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1	Investigation of PM ₁₀ , PM _{2.5} , PM ₁ in an unoccupied airflow-controlled room:
2	how reliable to neglect resuspension and assume unreactive particles?
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14 Abstract

15 It is now well recognized that Particulate Matter (PM) is one of the main air pollutants affecting 16 both ambient and indoor air quality. While ambient PM mass concentration measurements are 17 often performed by air quality monitoring networks, current regulations do not address their 18 indoor concentrations. The latter can be estimated nonetheless from a mass balance analysis 19 accounting for (i) the transfer of particles from outdoor and (ii) their typical indoor sources 20 (emissions, resuspension) and sinks (deposition, removal by air exchange). Inherently, the mass 21 balance analysis is valid with inert atmospheric species, i.e. mass is conserved and described by sources and sinks, thus assuming no physicochemical transformations. To check that the relative 22 23 imbalance (RI) is not significant over different size fractions, a series of careful measurements 24 were conducted in an unoccupied room within a building incorporating an energy-efficient design 25 (minimal heat losses, negligible air leaks, etc.). We show that by carefully characterizing the room properties, i.e. the air exchange rate, penetration factors and deposition rates for sized-26 resolved particles, the PM_{10} and $PM_{2.5}$ fractions, which are typically regulated outdoors, as well 27 28 as PM₁, can be well estimated indoors (with RI between measured and expected values < 19%) 29 under moderate ambient temperatures (< 22°C in this study). However, RI increases significantly, 30 especially for submicron particles, at higher temperatures, indicating possible transformations in 31 the particulate phase, which are not accounted for by the mass balance model. Therefore caution 32 is recommended regarding the mass balance analysis to estimate PM fractions indoors, especially 33 for PM₁.

34

35 Keywords: Indoor Air Quality, Particle penetration, Particle deposition, Aerosol budget, Mass
36 balance

37 **1. Introduction**

38 The infiltration of outdoor pollution combined with a range of indoor sources emitting volatile 39 organic compounds (VOCs) and particulate matter (PM) tends to lead to a lower air quality indoors than outdoors.¹ According to the World Health Organization (WHO) ² 3.8 million 40 41 premature deaths worldwide were attributable to household air pollution in 2016, accounting for 7.7% of the global mortality. The design of effective strategies aiming at reducing personal 42 43 exposure to harmful species requires a good assessment of pollutant concentrations in the indoor 44 environment, which in turn requires a good understanding of their sources, their sinks and all the 45 physicochemical processes leading to their transformations.

Among atmospheric pollutants, PM has been classified as carcinogenic for humans,³ and several 46 47 countries have set air quality standards on ambient mass concentrations of PM with an 48 aerodynamic diameter below 10 µm (PM₁₀) or 2.5 µm (PM_{2.5}, also known as fine particles). PM 49 of even smaller sizes, e.g. with a diameter lower than 1 µm (PM₁), is of particular interest since it 50 deposits more easily in the lower respiratory tract where it can penetrate into the circulatory 51 system, impacting other organs.^{4,5} Furthermore, PM₁ are observed at high number concentrations 52 compared to larger particles, leading to large surface areas that can carry adsorbed organic pollutants to the circulatory system.⁶ 53

To better understand the sources and properties of PM on indoor air, previous studies have been generally conducted either in simplified environments such as experimental chambers^{7–12} or proxy rooms,¹³ or in real buildings. In the former case, the focus is mainly on the formation of secondary pollutants while the latter is aiming at assessing the anthropogenic influence by investigating the impact of specific activities (such as cooking, candle combustion, use of cleaning products, etc.) on particle nucleation and growth,^{14–20} or by focusing on the possible transformations and origin of indoor particles under occupied conditions.^{20–25} Interestingly, only a few studies focus on the investigation of particle origin under unoccupied conditions in real rooms, therefore there is often a lack of understanding of the building influence itself.^{23,26–29}

63 Indoor concentrations of particles are typically driven by several processes acting either as64 sources or sinks as expressed in the mass balance model (Eq. 1).

