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Indoor dispersion of airborne nano and fine particles: main factors affecting spatial and temporal distribution in the frame of exposure modelling

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Abstract

A particle exposure experiment inside a large climate-controlled chamber was conducted. Data on spatial and temporal distribution of nanoscale and fine aerosols in the range of mobility diameters 8 – 600 nm were collected with high resolution, for sodium chloride, fluorescein sodium and silica particles. Exposure scenarios studied, included constant and intermittent source emissions, different aggregation conditions, high (10 h^{-1}) and low (3.5 h^{-1}) air exchange rates (AERs) corresponding to chamber Reynolds number respectively equal to $1 \cdot 10^5$ and $3 \cdot 10^4$. Results are presented and analysed to highlight the main determinants of exposure and to determine if the assumptions underlying two-box models hold under various scenarios. The main determinants of exposure found were the source generation rate and the ventilation rate. The effect of particles nature was indiscernible and the decrease of airborne total number concentrations attributable to surface deposition was estimated lower than 2% when the source was active. A near-field/far-field structure of aerosol concentration was always observed for the $\text{AER}=10 \text{ h}^{-1}$ but for $\text{AER}=3.5 \text{ h}^{-1}$ a single field structure was found. The particle size distribution was always homogeneous in space but a general shift of particles diameter (-8% to

+16%) was observed between scenarios in correlation with the AER and with the source position, presumably largely attributable to aggregation.

Key words

aerosol, nanoparticles, exposure determinants, two-box model, dispersion, model validation

1 Introduction

The consequences on human health of exposure to airborne fine particles ($<2.5\ \mu\text{m}$) have been a subject of concern for decades, leading notably to the US air quality standards for fine particles in 1987¹, and the 90's continued providing evidences of adverse effects on health of ultrafine ($<100\ \text{nm}$) particulate matter exposure²⁻⁴. With the emergence of nanotechnology, these concerns now extend to airborne fine and ultrafine particles of engineered nanomaterials⁵ (ENMs).

The tools currently available for the risk assessment of ENMs and their aerosols are of qualitative or semi-quantitative nature only, e.g. Stoffenmanager Nano, Nanosafer, CB Nanotool, Swiss Precautionary Matrix. Rather than quantitative air concentration predictions, they usually output a category of exposure potential, and, if hazard information is provided, a risk class. On the other hand, quantitative aerosol dispersion models developed range from complex models aiming to describe the concentrations in space and time with high resolution, e.g. Computational Fluid

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Dynamics (CFD) models⁶, to simpler models of reduced spatial and temporal resolution. Examples of the latter are so-called two-box models that assume the space to be dividable into two box-shaped volumes: a near-field (NF) box either around the person or the source of emission and a box for the far-field (FF, i.e. the rest of the room volume). The use of two-box models requires the contaminants to rapidly mix throughout the NF and FF in order to justify the assumption of homogenous concentrations in both boxes. However, there exists a general lack of knowledge on whether such simple dispersion models are applicable to fine/ultrafine aerosols despite the different behaviour observed for nano-sized and micro-sized particles. Once airborne, the dispersion of particles will depend on their size, surface area and charge distributions, as well as aggregation processes with background particles⁷. It is unclear to which extent these variables actually influence the NF and FF exposure estimates or how different the dispersion of nano-sized particles is compared to that of micro-sized particles, fumes or gases. Addressing these questions could help deciding if specific exposure models and mitigation strategies are needed for fine to ultrafine aerosol particles, including if they are composed of ENMs.

Small-scale experiments and field studies have been carried out to test the two-box models during scenarios involving the use of solvents^{8–14}. Overall, evaluations of the NF-FF model have found predicted values to be within 0.49–2.5 times measured concentrations in 99% of situations reported in the literature^{9,13,14}. The use of the two-box models in scenarios involving nano-size aerosols was tested by Koivisto *et al.*¹⁵ to predict exposure levels in a paint factory during pouring process. In that case,

the NF-FF model predicted values to be within 0.2–2.4 times measured concentrations. These studies provided initial data on the applicability and the general validity of the exposure models. However, there is a need to investigate the effect of changes in exposure determinants such as particle type (including ENM), emission pattern and ventilation conditions. Only experimental studies using exposure chambers with sufficiently large dimensions will be able to test the validity of assumptions on the spatial distribution, as well as the impact of changes in the scenario setting under controlled conditions.

Within the context of the EC FP7 project NANoREG (*A common European approach to the regulatory testing of Manufactured Nanomaterials*) we conducted a series of experiments to evaluate the extent of influence of the temporal emission profile (continuous, intermittent), the ventilation rate and emitted substance (including one ENM) on the temporal and spatial variability of airborne nanoparticles properties, for particles mobility diameters ranging between 8 nm and 600 nm.

This paper presents the experimental setup used in this project and the obtained results, i.e. time and space resolved number concentration and particle size distributions (PSDs) in the experimental chamber for the different scenarios. As a first analysis of the results, we propose to investigate the appropriateness of assumptions of spatially homogeneous exposure within the NF and FF boxes defined in two-box exposure models. Whenever possible, results are compared to

the one-box, two-box and three box modelling performed by Jensen *et al.*¹⁶ for the same experimental setup.

Acronyms used in the text are summarized in Table 3.

2 Materials and Methods

2.1 Aerosol chamber and air flows

The experiments were performed in an environmentally controlled chamber ($V = L \times W \times H = 6.45 \times 4.72 \times 2.65 \text{ m} = 81 \text{ m}^3$) at the Department of Civil Engineering at Danish Technical University. The chamber allowed full control of inlet and outlet airflows, temperature and humidity.

