Development and characterization of a sequential aerosol sampler by cascade impaction: application to the study of a fine particle pollution episode

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1. Context and objectives

During particulate matter (PM) pollution events, the relative influence of local vs. distant sources, coupled with variable meteorological conditions, could lead to very rapid variations of pollutant levels (Phung Ngoc et al., 2021). This is especially true in multi-influenced environments, as coastal and highly industrialized urban areas (Zhang et al., 2021). Aerosol Mass Spectrometers (AMS) and Aerosol Chemical Speciation Monitor (ACSM) are well suited for this, when dealing with long term measurements at a specific location. But increasing the spatio-temporal coverage of the monitoring requires more measurement sites and therefore the implementation of less expensive instruments that are quickly and easily operational. When coupled with individual particle analysis techniques such as SEM/EDX, cascade impactors appear particularly well suited. They can indeed allow short sampling, providing information on the size, morphology and chemical composition of the particles. For that, it seems relevant to have a system where the particle collection substrate can be changed very quickly, to perform a succession of time-limited samplings consistent with the evolution of the meteorological conditions and the dynamics of the boundary layer. The best solution is to use a system that automatically changes the particle collection surface, such as a rotary plate impactor.

In this context, we developed a home made Time Resolved Atmospheric Particle Sampler (TRAPS), designed for the collection of fine (PM1) and ultrafine (PM 0.1) particles. Samples can be collected on commercial membranes, as polycarbonate membrane or/and simultaneously on TEM grids. Combined with adaptable sampling times, this allows the collection of samples suitable for a wide range of single particle analyses including SEM, TEM, Raman etc. The TRAPS will also help, in some extent, to limit the sampling artefacts for semi-volatile species, occurring during long sampling times, thanks to the membrane rotation that protects deposited particles from the continuous air flow.

We firstly present here the description of the TRAPS and an overview of the methodology and experimental setup used for the determination of its sizing characteristics. Secondly, an example of the use of this instrument during the study of a fine particle (PM2.5) pollution episode on a multi-influenced urban site is presented.

2. Theoretical calculations and TRAPS Design

The flow regime during impaction is defined by the Reynolds number (**Re** - eq. 1) of the fluid (dimensionless), which represents the ratio between the inertial forces (due to the mass of gases present in the air) and the viscosity forces that characterize the resistance to the air flow.

For an impaction plate, **Re** is defined by:

$$Re = \frac{\rho \cdot U \cdot d}{\eta} \qquad \textit{(Equation 1)}$$

Where **ρ** is the density of the air (1.205 kg.m^{-3} at 293K), **U** is the air velocity inside the impaction nozzle (m.s^{-1}), **d** is the impaction nozzle "equivalent diameter" and **η** is the dynamic viscosity of the air (1.806.10^{-5} Pa.s at 293K). All the values used for Reynolds number calculations are reported in Table 1. In order, for the impaction traces, to be distinctly separated on the collection surface, we opted for rectangular acceleration nozzles, without any impact on the Reynolds number values.

Using eq. 1, Re values of 3540 and 12420 are obtained respectively for the first (PM 1) and second (PM 0.1) impaction stages. With Re > 3000, the flow regime inside the TRAPS has to be considered turbulent. In the case of turbulent flows, (Pui et al., 1987) empirically demonstrated that the transport efficiency **η** of a particle following a curvilinear motion making an angle φ=π/2 rad (90°) with the principal direction of the flow can be calculated following eq.2 :

$$\eta = 10^{-0.963\,Stk} \qquad \textit{(Equation 2)}$$

Where ***Stk*** is the Stokes number, which represents the ratio between the kinetic energy of a particle suspended in a fluid (here the ambient air) and the energy dissipated by friction with the fluid. If the particle kinetic energy is very high (***Stk*** >> 1), it will easily leaves the flow lines while approaching the collection collection surface (inertial regime). In the other hand, if the particle kinetic energy is dissipated by friction (***Stk*** << 1), the particle will then follow the flow lines regardless of the fluid direction (viscous regime). ***Stk*** is calculated by eq. 3 :

$$\textbf{\textit{Stk}} = \frac{U \cdot \rho_p \cdot C}{9\eta} \cdot \frac{dp^2}{d} \qquad \textit{(Equation 3)}$$

Where **ρ$_p$** is the density of the particle, **U** is the air velocity inside the impaction nozzle (m.s^{-1}), **C** is the Cunningham slip factor (dimensionless and depends on particle size and pressure - Marple and Willeke, 1976), **η** is the dynamic viscosity of the air (1.806.10^{-5} Pa.s), **d** is the equivalent diameter of the impaction nozzle (m) and **d$_p$** is the aerodynamic diameter of the particle (m). All the values used for Stokes number calculations are reported in Table 1.

