



Quantification of Low Pressure Impactor Wall Deposits during Zinc Nanoparticle Sampling

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ABSTRACT

A Dekati[®] low pressure impactor (DLPI) is used to determine the mass distribution of airborne particles from 7 nm to 10 µm as a function of aerodynamic diameter. Quantification of wall deposits inside this sampling device was performed for the first time using polydisperse zinc aerosol produced by a thermal spraying process. This aerosol is more representative of the size distribution found in occupational atmospheres than aerosols usually produced in laboratories. Each experiment (3 replicates) was carried out for two durations. The results showed that wall deposits were quite considerable and in relation with aerosol modes. 13% and 14% of the total mass was found on the wall after sampling periods of 5 and 20 minutes, respectively. Relative losses could reach 50% for certain impactation stages and these losses were also quite different between replicates.

Keywords: Dekati[®] low pressure impactor; Wall loss; Polydisperse aerosol; Zinc nanoparticles; Chemical analysis.

INTRODUCTION

Low pressure cascade impactors provide information on the size distribution of airborne particulate matter. They are widely used in various fields for characterizing aerosols, such as environmental dust and haze (e.g., Kang *et al.*, 2013), coals (e.g., Wang *et al.*, 2007; Kazanc *et al.*, 2013), urban aerosols (e.g., Maenhaut *et al.*, 2002; Salma *et al.*, 2002; Wagner *et al.*, 2008; Xie *et al.*, 2009), drugs (e.g., Klemmer *et al.*, 2013; McBride *et al.*, 2013; Sultana *et al.*, 2013; Zhu *et al.*, 2014), and bacterial aerosols (e.g., Simon *et al.*, 2013; Li 2013). In addition, impactors are commonly used for characterizing workplace environments (e.g., Cohen and Powers 2000; Chen *et al.*, 2007; Wu and Vincent, 2007; Dahlin *et al.*, 2008a; Dahlin *et al.*, 2008b; Dall'Osto *et al.*, 2008; Linnainmaa *et al.*, 2008; Birch *et al.*, 2011; Yang 2011; Coggins *et al.*, 2012; Ehrlich *et al.*, 2013). Most of them (MOUDI, ELPI, DLPI, BLPI, QCM impactor, etc.) are suited for nanoparticle sampling, which is a major advantage regarding the increasing presence of airborne nanoparticles, either produced intentionally or as a by-product of human activities in occupational environments. Many processes such as foundry casting (e.g., Cheng *et al.*, 2008), welding (e.g., Lehnert *et al.*, 2012; Zimmer *et al.*, 2002), and

thermal spraying (e.g., Bémer *et al.*, 2010) produce high concentrations of ultrafine particles. The main disadvantages of impactors are particle bouncing, multiple stage deposition (overlapping), and wall losses. The latter depend on many parameters such as impactor geometry, flow rate, particle size distribution and the nature of aerosol. For example, liquid particles are more subject to losses than solid particles (Marple *et al.*, 1991). Other parameters such as collection substrate (Fujitani *et al.*, 2006), relative humidity (Yamamoto *et al.*, 2002), and substrate greasing (Yamamoto *et al.*, 2002; Dunbar *et al.*, 2005) also affect wall losses by modifying stage collection efficiencies. Thus wall deposits must be characterized and quantified correctly to avoid the underestimation of particle mass concentration and mass size distribution.

Quantifying wall deposits is not systematic in impactor characterization. Most studies are limited to determining efficiency curves and few have focused on wall deposits which are generally considered negligible. When investigated, losses are essentially estimated from monodisperse (wet and/or dry) aerosols (Cheng and Yeh, 1979; Cushing *et al.*, 1979; Hering and Friedlander, 1979; Rubow *et al.*, 1987; Mitchell *et al.*, 1988; Vaughan, 1989; Marple *et al.*, 1991; Rader *et al.*, 1991; Marjamäki *et al.*, 1998; Michaud *et al.*, 1999; Kwon *et al.*, 2003; Demokritou *et al.*, 2004; Štefancová *et al.*, 2011; Liu *et al.*, 2013) which allow quantifying aerosol losses for a narrow range of particle size. Moreover, these laboratory studies provide data with model aerosols such as ammonium fluorescein (Cushing *et al.*, 1979; Rubow *et al.*, 1987; Marple *et al.*, 1991), which are not representative of

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the aerosols encountered in natural and occupational environments. Therefore there seems to be a lack of empirical data relating to other kinds of aerosols, and more specifically dry polydisperse aerosols for which computer modeling is particularly difficult compared to monodisperse ones. In particular, it would be helpful for users to study impactors with aerosols similar (in terms of concentration levels, number size distribution, chemical composition) to those expected to be sampled in workplace environments. This has already been suggested for multi-jet cascade impactors (Mitchell *et al.*, 1988). In addition, comparing polydisperse and monodisperse aerosols could highlight aggregation effects.

