obtained by the BLPI showed that Ca^{2+}, Na^+ and Mg^{2+} were dominated by coarse modes and for the case of K^+, the fine mode is the dominant one (suggesting biomass combustion as a possible emission source). As for Cl^-, the mass size distributions were clearly bimodal. The nano-Moudi outdoors had different size distributions from the BLPI for Cl^-, Na^+, Ca^{2+} and Mg^{2+}. Only for K^+ the size distribution is similar. Mass size distributions of Cl^- and Na^+ may have been influenced by filter contamination. The Ca^{2+} peak detected at around 100 nm obtained by the nano-BLPI in outdoor air may possibly be ascribed also to filter contamination, although no specific data are available to support this interpretation. Similar peaks at 10 and 50 nm were observed indoors with the nano-Moudi and nano-BLPI which may suggest bounce, contamination or blank variability.

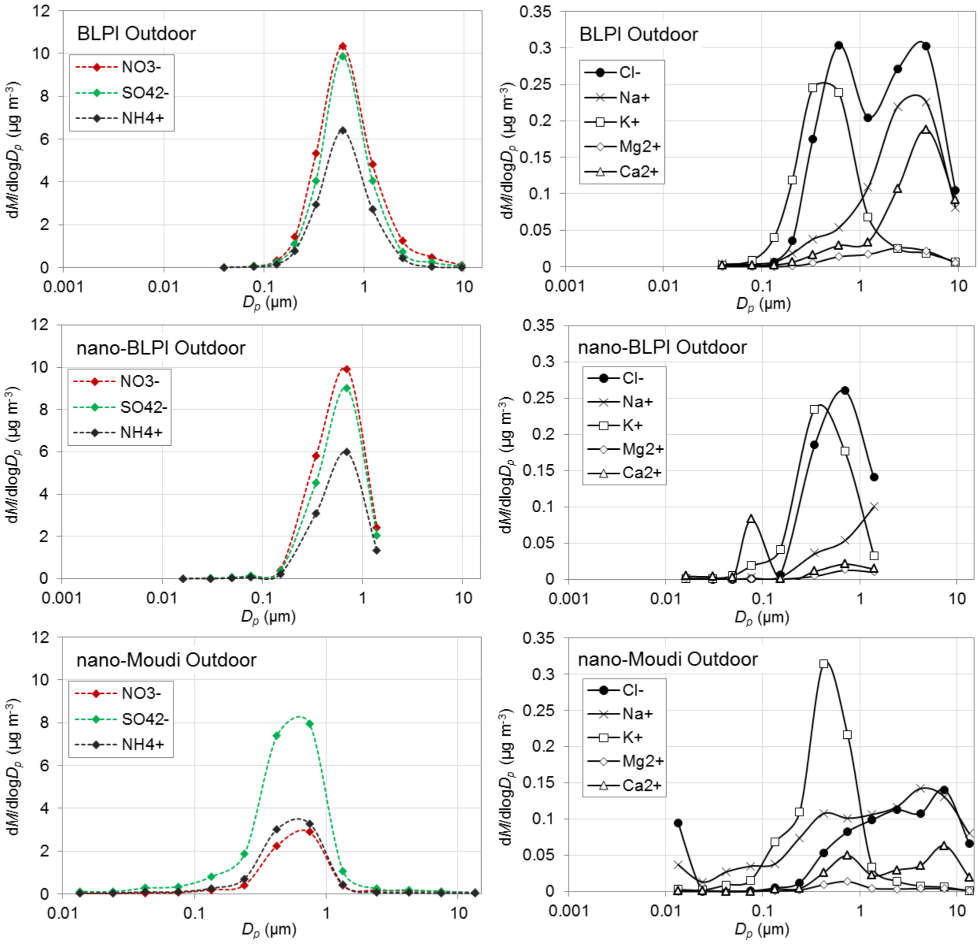

Figure 6. Average mass size distributions for different ionic species (left: SO_4^{2-}, NO_3^- and NH_4^+ and right: Cl^-, Na^+, K^+, Mg^{2+} and Ca^{2+}) during winter in outdoor air in Prague.