65
$$V\frac{dC_{in}}{dt} = \alpha PVC_{out} - (\alpha + K)VC_{in} + R + S$$
 {1}

where C_{in} and C_{out} are the indoor and outdoor concentrations (µg m⁻³), respectively, *V* the volume of the test room (m³), α the air exchange rate (h⁻¹), *P* the penetration factor (dimensionless), *K* the deposition rate (h⁻¹), R the resuspension rate (µg h⁻¹) and S the net results of additional processes acting as sources or sinks (µg h⁻¹). The latter can be due to either direct emissions from human activities such as cooking or cleaning, as well as physicochemical transformations of existing particles such as condensation, evaporation and chemical reactivity.

The first term on the right-hand side of Equation 1 can be considered as a source of particles originating from outdoor due to air exchange. The second term represents an indoor sink due to air exchange and deposition. The third term, resuspension, acts as a source mainly for particles larger than 1 μ m. For smaller particles the resuspension is likely negligible because they are more difficult to detach from the floor due to relatively higher adhesive forces compared to removal forces.^{30–32} The last term, S, aggregates additional sources or sinks due to indoor activities as mentioned above. The accuracy associated to indoor budgets of PM_x (x = 1, 2.5, 10) derived from Equation 1 depends on how well each term can be characterized, which is extremely challenging given their dependence on the particle size and their co-variability.³³

The mass balance equation (MBE) has been previously used in various studies to (i) estimate indoor concentrations of particles based on observed outdoor concentrations, (ii) quantify some parameters such as particle penetration, deposition, and resuspension rates,^{27,30–32,34–55} and (iii) estimate the strength of indoor particle sources resulting from human activities.^{46,47,56–62} In most of these studies however, two important simplifications are usually made: (i) particles are inert species and their number concentration and size does not change and (ii) particle resuspension is negligible under unoccupied conditions.

88 However, the size distribution of outdoor particles being transferred indoors could be impacted 89 by volatilization, condensation or coagulation processes due to a change in environmental conditions. Interestingly, Abt et al.,⁴⁷ performed measurements of continuous particle size and 90 91 mass concentration indoors and outdoors in four nonsmoking households located in the 92 metropolitan Boston area during winter and summer. The authors used the MBE to determine the 93 source emission and infiltration rates for specific particle sizes, as well as the contribution of 94 outdoor and indoor sources to indoor particle levels. The results showed that particles in the size 95 range $0.2 - 10.0 \,\mu\text{m}$ can be estimated within a median relative error of 20-50% using outdoor 96 concentrations and time-activity data, while for smaller particles $(0.02 - 0.2 \,\mu\text{m})$ the relative error 97 increases to 107%. This underestimation of the model may be related to the large variability in 98 indoor concentrations of smaller particles due to the contribution of specific activities to indoor 99 particle levels (cooking). In addition, transformations of particles penetrating indoors have been observed in some studies.^{43,57,59,63–66} For example Polidori et al., 2006 ⁵⁹ showed that 71-76% of 100

101 the organic carbon in $PM_{2.5}$ is most likely formed or emitted indoors rather than penetrating from 102 outdoors. Hodas and Turpin (2014)⁶³ estimated that changes in the organic aerosol mass indoors 103 due to shifts in the gas-to-particle partitioning can reach 11-27%, while Rim et al. (2016) 104 indicated that ignoring coagulation for the prediction of ultrafine particles clearly leads to an 105 underestimation of indoor concentrations. These observations challenge the assumption of 106 particles being inert species when the MBE is used.

107 Chan et al.⁶⁷ and Chithra and Nagendra⁶⁸ also performed measurements of particulate matter 108 indoors and outdoors (PM_{2.5} in homes, and PM₁, PM_{2.5}, PM₁₀ in classrooms respectively), where 109 human activities took place. They both used indoor models based on MBE in order to 110 characterize indoor particles and/or source strengths of known indoor activities and subsequently 111 predict the indoor concentrations. However, assumptions or limitations such as excluding 112 processes of fine particles in the model (i.e. particle transformations and resuspension) were 113 made. The authors found a good agreement between measured and estimated PM2.5 114 concentrations indoors when the human activities are well identified and characterized. However, the latter showed that PM₁ and PM₁₀ concentrations can be wrongly estimated by not including 115 116 reactivity and resuspension respectively in the model. From the above discussion, it is clear that 117 common assumptions in the use of the MBE, i.e. particles being inert species and particle 118 resuspension being negligible, need to be carefully reconsidered.