In this study, the wall deposits of a Dekati® Low-Pressure Impactor (DLPI) were quantified. This cascade impactor is composed of 13 impaction stages and a terminal filtration stage (optional) with a wide collection range (from 0.007 to 10 μm). Aerosols are sampled at a high flow rate (10 L/min) which allows the rapid collection of adequate quantities of airborne particulate matter for gravimetric and/or chemical analysis of impaction substrates. In addition, aerosol deposition on DLPI walls has not yet been subject to precise characterization, except by Marjamäki *et al.* (1998) who quantified losses of charged particles only.

Wall losses were determined for a dry aerosol produced by an electrical arc gun equipped with pure zinc wires that generate bimodal aerosols mainly composed of ultrafine particles (Bémer *et al.*, 2010). This setup was used under controlled laboratory conditions to mimic metal coating processes (electric arc, high velocity oxy-fuel, cold spray, or plasma). The choice of this source was justified by the wide use of thermal spraying processes and more generally thermal processes in various industrial sectors and thus the size of the population of workers potentially exposed.

METHODS

Aerosol Generation

The experimental set-up used for aerosol generation was described in Bémer *et al.* (2013), and is shown in Fig. 1. The electrical arc gun used is a Margarido® M25 supplied with zinc wires (2 mm in diameter). The operating principle of the electric arc gun is to apply a high intensity current (120 A; 21 V) between two wires to sublimate the metal. The metal particles are then propelled by compressed air at a pressure of 7 bar. The zinc wires are consumed at a rate of 2 m/min. The bench generates large quantities of polydisperse fumes composed of coarse (5 μm) and fine (0.08 μm) particles. This aerosol is adequate because it covers the entire size range of the cascade impactor investigated (0.007–10 μm on 13 stages, see below).

The gun is fixed to a vertically moving arm translated regularly along the rotating target (metal cylinder, $D = 40$ cm, $H = 40$ cm). The particles passing through the target (> 100 mg/m^3) are extracted via a 20 cm exhaust duct (approximate flow rate: 2000 m^3/h). This fully automated bench eliminates possible human errors and provides good stability and reproducibility in terms of particle mass size distribution. The collection point was located 5 m downstream to ensure aerosol stability. The aerosol was sampled isokinetically by metal elbow probes (internal diameter 16 mm) introduced into the duct. All the sampling devices were connected to the same collection point.

Sampling Devices

The Dekati® Low Pressure Impactor

The DLPI is a cascade impactor which collects airborne particles on 13 different impaction stages and one terminal filtration stage (the use of the filter stage is optional but

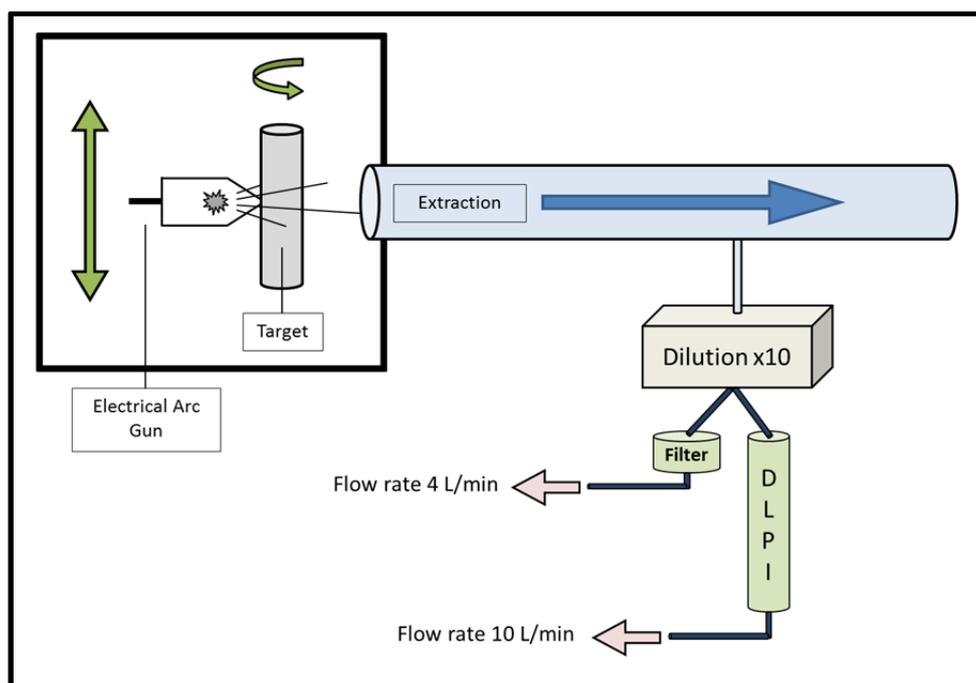


Fig. 1. Experimental set-up for aerosol generation and collection.

was employed here). Aerosol is collected from 0.007 to 10 μm according to aerodynamic diameter. 8 stages and the filter stage are dedicated to particles below 1 μm which gives quite good resolution for ultrafine particle distribution, and five stages collect particles from 1 to 10 μm . This device was used at a nominal flow rate of 10 L/min with a pressure drop of 100 millibar and ensured rapid particle collection. Different materials can be used as collection substrates, such as aluminum or polycarbonate foil, polyvinyl chloride (PVC) membrane, quartz fiber filter, etc., as a function of the environment and the information sought.