Filtered air using HEPA H14 filters was mechanically supplied to the room by two PKA 315 diffusers (Lindab A/S ventilation, Haderslev, Denmark) mounted as three-way supplies, located as shown in figure 1 (Technical drawings of the experimental setup are provided in figure i and ii in supplementary material). Air was mechanically exhausted through a single exhaust located in-between the inlets, also equipped with a PKA 315 diffuser in omnidirectional configuration. All supply and exhaust ports were computer-controlled individually by the ventilation system. Flow-rate calibration for each ventilation system was done using a balometer (SwemaFlow 125, Swema A.B., Farsta, Sweden). The total air supply Q was distributed to both inlets in equal shares. Three different ventilation configurations were studied corresponding to a zero, intermediate (3.5 h^{-1}) and high (10 h^{-1}) air exchange rate (AER), respectively

mimicking a non-ventilated room, a well-ventilated room and a laboratory. Table i, in supplementary material reports the essential information regarding the ventilation conditions in the chamber. The air velocity profiles around the diffusers were measured with high spatial resolution in a separate setup at INRS, France, using a 3D ultrasonic anemometer (Kaijo WA-390 “Microsonic”, Sonic Corporation, Tokyo, Japan). Detailed velocity profile measurements are also reported in supplementary. In all experiments, the room temperature was set to 20°C, the atmospheric pressure to 1018 ± 10 hPa and the relative humidity to 40%. In the conditions of the experiments, the airflows in the chamber were isothermal, incompressible and turbulent with Reynolds numbers based on the size of the room ranging between $3 \cdot 10^4$ and $1 \cdot 10^5$.

In addition to the measurement of velocity profiles at the air inlet, the airflow in the room was characterised for each AER by measuring the local mean age of air (AoA) at 12 locations in the chamber, following the concept and the tracer gas concentration decay method described in Sandberg and Sjöberg (1983)¹⁷. The whole procedure and measurement results are provided in supplementary. The age-of-air data showed that a good air mixing occurs inside of the room, at least at the chosen measurement positions: for the highest AER of 10 h^{-1} , the measured AoAs differ by less than 13% from the perfect mixing time constant equal to AER^{-1} , with an average difference of 5%, indicating a near ideal mixing ventilation setup. For the lowest AER of 3.5 h^{-1} , the measured AoAs differ more from AER^{-1} but remain close, with an average deviation of 9% and a maximum of 16%. These results are consistent with the ventilation setup intended to favour mixing.

2.2 Experimental Scenarios

The scenarios were designed to determine whether any of the factors varied within the experiment (ventilation, source material, and emission type) has a significant impact on the concentrations measured in the NF and FF, and subsequently whether these factors can be considered as main determinants of exposure.

The experimental scenarios analysed within the scope of the present paper are summarized in Table 1. They include two repetitions for three of the scenarios to assess repeatability (namely: instrument intercomparison - E02, E05 -, constant emission at high AER – E03, E04 - and at low AER - E06, E07 -). Regarding exposure determinants, two AERs (3.5 and 10 h^{-1}), two source positions (close and far from the exhaust) and two types of temporal emission patterns (constant or intermittent in the form of regular spikes) were tested. The influence of the type of particles (NaCl, SiO_2 and sodium fluorescein) was also investigated. The minimum duration of experiments was chosen so as to cover the transient regime, a sufficient period of steady-state situation when existing (i.e. a minimum of 3 to 5 AER^{-1}) and the decay period of similar duration.

2.3 Measurement Instruments

Aerosol concentrations and PSDs were measured at eight different locations inside the chamber. The corresponding monitoring locations were denoted as near field (NF) when located within an 8 m^3 source-centred cube, and four FF locations (FF1 - FF4) when located outside this cube. In addition, the concentration directly above the source (denoted SF for source field), at the source (denoted SM for source monitoring), at the inlet and at the exhaust (denoted EXH) were recorded.

The instruments used were located on the roof of the chamber and 2.60 m copper sampling tubes were used to reach the desired measurement location (figure 1). The aerosol sampling height in the chamber was chosen to be 1.60 m, i.e. “about nose height”. Instruments were explicitly labelled by concatenating their location ID, instrument type and number (i.e FF1-CPC(1) stands for CPC#1 placed in FF1, see table iii in supplementary). The inner diameter of the copper tubes was selected according to the aerosol volume intake rate: 8 mm for the FMPS and ELPI (10 L/min) and 4 mm for the CPC, SMPS, DiSCmini and NSAM (≤ 5 L/min). The length of the inlets resulted in a monitoring delay time below 7 s. Results were corrected for the particle losses induced by the copper inlets according to Cheng¹⁸. Further details are provided in the supplementary files. The range of instruments used cover particle sizes from 3 nm to 10 μ m: CPC (3 nm – 10 μ m); DISCmini (10-700 nm); FMPS (5.6-560 nm); SMPS (3-800 nm); ELPI (6nm-10 μ m).

Since calibrating the instrument on site was not possible, an instrument inter-comparison was carried out. The ventilation was switched off and the chamber was homogeneously filled with the NaCl test particles thanks to a nebulization aerosol generator (AGK 2000, Palas GmbH, Karlsruhe, Germany) and stirred with a mixing fan. Then all instruments recorded the concentration decay in the room. Although they reported different concentrations, all instruments were found linear with respect to each other with a proportionality coefficient close to one except for NF-NSAM and FF4-ELPI(2) (figure v, and table v, Supplementary material).

2.4 Aerosol sources

Three types of sources of airborne particles were used in the experiments. Sodium chloride (NaCl) and sodium fluorescein (Na-Fluorescein) nanoparticles were aerosolized using the aforementioned AGK 2000 nebulizer. The emission nozzle of the nebulizer was equipped with a t-shaped side-sampling junction. 0.3 L/min of the 10 L/min total emitted aerosol flow were sampled after dilution (factor 1:91 $\pm$ 3 %, VKL 100 diluter, Palas GmbH, Karlsruhe, Germany). For the experiments E01-E08 and E11-E13 the nebulizer was run in continuous mode to generate increasing and finally steady-state NaCl or Na-Fluorescein aerosol concentrations. For experiments E09 and E10, the nebulizer was run in spiking mode by feeding it intermittently with 10 standard litres per minute (SLM) of carrier gas. This was done by time-varying the set-point of supply air flow controller from 0 to 10 SLM, following a chosen schedule. For most experiments, the diluted source emission was monitored continuously by a condensation particle counter (CPC), referred to as CPC(8) (model 5403, GRIMM GmbH & Co., Ainring, Germany). The PSD of the source was likewise measured on demand by means of the scanning mobility particle sizer referred to as SMPS(1) (model 3081+3082+3088, TSI Inc., Shoreview, Minnesota, USA). As the total PSD at the source was found repeatable and stable over time for a given set-point, it was not found necessary to measure it continuously with the SMPS.