The objective of this work is to investigate whether the mass balance model described in Equation 1, assuming in a first step unreactive species (no particle processing) and negligible resuspension under unoccupied conditions (anthropogenic influence is negligible), could accurately predict indoor concentrations of PM_x (x = 1, 2.5, 10) if the parameters in this equation were accurately quantified for each PM size fraction, or whether physicochemical 124 transformations and resuspension need to be included in the model, unlike to what has been 125 mainly considered until recently. In this work, we first characterized each term of the mass 126 balance model (Eq. 1) for an empty room of a low-energy building (described in Sections 2.2 and 127 3.1), and we then used the MBE to investigate how indoor measurements of PM_x compare to 128 values inferred from the model constrained by particulate outdoor measurements (Sections 2.3 129 and 4). Subsequently, we investigate whether this approach can highlight physical and chemical 130 processes occurring in the room, and therefore if the equation can be applied directly in more 131 complex situations, as usually assumed.

- 132 **2.** Materials and methods
- 133 **2.1. Building and campaign description**

134 The measurement facility is located at the Institute Mines Telecom Lille Douai, on the Douai 135 Campus, in northern France. This facility is a wooden building, a so-called energy-efficient 136 building with the BBC label ("Bâtiment Basse Consommation"), which according to the French 137 thermal regulation RT2012 has a primary energy consumption of maximum 50 kWh of oil equivalent per m² per year on average.⁶⁹ This type of building is representative of recent 138 139 constructions aiming at significantly reducing the carbon footprint from heating and hot water 140 production, and therefore represents a trend for future constructions.⁷⁰ The building is composed of two similar rooms of approximately 12 m² of surface area and 2.4 m height, leading to a 141 142 surface to volume ratio of 0.4 m²/m³. Only one room (hereafter called "test room") was used to 143 perform all the characterization experiments (quantification of the parameters shown in Eq. 1) 144 and the simultaneous indoor/outdoor particle measurements, with no air exchange between the 145 two rooms. The test room is equipped with a dual-flow wall unit with heat recovery (KWL EC 60

Pro, HELIOS) providing an air flow rate of 17 m³ h⁻¹. This system includes a G4 filter for the 146 147 efficient removal of coarse particles. Ventilation was kept on during the whole measurement 148 period. In order to achieve the standards of energy-efficient buildings, the building is equipped 149 with thermally efficient windows (triple-glazed with Argon filling and an aluminum frame) and 150 the walls and roof are composed of wood, and insulated using wood wool and fibers. The floor is 151 covered by linoleum on top of wood wool and an OSB board. The external door is made of 33-152 mm thick aluminum profiles and a triple-glazed window. Given the negligible air leakage, each 153 term of the MBE can be accurately characterized.

154 The experiments – including the characterization of P, α and K from Eq. 1 and indoor/outdoor 155 measurements of size-resolved particles - were conducted over a long time period from June 156 2016 to October 2017 to encompass a complete 1-year weather cycle. The test room was kept 157 unoccupied and unfurnished during the experiments to minimize resuspension and emissions. 158 Particulate matter measurements were performed using two optical particle counters (OPC, 159 AEROTRAK 8220, Table 1), sampling air at a flow rate of 2.8 L/min. Particle number 160 concentrations were acquired for five different size bins (0.3-0.5, 0.5-1.0, 1.0-3.0, 3.0-5.0, 5.0-161 $10.0 \,\mu\text{m}$) at a time resolution of 10 s. A larger size bin (for particles larger than 10 μ m) has been 162 discarded due to very low counts. The two OPCs were cross-calibrated before, during, and after 163 the measurement campaign (see section 1 in the Supplementary Information). Collocated 164 measurements from the two OPC indicate deviations lower than 14% for all size bins. For 165 indoor/outdoor measurement periods, the two OPC were used simultaneously to record size-166 resolved C_{in} and C_{out} to test equation 1.