The DLPI collection substrates used in this study were 25 and 47 mm (filter stage) PVC membranes (GLA5000, PALL[®]). They were used because of their chemical purity and their simple preparation for analysis. Before use, they were washed with 0.1% Triton-X solution to remove electrostatic charges. After drying, the membranes were greased as recommended by Dekati[®] (DLPI user manual ver.3.62, 2010). We used Vaseline spray (Kontakt 701, Kontakt Chemie[®]) and a dedicated stencil was used to apply the grease over a defined area (15 mm disc centered on the membrane) to prevent the grease from entering under the substrate holders. The grease was applied to reduce the bouncing effect and was pure enough to perform chemical analysis without altering the performance of the spectrometer.

The membranes were weighed before and after sampling in order to assess aerosol total mass.

47 mm Filter Holder

A 47 mm filter holder made of stainless steel was used in parallel with the DLPI to provide a reference total mass concentration (without size separation) at a flow rate of 4.00 ± 0.03 L/min. These two filter holders were used in each experiment and were equipped with a 47 mm PVC membrane (GLA5000, PALL[®]). Unlike the 25 mm membranes, they were washed with 0.1% Triton-X solution but not greased.

Dilution

In order to avoid sampler saturation because of the high concentration of the aerosol in the main duct (> 100 mg/m^3), the aerosol was diluted. According to the manufacturer, it is advised not to exceed 1 mg of particulate matter per stage in order to ensure representative results (DLPI user manual ver.3.62, 2010). Thus, for the two experimental durations (5 and 20 minutes), a 10-fold dilutor (Palas[®] VKL 10) was used to accommodate aerosol concentration and distribution between the 13 stages according to impactor flow rate and sampling durations.

Conditioning

Before each experiment, the DLPI column was completely disassembled in order to clean each part in an ultrasonic bath with ultrapure water (MilliQ, Millipore[®]) for ten minutes. Every part was then rinsed and dried in an oven. Clean wipes wetted with isopropyl alcohol were then used to remove the potential remaining particles from the surfaces. This cleaning procedure was based on that described in the DLPI user manual (ver.3.62, 2010). Finally, the conditioned

(washed, greased and blank-weighed) membranes were placed on the impaction stages and the column was assembled.

Trials

Three trials were performed for both collection periods of 5 and 20 min. For each run, the electrical arc gun was turned on two minutes before starting the sampler to ensure aerosol stabilization in the duct. The DLPI and a filter holder were used for all the tests. These instruments were connected to one of the four outputs available on the flow splitter (TSI model 3708) adapted on the dilutor to provide equal flow distribution (Fig. 1).

Analysis

After collection, the membranes (from the DLPI, and filter holder) were placed in Petri dishes and stored overnight before gravimetric analysis with a 1 μg precision balance (Mettler[®] Toledo MX5).

After weighing, the PVC membranes were mineralized with 10 mL of bi-distilled nitric acid sealed in TFM containers in a microwave oven (Ethos ONE, Milestone[®]). The containers were heated at 230°C for 10 min before cooling. Once digested, the samples were diluted and analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES, Varian[®] 720-ES) or inductively coupled plasma mass spectrometry (ICP-MS, Varian[®] 820-MS) as a function of particle loading. Detection limits were 0.448 and 0.058 $\mu\text{g}/\text{sample}$ for ICP-OES and ICP-MS, respectively.

DLPI wall deposit characterization was performed for each impaction stage (1 to 13). The loss for one stage corresponds to the amount of zinc found on an impaction plate (without membrane) and a jet-plate (Fig. 2). Particulate matter on the inner surfaces was collected on a 37 mm mixed cellulose ester membrane (MCE, Millipore[®]). Two MCE membranes were used; one was dedicated to the impaction plate and the other to the jet-plate. Each MCE membrane was first cut into 4 equal parts in order to wipe each side of the plates and easily reach sharp parts. The membranes were wetted with ultrapure water and then were used to wipe the DLPI surfaces. The details for the wiped surfaces are presented in Fig. 2. The upper and lower surface of the jet-plate was wiped, especially the areas close to the nozzles where high amounts of particulate matter were noticeable (Fig. 3). After wiping, the four parts of the membranes were stored in clean tubes before mineralization. The MCE membranes were digested with 500 μL perchloric acid and 3 mL bi-distilled nitric acid before dilution and analysis. For each experiment, before collection and after surface sampling, several humidified 37 mm MCE membranes were used on the DLPI inner surface to check for the absence of residual contamination and to validate the efficiency of the wiping process.