In experiment E14, the silica powder (SiO₂) (EU Joint Research Centre Repository Test Materials: NM-203) was dispersed using a dry powder aerosol generator (Microdosing system, Fraunhofer ITEM, Hannover, Germany). The generator was operated in continuous mode at a pressure of 1.0 bar which generated an air flow-rate of 14.7 SLM.

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For all experiments except E11, the source was placed at the standard position 1 at a distance of 1.2 m from the front wall and 1.2 m from the left wall of the chamber, (figure 1). The nozzle height was 1.2 m. In E11, the source was moved exactly below the exhaust but kept the same elevation. The generation parameters of the aerosol sources that were employed in the various experiments are reported in Table 2. The total concentrations were obtained from instrument SM1-CPC(8) and the PSD parameters were obtained from instrument SM1-SMPS(1). Figure ix in supplementary material reports the detailed PSDs measured at the source. NaCl and Na-Fluorescein particles were log-normally distributed ($R^2 > 0.99$) and their PSD is hence fully described by their Geometric Mean Diameter (GMD) and Geometric Standard Deviation (GSD). Conversely, SiO_2 PSD departed more from log-normality with $R^2=0.94$.

2.5 Surface sampling

Surface sampling was performed to estimate deposition of particles in the chamber. Two types of samplers were used: electrically-conductive carbon tabs (sticky carbon tabs, Agar Scientific Ltd., Stansted, UK) and non-conductive polycarbonate membrane filters. Both types of sampling surfaces were flat and circular (diameter 2.5 cm) and were mounted on aluminium pin stubs. The stubs were fastened in plastic holders which were placed on walls, roof and ceiling of the chamber for collecting surface-deposited particles. For wall deposition experiment E12 with Na-Fluorescein particles, samples were first analysed with SEM before they were extracted in 2 ml of demineralized water and analysed with fluorescence spectrometry (480 nm in, 530 nm reflected).

3 Results

3.1 Particle deposition

Particle surface deposition was quantified through surface sampling only in experiment E12 which involved Na-Fluorescein particles with little dilution ($AER=3.5\text{ h}^{-1}$) and long exposure (13 h). For the conductive samples, the mass of deposited particles was found below the detection limit of $3\text{ }\mu\text{g.m}^{-2}$ even on the samples taken near the source, despite the long duration of E12 and fairly high aerosol concentrations (ca. $2.3.10^4\text{ \#.cm}^{-3}$). This detection limit corresponds to a deposition rate of $6.4.10^{-14}\text{ kg.m}^{-2}.\text{s}^{-1}$ given the duration of E12. Deposited particles were found on the non-conductive polycarbonate filters, but the difference between deposition on the conducting carbon and on the insulating polycarbonate surfaces indicates that electrostatic forces played an important role in the deposition on polycarbonate filters, and these samples are not believed to be representative of the surfaces of the chamber which were mostly made of painted sheet metal. Extrapolating to the whole surface of the chamber the conductive samples results suggests that the mass of Na-Fluorescein particles deposited in the chamber in E12 corresponds to less than 0.6 % of the mass of Na-Fluorescein exhausted by ventilation, given the air flow rate, exhaust particle concentration and PSD.

Considering the PSD of Na-Fluorescein particles in the chamber in E12 which was found homogeneous in space (lognormal distribution, $GMD = 68\text{ nm}$, $GSD = 1.6$, see Figure 5 and section 3.3.7), it is possible to extrapolate the detection limit in mass to a detection limit in number of particles. If we assume a purely diffusive deposition regime (compatible with the particle size range $10\text{ nm} - 200\text{ nm}$) and if we suppose

that the particle number deposition flux is size-dependent following a law in $Sc_b^{-2/3}$ (Sc_b being the size dependent Brownian particle Schmidt number, such as described in ¹⁹ and in many others sources), we may extrapolate the detection limit of $3 \mu\text{g.m}^{-2}$ for Na-Fluorescein deposition as being equivalent to a detection limit in number of $9.3.10^9 \text{ \#.m}^{-2}$. Given the 120 m^2 of surfaces in the chamber and the exposure duration of E12, this detection limit would have been reached only if the total deposition rate in the chamber had been greater than $2.4.10^7 \text{ \#.s}^{-1}$, which corresponds only to 1.3% of the Na-Fluorescein particle flow through the chamber.

This finding can be extended to experiment E08 which involved the same particle generation properties but a different AER (10 h^{-1} instead of 3.5 h^{-1}). According to PSD data from instrument EXH-FMPS(4) (figure 5 and supplementary table vii and figure xv) and to air flow data (supplementary table i), the particle mass flow rate at the exhaust was 20% lower in E08 than in E12, for an identical source generation rate, indicating a 20% increase of mass wall deposition. Consequently if less than $6.4 \cdot 10^{-14} \text{ kg.m}^{-2}.\text{s}^{-1}$ were deposited in E12, less than $7.7 \cdot 10^{-14} \text{ kg.m}^{-2}.\text{s}^{-1}$ were deposited in E08. Following the same reasoning than in E12, this can be extrapolated to a maximum total number deposition rate in the chamber in E08 of $4.10^7 \text{ \#.s}^{-1}$, i.e. 1.9% of the Na-Fluorescein particle flow through the chamber.