167 Table 1: Instrumentation information.

Instrument	Parameter	Specifications
TSI AEROTRAK Handheld Particle Counter 8220	Particle number concentration for 6 size bins (from 0.3 to >10.0 μm)	Counting efficiencies: $50\% \pm 10\%$ at 0.3μ m; 100% by 0.45μ m; $50\% \pm 20\%$ at all calibration cut sizes
Testo 480 probe	CO ₂ concentration	Range: 0 to 10,000 ppm

168

169 The spatial homogeneity of sized-resolved PM number concentrations within the test room was 170 investigated to ensure that local measurements performed at the center of the test room would be 171 representative of its whole volume, which is a prerequisite to constrain Eq. 1 with local 172 measurements of Cin. It was assessed by keeping one OPC at a fixed reference point (#1 in Figure 1) and moving the other to 13 different locations within the test room (#2-13 in Figure 1). 173 174 Each location was sampled for approximately 3 hours allowing particle counts to be 175 intercompared for each pair. The relative difference was less than 12% across the five different 176 size ranges, which is similar to differences observed when the two OPC performed collocated 177 measurements (14%), thus confirming a homogeneous PM distribution in the test room. Detailed 178 results are presented in section 2 of the SI.



179 Figure 1. 2-D and 3-D schematic diagrams of the building and positions of the OPCs.180 Measurements performed at heights of 0.60 and 1.30 m are in black and red, respectively.

181

182 **2.2. Determination of the Mass Balance Equation (MBE) parameters**

The air exchange rate was measured according to the ASTM E741-00 method⁷¹ using CO₂ (Air Liquide) as a tracer. Approximately 2,500 ppm (at the peak) of CO₂ was injected in the test room and fast mixing was ensured using a fan during injection. CO_2 was measured using a Testo 480 probe (Table 1) at a time resolution of 20 seconds during the subsequent 15 hours, which was long enough to reach its outdoor background concentration. The air exchange rate α (h⁻¹) was derived from the CO₂ temporal decay, using the slope of a linear regression between the natural logarithm of indoor CO₂ and time as shown in Equation 2:

190
$$ln[C_{CO2}(t) - C_{CO2,bkg}] = -\alpha \cdot t + ln[C_{CO2}(0) - C_{CO2,bkg}]$$
 {2}

191 where C_{CO2} and $C_{CO2,bkg}$ are the indoor air and background CO₂ mixing ratios (ppm), respectively, 192 α the air exchange rate (h⁻¹), and *t* the time (h).

The penetration factor (*P*) refers to the fraction of particles in the ambient air that passes through the building shell and ventilation systems.^{36,72,73} Since the chosen building is tightly sealed to meet energy-efficient standards, particles found indoor are assumed to be transported solely through the ventilation system. In this study, particle concentrations measured simultaneously at the entrance (i.e. outdoor) and exit (i.e. indoor) of the ventilation system were used in Equation 3 to quantify the penetration factor for each particle size bin.

199
$$P = \frac{C_{in,vent}}{C_{out}}$$
 {3}

where $C_{in, vent}$ and C_{out} are the particle number concentrations (# m⁻³) exiting the ventilation system indoors and outdoors, respectively. The initial 30 minutes of measurements were discarded to prevent contamination from resuspension while setting the instruments. It is worth noting that another existing (indirect) approach to determine penetration factors relies on using the MBE while assuming no direct emissions of particles indoors.⁷⁴ However a major caveat from this method is that it prevents any truly independent test of the MBE. Furthermore, 206 uncertainties from all other constrained parameters would propagate into the derived penetration207 factor.

208 Measurements of penetration factors were conducted twice for a duration of minimum 12 hours 209 each, at the beginning (October 2016) and toward the end (June 2017) of the measurement 210 campaign (see Supplementary Information, Table S2). For a given measurement period and 211 despite sometimes large variations of the average outdoor concentrations (from 9 to 85% 212 depending on the size fraction, likely linked to changing ambient conditions of temperature, 213 humidity as well as particle composition, mixing state and phase), the penetration factor stays quite stable without being affected by the outdoor situation (standard deviation less than 13%, 214 215 except for the larger size bin). When comparing the two periods, additional clogging of the air 216 filter as well as lower number concentrations led to higher relative standard deviations for the 217 larger size fractions. The size-dependent uncertainty associated to this parameter (variability due 218 to changes in ambient conditions and clogging of the air filters) was propagated during the 219 calculation of relative imbalance values. It is worth noting however that the penetration factor 220 showed stability over time for the smaller size bins corresponding to PM₁, which nonetheless 221 exhibit the strongest relative imbalance.