RESULTS AND DISCUSSION

Filter Holder Gravimetric and Chemical Analyses

Total dust and zinc concentrations on the filter holder PVC membranes were measured for every sampling sequence.

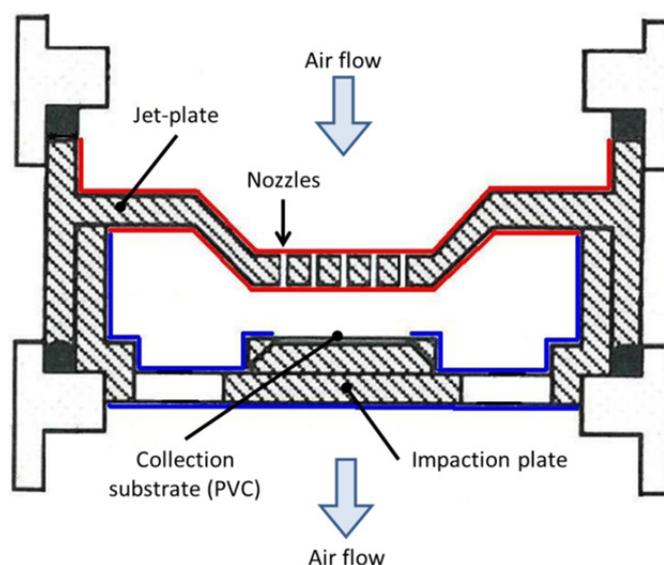


Fig. 2. Section of one DLPI stage to illustrate surfaces wiped for wall loss quantification. In red, surfaces wiped on the jet-plate, in blue on the impaction plate.



Fig. 3. Picture of jet-plate surface deposits on DLPI stage 4 ($D_{50\%} = 0.155 \mu\text{m}$).

Total dust concentration was quite stable for the different generations, with an average concentration of $12.3 \pm 1.0 \text{ mg/m}^3$ (8% RSD). Mean zinc concentration was $10.3 \pm 0.9 \text{ mg/m}^3$ (8.5% RSD). The difference between total dust and zinc concentrations was relatively high (16%). This difference could be explained by the fact that chemical analysis only reflects zinc mass while gravimetry gives the total mass of dust collected. Therefore, impurities, unexpected dust and zinc oxidation were not taken into account by chemical analysis.

Wall losses in the filter holder were not quantified or characterized; nevertheless it is reasonable to assume that losses occurred during sampling. Regarding the filter holder geometry, we assumed that losses would be low. Indeed, the surfaces involving particle losses in the filter holder were smaller than for the DLPI, while the air flow was straight in the sampler, thereby limiting contacts between the aerosol and the surface.

The results for the filter holder are compared directly with the DLPI gravimetric analysis presented hereafter.

DLPI Gravimetric and Chemical Analyses

Fig. 4 represents the average values ($N = 3$) for gravimetric and chemical analysis of PVC membranes for 5 and 20 min. Particle mass size distribution is approximately the same for the two durations. The main mode of the aerosol is centered on 80 nm, and the second around $5 \mu\text{m}$. Error bars correspond to one standard deviation and show relatively good reproducibility.

A slight difference can be seen between the two durations for stage 7, collecting particles from 0.64 to 0.987 μm . This must be an artifact on mass measurement regarding the good correlation ($r^2 = 0.97$) found for every sample between gravimetric and chemical analysis of the impaction media (Fig. 4). Deviations for gravimetry could be due to greater uncertainty from the balance, especially for the lowest loading on the collection medium. The average discrepancy between gravimetric and chemical analysis is 39%, with the larger relative discrepancies corresponding to the stages with the lowest particle loadings. For these stages, the difference is therefore less representative due to high uncertainties on gravimetric measurements.

Concerning the 20 min samples, the same observations could be made for the deviations on the replicates between the two kinds of analysis (Fig. 4). The mean difference between gravimetric and chemical analysis is 21% and could be mainly due to zinc oxidation. Indeed, this gap corresponds to the mass difference between ZnO and Zn. The conditions for aerosol generation were favorable for oxidation as sparks between the wires led to high temperature, and the compressed air supplied oxygen to the system. However, the zinc particles were probably not fully oxidized during this process as oxidation first occurs for surface atoms and would require more time (compared to particle generation duration) to reach the core of the particles (Delalu *et al.*, 2000). Although zinc oxidation kinetics has been studied for bulk material and coarse particles, data are lacking for nanoparticles. Nevertheless, the smaller the