Consequently, however significant as regards surface contamination, the deposition rate in E12 and E08 was too low to affect airborne number concentrations by more than respectively 1.3% and 1.9%. For experiment E14 where the flow configuration

was identical to E12 but which involved SiO₂ particles presenting a PSD closer to the accumulation mode (i.e. less depositable), the influence of the deposition rate on number concentration is expected to be lower (consequently less than 1.3%).

For experiments (E03, E04) and (E06, E07), that involved identical airflow configurations than E08 and E12 but NaCl particles instead of Na-Fluorescein particles, the small effect of deposition on number concentrations is likely to be comparable to that found for respectively E08 and E12, given the similarities of PSD for NaCl and Na-Fluorescein particles (particles mobility diameter only 17% smaller in average and identical GSD). Confirming extrapolations can be made following the theory of a deposition law in $Sc_b^{-2/3}$, suggesting a total number deposition rate lower than 2% of the NaCl particle flow through the chamber.

3.2 Normalization of the concentrations and GMDs

All measurements of aerosol number concentration are normalized to allow for comparison between scenarios independently of instruments variability. The concentrations are hence normalized according to the inter-comparison results obtained in experiments E02 and E05 (i.e. rescaled as equivalent SF1-CPC(2) measurements). Normalization coefficients are provided in Table v in Supplementary Material. SF1-CPC(2) was chosen as reference instrument arbitrarily: only relative changes between measurement points and experimental scenarios are looked for, hence the choice of the reference instrument did not matter as long as instruments responses correlated linearly.

In a similar manner, GMD measurements for all granulometers are normalized according to inter-comparison results from E02 and E05 and rescaled as equivalent SM1-SMPS(1) GMD, this instrument being used to characterize the PSD of particles emitted by the source. Normalization coefficients for GMD are provided in table vi in supplementary and corresponding intercomparison results for GMD measurements are shown on supplementary figure vi.

For steady-state situations, concentrations are also normalized by the total number concentration measured at the exhaust in steady-state, which is representative of the perfect mixing concentration. This exhaust concentration was obtained either by instrument EXH-CPC(7) or EXH-FMPS(4) depending on data availability, and rescaled as equivalent SF1-CPC(2) measurement. Normalization concentrations for the cases involved are provided in table vii in supplementary. The purpose of this second normalization is to compare different scenarios from the point of view of spatial distribution only. Indeed, this normalization neutralizes the dilution effect of the AER and the concentrating effect of the source generation rate. Hence it enables the comparison of cases involving different AER and different particle types and generation rate, without seeing the mere effect of a global increase or decrease of total number concentration due to a modification of the AER or of the particle generation rate.

3.3 Spatial and temporal variability in particle concentrations and PSD

The two-box model assumes the existence of two well-mixed zones within which the variations of particle concentrations and PSDs are small compared to variations between NF-FF zones. This assumption is questioned by testing the spatial heterogeneity of particle number concentration and PSD in the room in the various experiments, and by analysing the dependence of this heterogeneity with respect to the parameters that were varied between the experiments (AER, particle type, source position and emission pattern).

3.3.1 Reference situation: stationary emission rate and high AER

Experiments E03, E04 and E08 considered steady state scenarios with a high AER (10 h^{-1}) in the chamber. For these scenarios, a steady generation rate of the source and steady ventilation leads to a statistically stationary aerosol number concentration field in the experimental chamber. E03 and E04 were repetitions of the same experiment with NaCl particles, while E08 concerned Na-Fluorescein particles. The time evolution of concentration recorded by the instruments was analysed to compute the mean steady-state concentrations reported in figure 2a. A noticeable NF-FF heterogeneity appears in the room for the reference scenario in steady state when the emission source is on: The NF zone exhibits a mean aerosol concentration about 4 fold higher than the FF zones, where the concentration is generally homogeneous and close to the perfect-mixing concentration found at exhaust. The FF1 zone tends to depart a bit from this homogeneity, but the measured concentration is still noticeably lower than the NF concentration.

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As E03 and E04 are repetitions of the same experiment, comparison of measured values for these experiments and for the different instruments in figure 2 highlights the good repeatability of these stationary experiments, since the variation of means between identical experiments is well below 10% for most locations. This repeatability highlights the reliability of the experimental set-up.

These results may be compared to the simulation results of Jensen *et al.*¹⁶, who applied a multi-box modelling approach to the present experimental configurations. Jensen *et al.*¹⁶ thus propose modeling results for a one-box, a two-box and two three-box decompositions of space in the chamber. For a stationary source emission rate and a high AER the existence of a concentration difference between NF and FF is of course reproduced only by their two-box and three-box configurations, indicating a better representativity of models with more than one box. However they obtain a mean aerosol concentration in NF which is only 2 fold higher than in the FF zones, which is only one half of what is observed experimentally.

3.3.2 Steadiness over time of concentration hierarchy between NF and FF for the reference situation

In accordance with a NF-FF hypothesis, the hierarchy of particle number concentration between near-field and far-field must be maintained over time while the source is active. This steadiness of concentration hierarchy can be clearly seen for scenarios with a steady source and high AER (10 h^{-1}), as is illustrated in figure 3 which represents time series of concentration differences between all NF and FF instruments for experiment E03. For the first 48 minutes, when the source is active, the difference of concentrations between the NF and FF instruments is always noticeably higher than the difference between FF instruments. Soon after the

emission stops, the concentrations drop and the difference of concentration between the NF-FF and FF-FF positions becomes indiscernible. Note that this behaviour is satisfactorily reproduced by multizone modelling approaches, as shown in Jensen *et al.*¹² for the same configuration.

3.3.3 Influence of the ventilation rate

Experiments E06 and E07 were identical to experiments E03 and E04 (same source), except that the AER in the chamber was reduced from 10 to 3.5 h⁻¹. This allows analysing the effect of the ventilation rate on the spatial homogeneity of concentration. figure 2b thus shows the mean steady-state concentrations at the various positions in the chamber measured by the different instruments for an AER of 10 h⁻¹ (E03) and of 3.5 h⁻¹ (E06, E07). Results from E04 and E08 are not represented given the similarity of E03, E04, E08 shown in figure 2a.