The central point in the design of the TRAPS is the determination of the diameters of the acceleration nozzles, allowing to fix the median size ("Cut-Off" diameter) of the particles collected on a given impaction stage. As the TRAPS is specifically designed to study pollution aerosols, we have chosen median sizes of 1 µm for the upper impaction stage (called "PM1 stage") and 0.1 µm for the lower stage (called "PM0.1 stage"), as the majority of pollution aerosols are submicronic.

In the field of cascade impaction, the median size of the collected particles is defined as the aerodynamic diameter of collection at 50% efficiency (Dp$_{50}$), i.e. the equivalent diameter of a sphere of given density, having the same final impaction velocity as the studied particles for 50% efficiency. Since calibrated silica particles (ρ$_p$=1.80 g.cm^{-3}) are available in the laboratory, we used them to experimentally determine Dp$_{50}$ and compare them to theoretical values reported in Table 1.

	PM1 Stage	PM0.1 Stage
Nozzle Equivalent Diameter (mm)	1.940	0.564
Cut-off diameter (µm) for ρ$_p$=1.80 g.cm^{-3}	1.30	0.15
Air flow rate (L.m^{-1})	5.00	5.00
Air velocity inside the impaction nozzle (m.s^{-1})	28	333
Jet to plate distance ratio (S/W)	2.56	5.32
Nozzle Reynolds Number (Re)	3450	12420
Slip Correction factor	1.12	2.25
Stokes number (Stk)	0.30	0.31

Table 1: *Theoretical calculations of the stage parameters of the TRAPS at T=293K and P= 101.3 kPa*

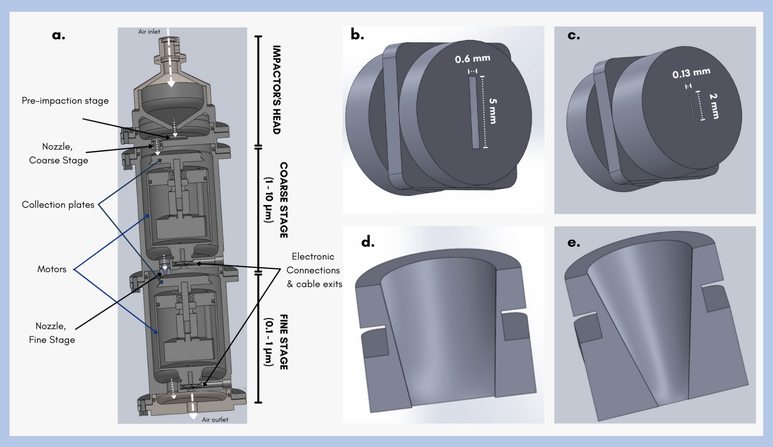

Figure 1: Internal view of the TRAPS impactor (a.) and scheme of the acceleration nozzles with their dimensions for the coarse-PM1 (b. and d.) and fine-PM0.1 (c. and e.) stages

3. Experimental determination of the Cut-Off diameters

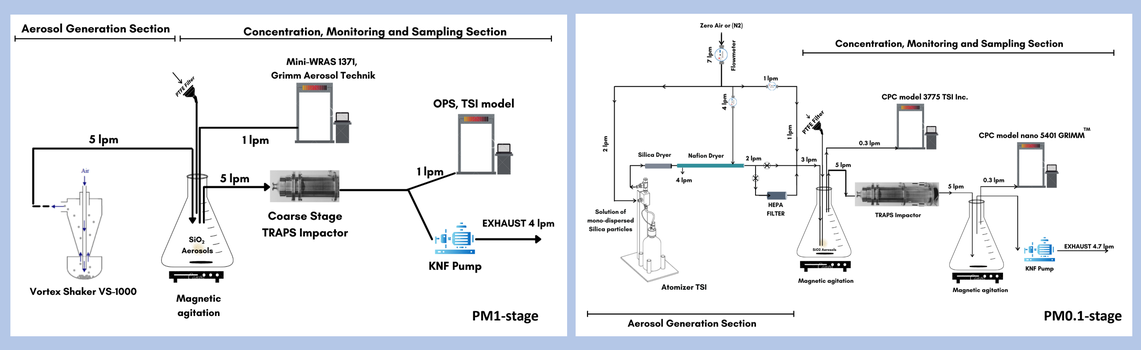

Figure 2: Schematic view of the experimental setup for determining collection efficiencies. On the left, the coarse- stage (1-10µm) using vortex shaking of powders of monodisperse silica spheres. On the right, the fine-stage (0.1-1µm)using nebulized monodisperse silica spheres