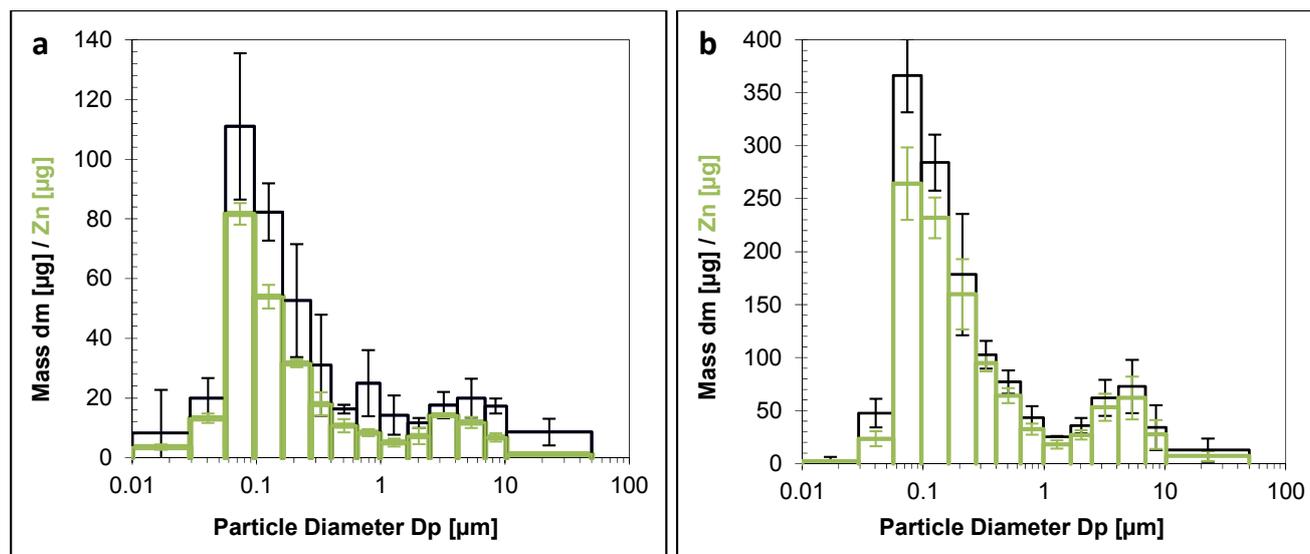


Fig. 4. Chemical and gravimetric results for DLPI impactation stages as a function of particle aerodynamic diameter. (a) 5 min and (b) 20 min sampling duration.

particle, the higher the proportion of surface atoms (Preining, 1998). Thus, oxidized surfaces could be larger for ultrafine particles than for micronic ones.

No final conclusions could be made on the origin of the differences between the two kinds of analysis. None of the hypotheses listed could be excluded. A significant part of the gap could be attributed to zinc oxidation, which appears to be consistent with the results (specifically for fine particles).

The average total masses collected on the substrates were 0.43 and 1.34 mg for 5 and 20 min, respectively. The mass ratio between the two durations was 3.11, slightly below the theoretical ratio of 4. Thus conformity of total mass proportionality on the collection medium was not fully achieved.

In addition, the quantity of zinc in the entire column given by chemical analysis was proportional to sampling duration ($Zn_{20}/Zn_5 = 3.99$). This ratio was 4.04 ± 1.52 on average for the calculation of individual stages. The results obtained with chemistry therefore appeared more reliable than the results of gravimetric analysis, regarding mass proportionality as a function of sampling duration. In the remainder of this paper, only the results from the chemical analysis will be reported.

Mean zinc concentrations for the filter holder and DLPI were compared for the two sampling durations and the results are presented in Table 1.

Zn concentration from the DLPI analysis was systematically lower than that found on the filter holder, with an average relative difference of 44%.

The differences between the two samplers could be due to particle losses along the transfer tubing, because the tubing connected to the DLPI was 6 times longer than that connected to the filter holder. Von Der Weiden *et al.* (2009) developed the Particle Loss Calculator (PLC), a software application which allows estimating losses inside transport tubing, using several physical mechanisms such as diffusion, settling, and inertial deposition. We ran PLC under our

Table 1. Results of zinc quantification on the filter holder and DLPI for 5 and 20 minute samples. ND for Non-Determined.

	Experiment #	Filter Holder	DLPI
		mg(Zn)/m ³	
5 min	1	10.5	5.6
	2	11.6	6.0
	3	9.4	ND
Average		10.5	5.8
20 min	1	11.1	5.9
	2	10.0	5.3
	3	9.5	6.0
Average		10.2	5.7

settings, adding all the effects proposed and the results show very low losses for both short (≈ 10 cm) and long (≈ 60 cm) tubing. Indeed, total loss during transport was estimated below 1% for particles larger than 20 nm and from 1 to 2% for particles smaller than 20 nm. Thus we could consider particle loss in tubing as negligible.

Considering the mass sampled by the DLPI substrates, it was lower by 44% than the total mass sampled by the filter holder substrate. Thus DLPI wall losses were investigated.