In the present setup, it is found that decreasing the AER increases the homogeneity of concentration in the room: the differences between NF and FF become indiscernible at 3.5 h⁻¹ compared to 10 h⁻¹. The FF3 position tends even to present a higher concentration than the NF position at 3.5 h⁻¹. This situation can be attributed to a higher influence of internal mixing over renewal rate when the ventilation rate is decreased. For this setup, it is apparent that the concept of NF and FF finds its limit for the lowest AER.

Once again, comparison between E06 and E07 results highlights the repeatability of these experiments.

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For the same configurations, Jensen *et al.*¹² reported the following modelling results: all modelled multizone-configurations predicted a similar NF-FF hierarchy for particles concentrations, with a concentration in NF 2 fold higher than in the FF zones in steady state. This is not consistent with measurements which show no significant separation of space in two zones for an AER of 3.5 h⁻¹. In this case, the multizone modelling such as proposed in Jensen *et al.*¹² does not reproduce the effect of a change of AER.

3.3.4 Influence of the particle type

If Na-Fluorescein particles (GMD=35 nm, bulk density 1600 kg·m⁻³) are generated instead of NaCl particles (GMD=29 nm, bulk density 2170 kg·m⁻³, see Table 2), while keeping the same experimental setup (same generator, same position and identical AER), no significant changes in the spatial distribution of measured concentrations appear. This can be seen in figure 2a for the stationary emission at the higher ventilation rate (10 h⁻¹) (E03, E04 vs. E08). This is also observed at the lower ventilation rate when comparing E06, E07 vs. E12 on figure 2b. When SiO₂ nanoparticles are used, the same trend is observed (E14, figure 2b), however the instability of the SiO₂ generator (50% variation of the generation rate) leads to more unstable measured concentrations and hence to less conclusive observations (in E14, the variations between instruments at the same measurement point are noticeably higher than in other experiments, especially CPC vs. DiSCmini between which a 4:1 ratio is observed at NF and FF4 positions). When comparing E03 to E06, E07 and E12 on figure 2b, it is clear that the effect of the AER on the spatial distribution of concentration predominates over the effect of particle type.

3.3.5 Influence of source position

In experiment E11, the source position was changed with respect to other experiments, while keeping an AER of 10 h^{-1} such as in the reference situation (E03, E04). The new source position was situated below the exhaust, as to mimic the effect of local exhaust ventilation. The new source position was hence immediately below the EXH sampling and noticeably closer to FF2, FF4 and FF3 samplings. This led to significantly lower concentrations than in the reference situation (figure 2c), as an important part of the emitted particle was directly removed by the exhaust. Regarding the heterogeneity of particles concentration in the room, it is obvious that the highest concentrations are always reached around the source and that concentrations decrease further away, the minimum being measured nearby the old source position (NF) and in FF1. The existence of a 4:1 ratio between the most concentrated zone (nearby the source: NF sensors in E03 and EXH-CPC(7) sensor in E11) and the least concentrated zone in the chamber, away from the source, indicates a spatial variability which is compatible with a NF-FF hypothesis, as in the reference situation.

3.3.6 Influence of the emission pattern

Experiments E09 and E10 aimed at studying the influence of a time-varying source generation rate on the space and time dispersion of nanoscale aerosols in the chamber. Figure 4 thus shows the time-resolved concentrations measured at the different positions for the spiking generation rate of E09 at an AER of 3.5 h^{-1} . In E09, emission spikes are separated by a decay period of 15 min, which is very close to the

air renewal time constant AER^{-1} of 17 min. For all positions, the typical dilution/diffusion/time-shifting effect of turbulent transport by ventilation is clearly seen. It is apparent that the emitted particles first reach the NF position, the exhaust and the FF3 position before reaching the FF1 and FF4 zone, these two last regions presenting a much weaker source/response transfer coefficient than the other regions. These time series do not highlight the existence of a clear separation between a NF region, where the response to the source would be high and quick, and a FF region presenting a dampened and delayed response to the source. It is likely that the sensor responses are largely explained by the complex organization of flow patterns in the chamber, rather than by mere distances to the source: the FF3 and exhaust positions react more and more quickly to an increase of the source generation rate than the FF1 position which is closer to the source. These observations can be highlighted by signal cross-correlations (supplementary figure xvi). It is worth noting that when applying multi-zone modeling to this configuration, Jensen *et al.*¹² observed that the dynamic of aerosol transport in the chamber could only be reproduced satisfactorily by using three boxes rather than two boxes. In particular, the 3-boxes layout composed of cylindrical nested boxes centred on the source performed best in its ability to reproduce the broadening of the concentration profiles and the timing of the peaks at the FF positions. This observation is consistent with experiments for which a multi-zone behaviour is observed.

The effect of the emission pattern on the space-time dispersion of aerosols was also investigated for a higher AER of 10 h^{-1} in experiment E10 (not shown here for brevity). In E10, the period between emission spikes was shorter than the air renewal time constant AER^{-1} in the chamber (2 min between spikes, i.e. one third of AER^{-1}). It was observed that the concentrations in FF points did not have enough

time to decay significantly between spikes, thus the effect of source intermittency was blurred in FF, leading to a rather steady situation in FF. Meanwhile, the effect of source intermittency remained clearly visible in NF. Hence, a clear separation between a NF region where the response to the source is high and quick and a FF region presenting a dampened and delayed response to the source exists in E10. A generalization is not possible only from these two experiments with unsteady sources; however, we can advance that the effect of source intermittency is unlikely to be noticeable in FF if the source intermittency period is small compared to the air characteristic mixing time, which is close to AER^{-1} for mixing ventilation. The existence of a clear separation between NF and FF for intermittency is hence probably largely dependent on the ratio of the source and mixing time scales.