Size-resolved deposition rates (K) were derived after triggering a sharp increase in aerosol concentrations (e.g. by vacuuming) and following its temporal decay, using the slopes of a linear regression between the natural logarithm of particle number concentrations and time, subtracting the air exchange rate (Equation 4).³⁷

226
$$\ln[C_{in}(t) - C_{in,bkg}] = -(\alpha + K) \cdot t + \ln[C_{in}(0) - C_{in,bkg}]$$
 {4}

where C_{in} and $C_{in,bkg}$ are the particle indoor and background concentrations (# m⁻³), respectively, 227 228 α the air exchange rate (h⁻¹) and t the time (h). Given the complexity of generating large amounts 229 of particles within the whole volume of the room in all the relevant size bins, several criteria were 230 taken into consideration to ensure an accurate quantification of size-resolved PM deposition 231 rates: (i) a peak of particle number concentration during resuspension at least twice higher than 232 the background concentration for each size bin; (ii) a steady background, with a peak-to-peak 233 background fluctuation lower than 10% of the concentration enhancement during resuspension for each size bin; (iii) a determination coefficient (r^2) higher than 0.9 for the linear regression 234 235 analysis (Eq. 4). Details on the deposition rate experiments are given in the SI.

236

2.3. Size-resolved investigation of the aerosol budget indoors

237 Simultaneous indoor and outdoor particle measurements were conducted on 24-hour periods 238 which were repeated 25 times during the campaign to investigate the aerosol budget indoors, that 239 is to say to characterize both sources and sinks and investigate mass closure under steady state 240 conditions. The dataset was screened for short-time windows (at least 1.5 h), characterized by 241 steady outdoor particle concentrations. A time window was considered suitable when the relative 242 standard deviation (RSD) of the measurements was less than 35%. This threshold was chosen to 243 ensure a statistically significant number of time windows for a robust analysis. Calculations using 244 other RSD thresholds (< 35%) do not present improved results (Figure S3). Those short-time 245 windows allowed the application of the MBE, which otherwise would require to integrate the 246 measurements over significantly longer time-scales (days, weeks) to use the steady state assumption.⁷⁵ Overall, 23 short-time windows (on 17 different days) were identified. The 247 parameter Cout was derived by averaging the outdoor concentration over the time windows while 248 249 Cin was averaged from the subsequent half hour (due to outdoor-indoor lag time discussed in

section 4 of the SI). All the other parameters required in Equation 1 were taken from the characterization results described previously (α , P, K) and the geometry of the room (volume V). Negligible resuspension (unoccupied room) and no direct emission or formation (empty room, no human activities) were assumed for the MBE calculation as previously discussed.

254 **3.** Results

255 **3.1.** Size-resolved assessment of the MBE parameters

256 The average air exchange rate in the test room was 0.54 ± 0.05 h⁻¹ (out of 9 experiments using 257 CO₂ time decays, see an example in section 5 of the SI). The values reported in the literature for different types of buildings typically range from 0.12-3.5 h⁻¹ under mechanically-ventilated 258 conditions, reaching up to 7.9 h⁻¹ when air exchange is forced by the opening of windows and 259 doors^{23,76,77}. Our results show that air exchange is reasonably low, which is a feature of energy-260 261 efficient buildings where heat loss is also minimized through reduced air exchange. In addition, the air exchange rate calculated from the ventilation unit flow rate of 17 m³ h⁻¹ (value provided 262 by the manufacturer) and the room volume of 29.2 m³ leads to a value of 0.58 h⁻¹ (see section 5 263 264 of the SI). This value is in good agreement and falls within the uncertainties of the measured 265 AER of 0.54 ± 0.05 h⁻¹. This excellent agreement (i) confirms the air flow rate of the ventilation 266 unit is stable over time, and (ii) supports our assumption to neglect air leakages.