DLPI Wall Losses

It is noteworthy that the main loss phenomena were due to particles rebounding on the substrate, the diffusion of fine particles, inertial deposition, and charge effects. One effect predominates according to the characteristics of the particles, particularly size. For example, large particles are more subject to inertial deposition than diffusion. On the contrary, fine and ultrafine particles are mainly subject to diffusion. It should be noted beforehand that the amounts of particles collected (0.4 mg maximum) were well below the maximum recommended by the manufacturer (1 mg maximum per stage). Consequently, losses could not be attributed to overloading, especially in the upper stages. In

the lower stages, this limit was less relevant because 1 mg of ultrafine particles represents a larger volume than for coarse ones. The amounts of particles may slightly modify aerolic characteristics and thus wall losses.

The results showed that generally, for one stage, 80% of loss was found concentrated around the jet-plate nozzles (Fig. 3) and 20% on the impaction plate. For stages 12 and 13 ($> 8 \mu\text{m}$), ratios were closer to 50–50%. This was higher compared to previous results (Cushing *et al.*, 1979) where nozzle wall loss represented about 40% of the total wall loss on different impactors, based on the generation of monodisperse particles of ammonium fluorescein (diameters from 1 to $18 \mu\text{m}$). Michaud *et al.* (1999) also noticed evidence of deposition around orifices and on substrate retaining rings. Liu *et al.* (2013) developed smoother shaped nozzles that avoid clogging effects and thus slightly reduce losses for particles smaller than 105 nm (from 2.9 to 26.1% for an impactor (MOUDI) with non-modified nozzles, and from 3 to 23.5% for an impactor (NCTU micro-orifice cascade impactor) with modified nozzles). Moreover, Vaseline greasing could be replaced by silicone to enhance particle retention (Pak *et al.*, 1992). Nevertheless, Marple *et al.* (1991) used silicone on substrates and found up to 35% loss for liquid particles ranging from 0.03 to $20 \mu\text{m}$, and up to 20% loss for solid particles ranging from 7 to $20 \mu\text{m}$. Dunbar *et al.* (2005) concluded that substrate greasing reduces but does not minimize bounce effects and thus related particle loss. Therefore it seems difficult to avoid particle loss even under the best condition of use.

The results presented in Fig. 5 show that a non-negligible amount of particles was found on the walls (e.g., for stage 13 during the 5 min. generation period, average particle loss was 48%). Particle deposition seems to be correlated with aerosol modes. The maximum wall deposit is located around stages 3 and 4, which correspond to the collection of particles 150 nm in diameter.

If the size of the particles deposited on the walls cannot be strictly attributed to one of the corresponding stages we can presume these particles are rather small, considering

that larger particles should have deposited on the upper section of the impactor. The other significant deposition area was located around stages 12 and 13 (particles collected of $10 \mu\text{m}$ on average). They could be either large or small particles; however, Mitchell *et al.* (1988) noticed that particles larger than $5 \mu\text{m}$ were responsible for increased wall losses in stages 0, 1 and 2 of the Andersen Mark-II, with effective cut-off diameters of 9, 5.6 and $5 \mu\text{m}$, respectively.

These losses occurred before the most impacted stages, as already observed for other impactors and sampling conditions (Kwon *et al.*, 2003). According to the authors, the shift between aerosol mode and maximum wall deposit can be explained by the fact that a particle of a given size will mainly settle on its “dedicated” impaction plate. However, as efficiency curves are not perfect, particles of given sizes can impact on several stages (overlapping). Thus some of the particles will deposit on the previous stage and probably also on the walls. This phenomenon has already been observed in the Andersen Mark-II impactor (Mitchell *et al.*, 1988).

The amount of particles found on the walls was relatively repeatable. Total zinc losses were almost the same between samples (20% and 11% RSD for 5 and 20 min sampling durations, respectively), but were more scattered when considered stage by stage.

These losses represent a non-negligible amount of particles that are not collected on the impaction substrates. This results in underestimating particle mass concentration and, depending on the aerosol considered, in inaccurate mass distribution. Collection conditions such as substrate and substrate holder could modify particle behavior (e.g., bouncing effect) and thus alter aerosol distribution. Cheng and Yeh (1979) concluded that particle rebound reduced collection efficiency which increased wall losses and shifted aerosol distribution in the Sierra radial slit jet impactor. Dunbar *et al.* (2005) observed statistically significant ($P < 0.05$) differences in mass median aerodynamic diameter between uncoated and coated impaction plates; they also minimized bounce effects with the use of saturated glass