3.3.7 Spatial variations of the particle size distribution

The mean PSD measured by all granulometers in the room for all steady experiments are reported in supplementary figure xiv, together with log-normal fits and associated determination coefficients R^2 . The high R^2 values with respect to log-normality indicate that the description of the PSD can be efficiently summarized by considering only the evolution of the GMD and GSD throughout the chamber. It is also found that the GSD of particles diameters is homogeneous in space for all experiments with continuous source (figure xv in supplementary), the GSD being even the same than that of the generated particles (within experimental fluctuations of Table 2). As a consequence, the particles GMD is a sufficient parameter to describe the behavior of the PSD under the different scenarios and positions in the chamber.

Figure 5 reports the mean GMD of particles measured by all granulometers in the room for all steady experiments, rescaled as equivalent SM1-SMPS(1) measurement. The GMDs of the particles emitted by the source are reported as a reference.

From figure 5, we observe that for a given experimental scenario the variations of the GMD in space are small ($< 6\%$ for NaCl and Na-Fluorescein particles, and $< 10\%$ for SiO_2 particles) and smaller than their temporal fluctuation (and than the temporal GMD fluctuation of the source for SiO_2), hence there is no significant variations of GMD in space, if we disregard temporal fluctuations. The PSD then appears homogeneous in space (apart total concentrations).

Conversely, we observe a noticeable effect of the AER and of the source position on the particle GMD in the chamber. Thus, decreasing the AER from 10 h^{-1} to 3.5 h^{-1} , such as between (E03, E04) and (E06, E07), or between E08 and E12, leads to an increase in particles GMD of 16% to 11%.

Similarly, when the source is situated immediately below the exhaust in E11, the GMD of particles in the chamber is reduced by 8% with respect to (E03, E04), while the AER remains the same. In E11, the observed GMD is indeed closer to the GMD measured at the source by the SMPS.

Overall, the GMD increase with respect to that of the generated particles ranges between 13% for SiO_2 particles to 92% for Na-Fluorescein particles in E12, with intermediate values of 35% to 72% for NaCl particles. For particles in this size range, which are insensitive to sedimentation and to inertial separation induced by

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accelerations of the carrier airflow, the observed PSD shift towards higher diameters can only be explained by size-dependent deposition or aggregation. If we assume a purely diffusive deposition regime (compatible with the involved PSD range 10 nm – 300 nm) and if we suppose that the particle number deposition flux is size-dependent following a law in $Sc_b^{-2/3}$, as already introduced in section 3.1, it is possible to show that pure deposition cannot explain an increase of GMD greater than a certain extent when the source is active. Considering the PSD of the particles when they are released in the chamber (Table 2), this maximum deposition-induced GMD increase is equal to 31% for NaCl and Na-Fluorescein particles and 52% for SiO₂ particles. Consequently the observed evolutions of GMD are likely to be attributable to a combination of both deposition and aggregation, which respective influence is not precisely measured with the current setup, given the absence of quantified wall deposition. However, if the assumptions underlying the deductions from the deposition result section are verified, the deposition rate is smaller than a few percent of the source generation rate and hence most of the PSD shift is attributable to aggregation.

4 Summary and discussion

The observations made for the specific situations of the experiments can be summarized as follows:

- When measured, the deposition rate is found too low to affect airborne number concentrations by more than about 1%, for the 8 – 100 nm Na-fluorescein particles when AER = 3.5 h⁻¹. Physical considerations suggest that this can be generalized to all the considered scenarios i.e. to particles with

mobility diameter in [8 – 600 nm] and to AER in [3.5 – 10 h⁻¹];

- apart total concentrations, no spatial variation of PSD in the room is observed for any of the studied scenarios : the log-normality is preserved and GMD and GSD spatial variations are below their temporal fluctuations. This means that size-dependent number losses like deposition and aggregation are not locally influent in the present cases ;
- a slight and general PSD shift (-8% to +16% modification of the particle GMD) is however observed in correlation with the AER and with the source position, for a given source generation properties. This indicates a direct effect of total number concentration and particles residence time on the PSD, a shift towards higher diameters being observed for higher concentrations and residence time (i.e for lower AER and increased distance of the source with respect to the exhaust). This can be attributed to a combination of size-dependent deposition and aggregation, the estimated low deposition rates suggesting a predominant role of aggregation ;
- For given ventilation rate, emission scenario and number generation rate of aerosol sources, the particles composition and size (NaCl, Na fluorescein and SiO₂) were not seen to influence dispersion. This is expected considering the predominance of flow transport over particle dependent phenomena (diffusion, aggregation) for the considered PSD, number concentration and ventilation conditions.

Concerning two-zone modelling, the results suggest the following conclusions, for the specific situations of the experiments:

- the spatial heterogeneity of concentrations between the NF and FF depends

strongly on the ventilation rate. For the studied room volume of 81 m^3 , a rather indiscernible difference between the NF and FF total particle number concentration was found when the ventilation rate was lowered to 3.5 h^{-1} . This is not reproduced by the two-zone modelling reported in Jensen *et al.*¹⁶ which predicts a systematic difference between zones ;

- the difference in NF-FF concentrations depends on the source activity: the differences of concentrations between NF and FF become insignificant shortly after the emission stopped. This aspect is properly reproduced by two-zone modelling¹⁶;
- the spatial heterogeneity between the NF and FF is observed independently of the source position. It exists either when the source and exhaust are 3 m apart (general ventilation scenario) or when the source is just below the exhaust (scenario with a single local exhaust ventilation);
- for unsteady emission scenarios, the existence of a clear separation between a NF region (highly and quickly source-responsive) and a FF region (presenting a dampened and delayed response to the source) seems to be largely dependent on the ratio of the emission period over the ventilation time scale. This separation is hence case dependent and a modelling based on an immutable separation of space in two zones is unlikely to reproduce all situations. Thus, for the present experimental setup, results are reproduced by multi-zone modelling only if more than two boxes are used¹⁶, following a specific layout;
- Given the observed homogeneity of the PSD in the room for the different scenarios, there does not seem to exist a NF or FF from a PSD point of view.