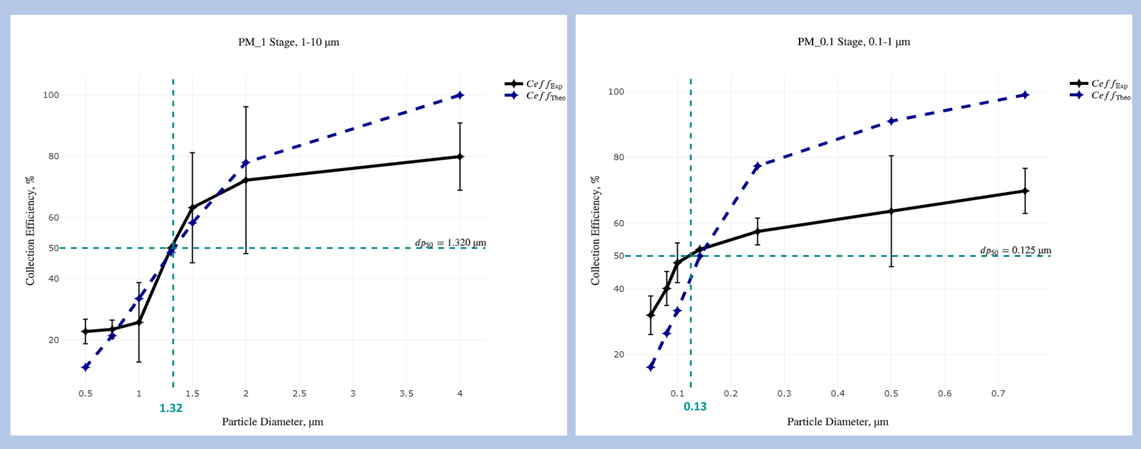

Figure 3: Theoretical (blue dotted line) and experimental (black line) collection efficiency curves on the left for the PM1 stage and on the right for the PM0.1 stage.

The Dp50 values of 1.32 µm and 0.13µm were graphically determined from the experimental curves and coincided with the theoretical value of 1.30µm and 0.15µm for the coarse and fine stages, respectively.

4. Case Study and Implications

The TRAPS was used during a PM$_{2.5}$ pollution episode from April 20th to 21st 2021 at Dunkerque, France (51°03'N, 2°38'E ; 200 000 inhabitants). Samples were collected for eight periods (P1-P8)during the episode (Figure 4) and two of them are of particular interest: **P1** and **P5** characterised by PM$_{2.5}$ mass concentrations ≥ 15 µg/m^3 (WHO daily limit). Results obtained for these two periods are presented here.

On each stage, particles were collected on TEM grids and analysed by SEM/EDX (Beji et al., 2020) allowing to identify six types of particles according to the main chemical elements present: Metals-, Carbon-, Sulfur-, Calcium-, Sodium- and (Si+Al)-rich particles (Figure 5).

During the period(**P1**), metals-bearing particles dominate the coarse fraction, which is consistent with the fact that the studied site was influenced by metallurgical industrial activities during this period. The identified particles are mainly Fe and Mn oxides of circular or nearly circular shapes, mixed with small amounts of others elements (Si, Al, K, Ca, Na). Carbonaceous and S-rich particles from local industrial sources are also abundant in the fine fraction.

For the second pollution event (**P5**), meteorological conditions had evolved and the studied site was no longer under the influence of local industrial emissions, but was impacted by regional air masses. Carbonaceous particles account for nearly 50% of all analysed particles, and these particles mostly come from the incomplete combustion of fossil fuels (car traffic) or from biomass burning. In addition, their particular abundance in the finest fraction of the atmospheric aerosols is consistent with long-range, non-local transport.

Through this case study, still exploratory, we were able to demonstrate that TRAPS, associated with the SEM/EDX technique well skilled in the LPCA laboratory, is well adapted for the description of temporal evolution of aerosols during pollution events.

This work is ongoing with the quantification of the mixing state of atmospheric particles, which should make it possible to assess the degree of ageing of particles during transport.

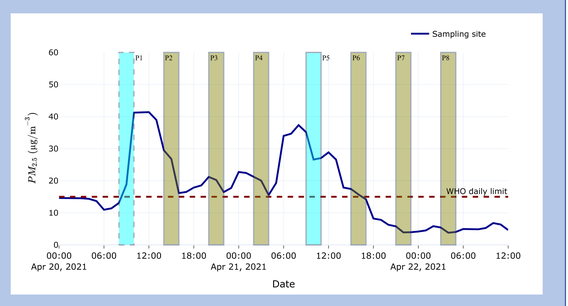

Figure 4: Time series evolution of PM2.5 mass concentrations at our sampling site. Colored rectangles represent TRAPS sampling periods.

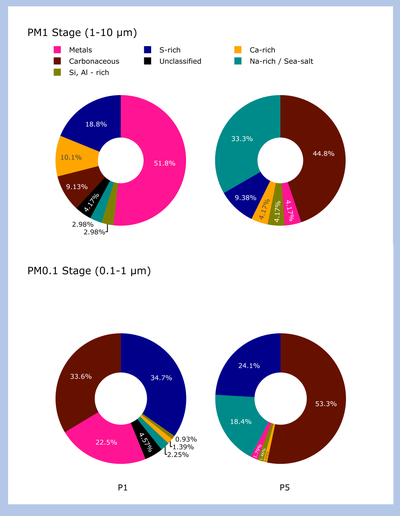

Figure 5: Relative contribution of different particle types during the P1 and P5 periods, for the PM1 and PM0.1 stages.

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