Results for the penetration factor are shown in Figure 2, depicting a significant decrease with increasing size (> 0.8 for PM₁ down to less than 0.2 for the coarse fraction). This trend, in agreement with the literature,^{34,36,37,78,79} is expected since penetration depends on the particle size, with fine particles (< 2.5 μ m) typically penetrating more easily than coarse (> 2.5 μ m) ones. Other parameters influencing penetration factors are the chemical composition, phase state and 272 mixing state of particles,^{36,72,73} as well as the nature, thickness and shape of the filter, which may 273 explain some of the variability observed in the literature for each size bin (grey area in Figure 2).



Figure 2: Average values of penetration factors (open markers) as a function of particle size. Error bars represent $\pm 1\sigma$ (n = 235). The shaded area represents the range of values obtained in previous studies.⁷²

Deposition rates were found to range from 0.2 h⁻¹ up to 1.2 h⁻¹, as shown in Figure 3 and section 274 6 of the SI. Concentration levels reached upper values of 2.0×10^8 and 2.9×10^5 particles m⁻³ for 275 276 the smallest and largest size bins, respectively and each experiment lasted for 5-6 hours. The lowest deposition rate was observed for 1.0-3.0 µm particles, and values reported for the different 277 278 size bin in this study are in agreement with previously reported deposition rates, although large 279 variability exists in the literature depending on room occupancy, air flow, aerosol type and building materials.^{78,80} From Figure 3, and considering an air exchange rate of 0.5 h⁻¹, deposition 280 281 represents approximately one third, half and the total sink for particles smaller than 3.0 µm, 282 ranging from $3.0-5.0 \,\mu$ m, and larger than $5 \,\mu$ m, respectively.



Figure 3: Average values of deposition rates (open markers) as a function of particle size. Error bars represent $\pm 1\sigma$ (9-15 measurements). The shaded area represents the range of values obtained in previous studies.^{37,78}

During deposition rate experiments, a three-fold increase was observed for submicron aerosols when ambient temperatures increased from 22°C to 29°C (Figure S7), a result that requires further analysis and, to the best of our knowledge, was not previously reported, although the influence of temperature gradients has been also suggested by Abt et al. (2000).⁸⁰

287 **3.2.** Indoor and outdoor particle concentrations

In Europe, the European Environmental Agency (EEA) and the World Health Organization (WHO)^{81,82} have regulated the PM₁₀ and PM_{2.5} fractions in ambient air, thus the mass concentrations of PM₁₀, PM_{2.5} (considered as particles with optical diameter below 3.0 μ m for this study) and PM₁ were calculated assuming a spherical shape and an aerosol density of 1.4 g cm⁻³.⁸³ An example of indoor and outdoor particle concentrations is given in Figure 4 together with a time window selected for the mass budget analysis. The time series show that outdoor concentrations are highly variable due to the proximity of emission sources, while the indoor 295 concentration is buffered due to the slow air exchange rate and the absence of primary indoor 296 sources.



Figure 4: Example of time series of 6-minute averaged particle concentrations for the different particle size fractions on 29-30 June 2017 measured indoor (filled) and outdoor (open circles). A

time window selected for mass budget analysis following the criteria described in subsection 2.3 ishighlighted in pink.

301 4. Discussion

302 4.1. Impact of infiltration on indoor particle concentration

303 Based on the air exchange rate and the size-resolved penetration factors and deposition rates, the 304 infiltration factor, i.e. the fraction of outdoor particles that penetrates and remains suspended 305 indoors, is defined as:

$$F_{\inf} = \frac{\alpha P}{\alpha + K}$$
 {5}

and can be calculated considering an unoccupied room (particle sources and sinks only related to air exchange and deposition). The indoor concentration of the finer fraction (0.3-0.5 μ m) would reach approximately 50% of the outdoor concentration, whereas for the 1.0-3.0 μ m and 5.0-10.0 μ m size bins this value would decrease to 43% and 4%, respectively. Such a marked decrease is consistent with a combined effect of penetration factors decreasing with particle size and the opposite behavior for deposition rates.