These findings might have been affected by experimental data gaps and

irregularities, unavoidable during the operation of up to 18 instruments in a limited time. A summary of experimental irregularities is provided in supplementary material. Concerning the estimation of deposition through surface sampling, it is obvious that it is truly limited by technical constraints, only one product being sampled, the limit of quantification not being reached and a limited area being covered by samples. This limits the possibility of distinguishing the effects of deposition and aggregation.

Beyond these experimental uncertainties, generalizing the conditions under which a two-zone exposure pattern exists from these results is hazardous, since these experiments are necessarily limited regarding ventilation design, geometrical layout and particles used, among others. In particular, the effect of ventilation was considered only from an AER point of view. It is known however that the ventilation design is also decisive on the control of airborne contaminants, and current setup reproduces only the case of mixing ventilation, without considering the effect of a different concept such as displacement ventilation, or simply of a different layout. Consequently, results should rather be considered as forming necessary but not sufficient validation test-cases for dispersion models.

Some aspects of these results can be put in perspective with several real scale studies in the literature. In the vast majority of situations, which can be either field studies^{11,14,15} or lab studies^{10,12,13,20–30}, the existence of a NF around the source presenting a noticeably higher contaminant concentration than the rest of the indoor environment is attested in steady-state. This was actually the main motivation for the development of the two-zone modelling concept⁹. Note however that if that is the case for point sources, such as in the present experiments, it is not always the case

for wide sources such as surface sources³⁰. Not also that situations where NF concentration are lower than FF concentrations are rare though they occasionally happen^{13,24}. It is also interesting to observe that while investigating the consequences of the two-zone model, Cherrie^{31,32} performed parametric simulations involving variable room size and AER, encompassing the current experimental situations. These theoretical computations predicted that decreasing the AER from 10h^{-1} to 3h^{-1} for a chamber of 100m^3 (which corresponds approximately to switching from experiments E03,E04,E08 to E06, E07 E12) would lead to an increased homogeneity of concentration in the chamber, that is to a decrease of the NF/FF concentration ratio. This theoretical prediction of the two-zone model is hence fully in accordance with our experimental results. More specifically, according to computations from Cherrie^{31,32}, the current experimental setup which yields to a NF/FF ratio of 4 at and AER of 10h^{-1} would correspond to an intermediate airflow between NF and FF (about $0.12\text{m}^3.\text{s}^{-1}$). This finding actually supports the two-zone modelling concept. Regarding the sensitivity of the aerosol distribution in the room with respect to the source position, for a given ventilation layout, this was also widely reported in the literature^{20,21,30,33,34}. As for the negligible contribution of deposition on particle balance in the room, this was already observed by the pioneer work of Murakami et al.³³ for a chamber of similar size (54m^3) and particles with diameter ranging between 0.31 and $4.5\text{ }\mu\text{m}$, but for a notably higher AER (42h^{-1}) and a number concentration 4 order of magnitude lower (that is, in conditions where the particle wall deposition is likely to be more influent due to increased friction velocity, and where aggregation is negligible). The same author also observed the similarity of transport for passive scalar and aerosol particle in the same conditions. Considering this state of the art, the observations made in the present experiment

are not surprising but they extend the available datasets to situations involving polydisperse aerosols in the sub-micron range, and time-varying situations.

Some of the results regarding particle transport are likely to be generalizable to situations involving similar AER and PSD and smaller or equal particle concentrations. This is probably the case for deposition, which was not found to affect total number concentrations by more than 1-2% percent when the source is active, independently of AER and particle type and size (within the range 8–600 nm) and for an average total number concentration below $4 \cdot 10^4 \text{ cm}^{-3}$. Indeed, particle deposition rate is known to be proportional to bulk concentration, surface areas, wall friction velocity and to the deposition velocity. This deposition velocity depends on particle inertia and diffusivity, surface orientation and friction velocity¹⁴. In current experiments, the values of these deposition parameters are likely to be similar to those frequently met in indoor air, considering the experimental setup. Hence for similar situations (PSD and concentration, room volume, AER and exposure duration) the effect of particle wall losses on number concentrations is likely to be indiscernible, as long as the source of particles is active.

The same reasoning applies to aggregation: the aggregation rate scales with the square of particles concentration and is mostly particle size dependent and Kolmogorov's time scale dependent³⁵. Associated with the aerosol residence time, this rate determines the growth of particle size and the decrease of number concentration due to aggregation. The values for the aggregation parameters involved in current experiments (residence time, Kolmogorov's time scale, PSD and concentration) are typical of many situations for which aggregation should hence play a similar role. Since no spatial variations of the PSD are observed between the

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measurement points, the only region where aggregation is likely to be significant is the aerosol source, given the high concentration involved ($\sim 10^7 \text{ cm}^{-3}$). However this effect must be limited by the rapid dilution of particles in the room to concentrations below $4 \cdot 10^4 \text{ cm}^{-3}$.

5 Conclusion

A particle exposure experiment inside a large climate-controlled chamber was conducted. Data on spatial and temporal distribution of nanoscale and fine aerosols in the range of mobility diameters 8 – 600 nm were collected with high resolution. Different exposure scenarios were studied, including constant and intermittent source emissions, aggregation conditions, high (10 h^{-1}) and low (3.5 h^{-1}) AERs corresponding to chamber Reynolds number between $3 \cdot 10^4$ and $1 \cdot 10^5$. The main aim was to collect a set of data allowing evaluating how close to reality is the two-box model simplification, in terms of spatial resolution, and how the AER, type of emission (continuous to intermittent) and type of particle influence its representativeness.

We observed that under the conditions of the experiment the NF-FF simplification generally holds and that the source position in regard to the exhaust does not affect the NF-FF structuring.

In present experiment, the NF-FF spatial simplification could even be reduced to one box for AERs lower or equal to 3.5 h^{-1} , with constant emission sources and after reaching steady state. However, in actual situations involving nanoparticles sources indoor, emission rates will not be constant and steady state will not be reached, hence a difference between NF and FF is likely to exist.