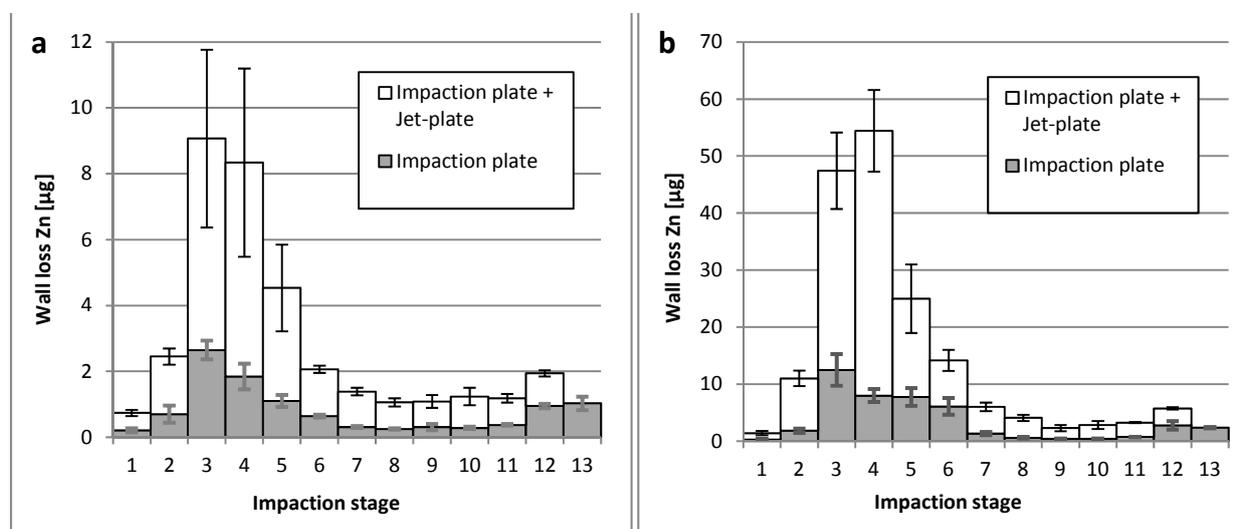


Fig. 5. Wall deposits for (a) 5 min ($N = 2$) and (b) 20 min ($N = 3$) generations as a function of impaction stage.

fiber filters. However, they considered interstage losses to be intrinsic to the impactor despite the link between rebound and loss. Fujitani *et al.* (2006) showed that the modal diameter of a DLPI equipped with aluminum foils or polycarbonate filters was shifted two stages downward compared to the same impactor equipped with quartz fiber filters.

Fig. 6 represents the relative amounts of zinc found on walls. No difference in the relative amount of zinc was found between sampling durations of 5 and 20 min. Hence relative wall deposits do not depend on sampling duration and thus on the amount of zinc provided to the system. Deviations on replicates were 15% and 17% for the 5 and 20 min samples, respectively, thus non-negligible. However, very low particle loading on some stages causes very large deviations that may not be representative of the effective deposit on those stages. It should be noted that inner surface zinc quantification before and after each experiment showed low levels of zinc (from 1 to 6 μg for the whole column), which confirms the efficiency of the protocol and the robustness of the results relative to quantities deposited on the wall.

Three main deposition areas could be distinguished from the general trend. The first deposition area was located around the fourth and fifth plates, probably resulting from the diffusion of the smallest particles because coarse particles had been collected earlier and were theoretically absent in this section of the impactor (unless the fine particles had aggregated). The second deposition area was located around the eighth stage where both small and large ($\approx 1.5 \mu\text{m}$) particles could contribute to wall deposits. Finally, considerable relative wall deposits were observed for the last two stages (12 and 13). Nevertheless, in this case, relative wall deposits may not be representative regarding the very low loading on the collection medium, leading to high relative ratios.

The mean loss by wall deposit for the whole DLPI column represented 13% (9% RSD) and 14% (6% RSD) of the total mass collected for the sampling durations of 5 and 20 min,

respectively. Thus, total wall losses were reproducible for the parameters investigated. More specifically, the deposition rates on the walls could reach 48% and 35% for the durations of 5 and 20 min, respectively, but were less reproducible (15% and 34% RSD for the 5 and 20 min sampling durations, respectively) from one stage to another compared to total particle losses.

For most impactors (Andersen Mark I, II, III, SIERRA model 226, MOUDI, BLPI, Marple), wall losses tend to decrease when particle size decreases from tens of micrometers down to 1 μm (Cushing *et al.*, 1979; Vaughan, 1989; Rader *et al.*, 1991). On the contrary, Horton *et al.* (1992) found that losses increase with particle size reduction. The very low flow rate (0.24 L/min) of the impactor investigated by Horton *et al.* (1992) may be responsible for this unusual behavior compared to other studies. Losses seem to be lowest between 0.3 and 1 μm regardless of aerosol, and start to increase again for particles below 0.3 μm (Marjamäki *et al.*, 1998; Demokritou *et al.*, 2004; Štefancová *et al.*, 2011; Liu *et al.*, 2013). The latter behavior has been observed for every impactor investigated, except for the QCM impactor studied by Horton *et al.* (1992). The results on the MOUDI obtained by Marple *et al.* (1991) showed that losses reached 35% for liquid particles, 20% for solid particles of about 15 μm , and were below 2% for particles between 0.2 and 3 μm . Losses increased again for particles below 0.1 μm .