Actually, given that short-time windows were chosen under steady-state conditions, indoor particle concentrations are always expected to be smaller than outdoor ones. While this was observed to be true for particles larger than $1.0 \,\mu$ m, indoor PM₁ concentrations were sometimes similar to or even higher than outdoor ones, suggesting that processes other than penetration and deposition are required to estimate concentrations of submicron particles in this unoccupied 318 room. The validity of the MBE for different particle sizes, including submicron particles, and 319 under different environmental conditions was therefore evaluated below.

320 4.2. Evaluation of the relative imbalance for PM₁₀, PM_{2.5} and PM₁

321 Under steady-state conditions in an unoccupied and empty room, the source and sink terms of the 322 MBE (Equation 1) should cancel each other out and PM concentrations should only depend on 323 the introduction of particles from outdoors (α PVC_{out}) and their loss through a combined effect of 324 air exchange and deposition ((α +K)VC_{in}). In order to check this aspect, we define the Relative 325 Imbalance (RI) as the difference between source and sink rates, normalized to the source rate:

326
$$RI = \frac{\alpha PVC_{out} - (\alpha + K)VC_{in}}{\alpha PVC_{out}}$$
 {6}

Figure 5 depicts the averaged relative imbalance for the three PM fractions calculated for the 23 short-time windows during the campaign, with a distinction between warm ($T_{in} > 28$ °C, eight time-windows) and cold ($T_{in} < 22$ °C, 15 time-windows) periods. This distinction has been arbitrarily done based on the based on the observation of different results (in terms of RI) during these two periods.



Figure 5: Averaged relative imbalance (RI; see equation 6) for the PM₁, PM_{2.5} and PM₁₀ size fractions (from left to right) considering the campaign average, warm periods only ($T_{in} > 28 \ ^{\circ}C$) and cold periods only ($T_{in} < 22 \ ^{\circ}C$). Error bars represent 1 σ .

332 This figure shows that indoor PM₁₀ and PM_{2.5} concentrations are reasonably well estimated on a 333 campaign average when both size-resolved penetration factors and deposition rates are correctly 334 characterized (mean RI < 18%), whereas a larger variability is observed for RI on PM₁. A close 335 look at the RI temporal variability for the three PM fractions indicates an important impact of 336 temperature (Figures 5 and S8), with larger positive RI values at higher temperatures. This is also 337 clearly depicted in Figure S8 (panel b), where RI values observed for each PM fraction during the 338 warm period are larger than the means+1 σ observed during the cold period (shaded area). These 339 positive RI values indicate additional sources of PM in the building during the warm period, 340 which are not observed during the cold period. It is interesting to note that the only parameter of 341 the MBE that showed some temporal (or temperature) dependence was the deposition rate (more 342 details in the SI), which was considered in this analysis. Not accounting for the larger deposition rates of PM₁ at higher temperatures would lead to even larger RI values for this PM fraction. 343

344 We speculate that different physicochemical processes, not accounted for in the mass balance analysis performed above, are affecting the PM fractions. One assumption made in the above 345 analysis, similarly to other studies,^{42,60} is the negligible impact of resuspension on PM loading in 346 347 an unoccupied room. When temperature increases and relative humidity decreases, adhesive 348 forces between particles and surfaces become looser, thus an increase of resuspension rates can 349 impact the ambient PM concentrations. While this assumption may hold for the cold period, 350 higher ambient temperatures and lower relative humidity, can lead to an increase of resuspension rates on hard floors (such as in this test room)⁸⁴ and, as a consequence, a significant impact of 351 352 resuspension on ambient PM concentrations. Interestingly, previous studies have shown that the resuspension rate increases with particle size,^{31,32,38,84-86} indicating that resuspension rates for 353 354 PM₁ are orders of magnitude lower than for PM_{2.5} and PM₁₀. As a consequence, while an increase of resuspension rates with temperature could explain the RI observed for PM_{2.5} and PM₁₀, 355 356 resuspension alone is unlikely to explain the larger RI observed for PM₁.