Accepted Article

For the considered scenarios, the effect of particle aggregation is visible but is not a major determinant of exposure (-8% to +16% variations of the GMD across scenarios), thus its modelling could be avoided if only first order effects are considered. In addition, deposition was not found to affect total number concentrations by more than 2% when ventilation was running and source was active. This secondary yet noticeable role of aggregation and deposition on aerosol space-time dispersion is likely to be generalizable to situations involving similar AER and aerosol residence time, PSD and total number concentration.

The conditions under which a two-zone model is applicable to nanoscale and fine aerosols cannot be deduced only from these experiments: the best possibility of obtaining generalizable results relies on using the collected data to validate various dispersion models, including more generalizable models such as CFD-based models.

6 Supporting information

The data generated within these experiments is available publicly. The NANoREG results repository is available at <http://www.nanoreg.eu/>

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- Accepted Article
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Tables

Table 1: Exposure determinants for each scenario.

Scenario ID	Task	Source material	AER (h ⁻¹)	Emission pattern	Source Position*	Emission Duration (min)	Decay duration (min)	total time (min)
E02	Instrument inter-comparison	NaCl	0	Constant	Position 1	4	242	246
E03	Constant emission at high AER	NaCl	10	Constant	Position 1	48	30	78
E04	Constant emission at high AER	NaCl	10	Constant	Position 1	40	30	70
E05	Instrument inter-comparison	NaCl	0	Constant	Position 1	49	865	914
E06	Constant emission at low AER	NaCl	3.5	Constant	Position 1	159	85	244
E07	Constant emission at low AER	NaCl	3.5	Constant	Position 1	90	84	174
E08	Constant emission at high AER	Na-fluorescein	10	Constant	Position 1	30	30	60
E09	Intermittent emission at low AER	NaCl	3.5	Spiking	Position 1	1 min each every 15 min	15 min betw. spikes, 105 min after last spike	202
E10	Intermittent emission at high AER	NaCl	10	Spiking	Position 1	1 min each every 2 min	2 min betw. spikes, 45 min after last spike	75
E11	Constant emission at high AER with moved source	NaCl	10	Constant	Position 2	64	32	96
E12	Wall deposition experiment: constant emission at low AER	Na-fluorescein	3.5	Constant	Position 1	783	0	783
E14	Silica	Silica	3.5	Constant	Position 1	88	62	150

(*) Position 1 : away from exhaust (Figure 1), Position 2 : below exhaust

Table 2: Properties of the aerosol sources

Material	Experiment	Carrier flow rate (SLM)	Total concentration at injection - SM1-CPC(8) (#.cm ⁻³)		Generation rate (#.s ⁻¹) SM1- CPC(8)		GMD (nm) SM1-SMPS(1)		GSD SM1-SMPS(1)	
			mean	std. dev.	mean	std. dev.	mean	std. dev.	mean	std. dev.
NaCl	E01-E07, E09-E11, E13	10±0.6%	2.24E+07	2.13E+05	3.73E+09	3.55E+07	29.3	0.57	1.59	0.006
Na-Fluorescein	E8	10±0.6%	9.54E+06	1.28E+05	1.59E+09	2.13E+07	35.3	0.37	1.59	0.003
Na-Fluorescein	E12	10±0.6%	9.06E+06	4.72E+05	1.51E+09	7.87E+07	35.3	0.37	1.59	0.003
SiO ₂	E14	14.7	4.02E+05	1.89E+05	6.70E+07	3.15E+07	121.7	8.25	1.94	0.12

Table 3: list of acronyms

General purpose acronyms					
AER	Air Exchange Rate	ENM	Engineered nanomaterial	CFD	Computational Fluid Dynamics
AoA	Age of air				
Measurement location		Measurement instrument		Measurement properties	
EXH	Exhaust	CPC	Condensation Particle Counter	PSD	Particle Size Distribution
FF	Far Field	DISCmini	Handheld Nanoparticle Counter	GMD	Geometric mean diameter
NF	Near Field	ELPI	Electrical Low Pressure Impactor	GSD	Geometric standard deviation
SF	Source Field	NSAM	Nanoparticle Surface Area Monitor	SLM	Standard liters per minute (0°C, 1.013e5 Pa)
SM	Source Monitoring	SMPS	Scanning mobility particle sizer		

Figure legends

Figure 1: Schematic representation of the experimental chamber and position of the instruments and ventilation (SRC: source, NF: near field; FF: far field, IN: air supply, EXH: air exhaust)

Figure 2: Effect of AER, particle type and source position on NF-FF mean total number concentrations (TNC) for steady-state situations. Concentrations are normalized by the mean TNC at exhaust. Error bars correspond to 95% confidence intervals on means. (a) Reference situation: high AER scenarios, (b) scenarios for different particle types and AER compared to reference situation, (c) effect of moved source.

Figure 3 : Time series of the concentration differences between NF and FF sensors and between the FF sensors, for experiment E03 (continuous emission of NaCl, AER of 10 h^{-1}). Signals are filtered with a 5 min moving-average filter to improve readability. Considered instruments are NF-CPC(3), NF-DiscMini (1), NF-FMPS(1) and FF1-CPC(1), FF1-SMPS(2), FF2-CPC(4), FF2-FMPS(2), FF3-CPC(5), FF4-CPC(6), FF4-DiscMini(2), FF4-FMPS(3). Differences are normalized by mean total number concentration at exhaust.

Figure 4: time-resolved total number concentrations measured at the different positions in case of a spiking source at AER of 3.5 h^{-1} – Experiment E09

Figure 5 : Particles geometric mean diameter in steady-state at the different measurement points for steady-state experiments with continuous emission source. Error bars correspond to two standard deviations of measurements. Extra horizontal grid lines correspond to particles GMD of the different sources such as measured by SM1-SMPS(1)

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5