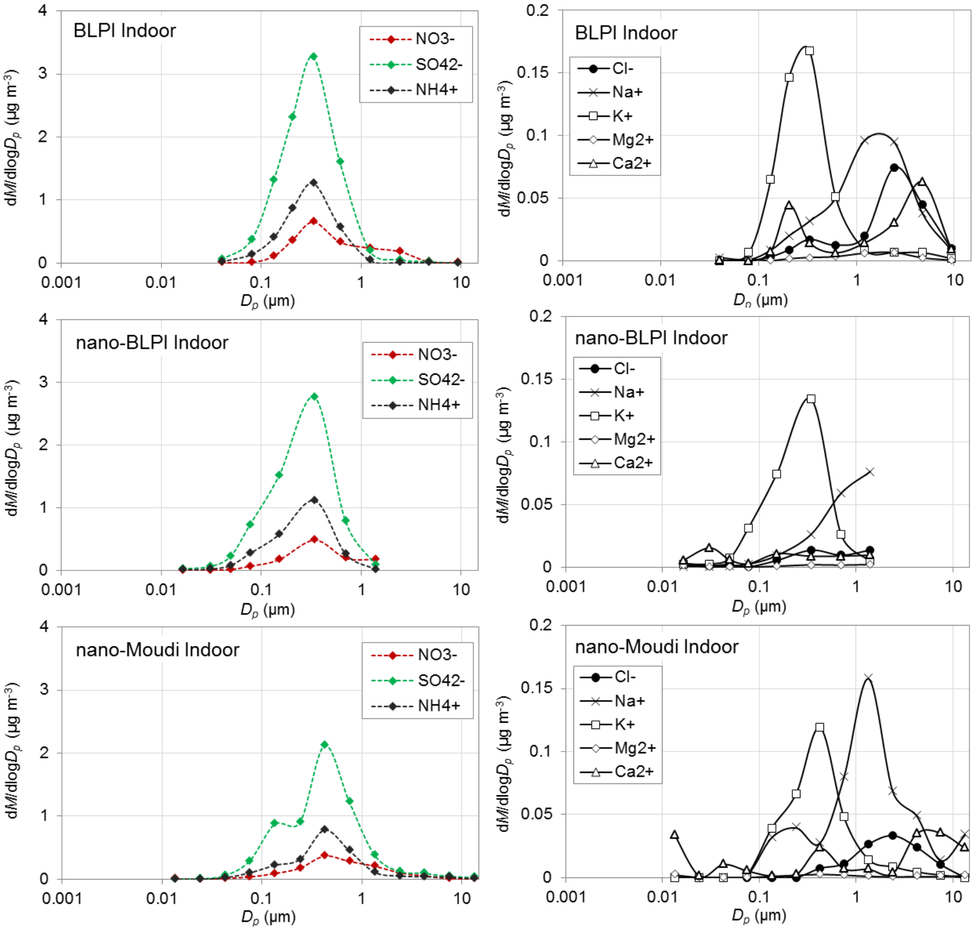

Figure 7. Average mass size distributions for different ionic species (left: SO_4^{2-}, NO_3^- and NH_4^+ and right: Cl^-, Na^+, K^+, Mg^{2+} and Ca^{2+}) during winter in indoor air in Prague.

4 Conclusions

This work aimed to assess the performance of four conventional and nano-range impactors, by means of two intercomparison exercises in Prague, during winter 2015 and in Barcelona during summer 2015. The aim of the campaigns was to test the instruments' performance with regard to the particle mass size distributions under different aerosol compositions resulting from different emission sources, meteorology, seasons, and air mass origins. All the cascade impactors were loaded with uncoated substrates excepting for the case of BLPI which foils were coated.

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Taking the BLPI as an internal reference, the best agreement regarding mass size distributions was obtained with the nano-BLPI, especially for particles larger than 250 nm. The nano-Moudi showed a good agreement for particle sizes >320 nm, whereas for particle diameters <320 nm this instrument recorded larger mass concentrations than the internal reference. Different particle effects may have caused the differences regarding particle mass concentrations collected in indoor and outdoor air by the nano-Moudi. Particle volatilisation may have occurred due to the internal rotating mechanisms which heat the impactor casing up. Decomposition of ammonium nitrate and chloride, as evidenced by the lower nitrate and chloride concentrations in the accumulation mode, is probably also enhanced in the nano-Moudi due to the spreading of the sample on the whole filter surface, in comparison with thick individual spots of material obtained with the BLPI and nano-BLPI impactors. Further research is needed to clarify this issue. With regard to the PCIS, their size-resolved mass concentrations were comparable with other impactors for PM_1, PM_2 and PM_{10}, but the cut-off at 0.25 μm was not consistent with that of the internal reference.

In Barcelona, the sampling took place under dry conditions (< 50% RH) and thus, particle bounce would be expected since some particles (depending on composition) could get dry. Inversely, bounce can be probably neglected for the Prague outdoor intercomparison since the RH was always >50 % indicating the presence of droplet aerosols that tend to adhere to the impaction substrate. To avoid such an effect impactor substrates should always be greased especially in areas with low humidity.

Aerosol mass size distributions were assessed for the Prague and Barcelona campaigns. During winter in Prague (outdoors), the mass size distributions showed a predominantly fine mode, with the coarse mode being almost negligible (by all impactors). However, in Barcelona, the coarse size fractions showed larger mass concentrations, evidencing the higher influence of mineral and marine aerosols.

This study concludes that comparability between the different types of impactors assessed was dependent on particle size. Specifically, the influence of the differences in impactor construction (number of jets, flow, vapour pressure, etc.) on UFP mass concentrations should be further addressed. In addition, further research is necessary with regard to the particle processes (evaporation, bounce, etc.) behind the differences in particle mass observed across size fractions in this study.

Acknowledgements

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