Our results on the DLPI were slightly different. Indeed, there were high wall losses in the upper stages (12–13) and also in the lower stages (3–5), but losses were non negligible between these deposition modes, (17 and 18% on stage 8 for samples taken over 5 and 20 min, respectively), as shown by other authors. The most noticeable singularity was the decrease of losses below stage 4 which has not been shown yet.

The deposits found around the nozzles were probably due to the diffusion of fine and ultrafine particles. Even in the upper section of the impactor, diffusion was probably

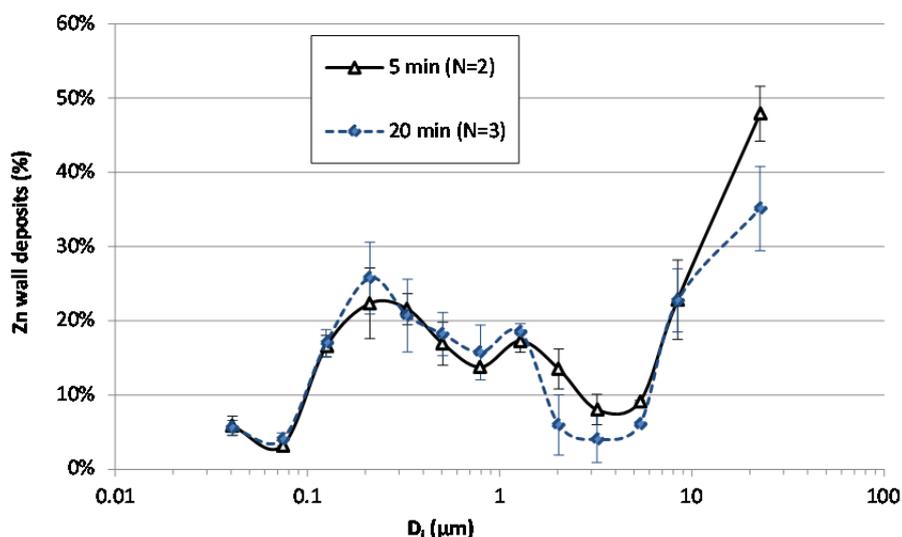


Fig. 6. Relative amount of wall losses for 5 min (solid line) and 20 min (dashed line) samples.

involved in wall losses. Marjamäki *et al.*, (2005) showed that low amounts of ultrafine particles ($<0.1\ \mu\text{m}$) were collected (up to 5% for 10 nm particles) in the upper stages of the ELPI. For the upper stages, the size of the particles found on the walls could be highly variable since low amounts of ultrafine particles ($<0.1\ \mu\text{m}$) were collected (up to 5% for 10 nm particles) in all the stages (Marjamäki *et al.*, 2005). Furthermore, the number and the size of the nozzles varied per stage. The maximum particle deposition was found on stages 3 and 4, equipped with a large number of narrow nozzles (21 and 19 respectively). The air flow was faster at this level of the impactor than at its top. This could lead to greater turbulence and, consequently, to a higher probability for particles to deposit on the walls. It is consistent with the upper parts of the DLPI where wall losses were relatively high and where the nozzles were few and wide. In this case, the diffusion, impaction and interception of particles were all probably involved in wall deposition.

The use of the DLPI is therefore interesting and gives relevant information, but it should be used in synergy with another sampling device to obtain additional information on particle size distribution, and wall losses should be quantified systematically for each sample or aerosol source. Solutions like the use of greased substrates will clearly decrease wall losses and thus improve particle collection efficiency (Vaughan, 1989; Michaud *et al.*, 1999). However, greasing is certainly not sufficient in view of our results. More generally, sampling conditions (humidity, substrate greasing, temperature, characteristics of aerosol sampled, etc.) can lead to specific wall losses that are difficult to predict and could result in inaccurate assessments of mass size distribution.

CONCLUSION

DLPI wall losses were quantified with a bimodal metallic aerosol reflecting atmospheres that could be encountered in occupational environments where thermal processes are used. This kind of aerosol is comparable to those produced by welding and smelting, and was chosen to investigate potential synergetic effects between small and large particles. The results showed that losses were independent of collection duration and slightly different between replicates. Relatively high amounts of particles were found on the walls and especially close to jet-plate nozzles situated before the most impacted stages. Moreover, these deposits were found to be associated with the aerosol mode. This is an important result which justifies the use of such multimodal aerosols that do not appear to present interactions between the two size modes. Mean losses (13% and 14%) agreed with those found in the open literature for this kind of impactor. These losses remain relatively high considering that greased substrates were used and that the DLPI is an impactor designed to ensure low inter-stage losses. Although cascade impactors (including the DLPI) are effective tools for both qualification and quantification, users should bear in mind that non-negligible and inevitable losses slightly modify these two types of information. Apart from systematic quantification of wall losses, it appears quite difficult to

propose data correction without performing prior laboratory characterization of the impactor under well-defined conditions in the laboratory.

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