357 Additional physicochemical processes such as gas-to-particle conversion (condensation of low-358 volatility gases) for particles smaller than 0.3 µm (lowest size measurable with the OPC) or the reverse (volatilization of NH₄NO₃ or water soluble and semi-volatile organics)^{28,87–91} for particles 359 360 larger than 1 µm could also impact the PM₁ fraction. On one side, building materials covering 361 large surface areas in the building, mainly wood and linoleum, are known to emit monoterpenes and SVOCs, respectively.⁹² Monoterpenes and other unsaturated VOCs are rapidly oxidized by 362 363 ozone, which in turn lead to the formation of oxygenated VOCs (OVOCs) of lower volatility. A 364 subsequent winter field campaign performed in the same test room (whose results are beyond the scope of this publication but will be presented in a forthcoming paper) has shown that the total 365 concentration of the 60 measured OVOCs was at least eight times higher indoors (average C_{in} = 366

146.6 μ g m⁻³) compared to outdoors (average C_{out} = 18.3 μ g m⁻³), which supports that low-367 368 volatility species could interact with the submicron aerosol population and could lead to a 369 modification of their size distribution. Besides, the emission of condensable gases from building 370 materials and gas-phase oxidation reactions are expected to become more significant at higher 371 temperatures (higher emission rates, higher ozone concentrations). On the other side, 372 volatilization of ammonium nitrate, leading to the formation of ammonia and nitric acid, has 373 already been observed indoors, with volatilization rates depending on temperature, relative humidity and ventilation rate.²⁸ Both physicochemical processes (condensation and volatilization) 374 375 will be strongly linked to gas-particle equilibrium and temperature differences between indoors 376 and outdoors.

377 While the above discussion remains speculative, the results from this study indicate that 378 assuming no resuspension in an empty room seems to be a valid assumption for the use of the 379 mass balance model as long as ambient temperature remains under moderate values (< 22°C in 380 this study). Under these conditions, PM_x (x = 1, 2.5, 10) concentrations indoors can be accurately 381 assessed from outdoor measurements, providing that air exchange and deposition are well 382 characterized. However, this assumption may break down for higher temperatures, especially for 383 PM_{2.5} and PM₁₀. In addition, the larger imbalance observed for PM₁ indicates that this PM 384 fraction is significantly more impacted by aerosol dynamics and thus requires a more detailed 385 knowledge of the complete aerosol size distribution, its physicochemical characteristics, and 386 physicochemical processes occurring indoors.

387 **5.** Conclusions

388 Particulate matter (PM) concentration indoors is the result of a balance between sources and 389 sinks, including potential physicochemical transformations, and a good understanding of these 390 processes is needed to correctly assess PM budget indoors. During this study the air exchange 391 rate, the penetration factor and deposition rates of size-resolved PM were measured in an 392 unoccupied energy-efficient building as defined by the French RT2012 regulation. Those parameters were used to constrain a MBE linking indoor concentrations of PM to outdoor ones. It 393 394 was found that indoor concentrations of PM_x (x = 1, 2.5, 10) can be well estimated by the MBE 395 based on measured outdoor concentrations, as long as temperature does not exceed a certain 396 threshold of ambient temperatures (around 22°C in this study). In contrast, the indoor PM budget 397 was significantly less accurate, with measured indoor concentrations being up to four times 398 higher than values calculated from the mass balance approach for lower temperatures. This 399 disagreement is attributed on the one hand to the co-variability between processes and 400 transformations (gas-to-particle conversion, condensation, evaporation) taking place indoors for 401 the PM₁ fraction, and on the other hand to the assumed negligible resuspension mainly of coarse 402 particles (PM_{2.5} and PM₁₀) even in a relatively source-free indoor environment.

The results of our study point out the limitations of a simplified mass balance analysis to estimate PM fractions indoors, as can be classically found in the literature. It is usually assumed that under unoccupied conditions the particles do not undergo transformations when they penetrate indoors. Even under conditions where all the terms of the MBE have been carefully characterized and air leakage is strongly minimized, such as in this work, this assumption leads to a miscalculation of particle concentrations indoors during warm periods, whereas it seems to be accurate enough (within uncertainties) for cold ones. Such results suggest that (i) the approximation of indoor 410 species as inert and (ii) the assumption of negligible resuspension are not valid depending on the 411 meteorological conditions, and an accurate budget analysis requires significantly more 412 sophisticated methodologies including a detailed chemical composition characterization of the 413 gas and particulate phases, even in a well-characterized and unoccupied environment.

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Energy Efficient Building





Deposition / Resuspension

Air exchange