Investigation of PM10, PM2.5, PM1 in an unoccupied airflow-controlled room: How reliable to neglect resuspension and assume unreactive particles?

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Abstract

It is now well recognized that Particulate Matter (PM) is one of the main air pollutants affecting both ambient and indoor air quality. While ambient PM mass concentration measurements are often performed by air quality monitoring networks, current regulations do not address their indoor concentrations. The latter can be estimated nonetheless from a mass balance analysis accounting for (i) the transfer of particles from outdoor and (ii) their typical indoor sources (emissions, resuspension) and sinks (deposition, removal by air exchange). Inherently, the mass 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 imbalance (RI) is not significant over different size fractions, a series of careful measurements were conducted in an unoccupied room within a building incorporating an energy-efficient design (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-resolved particles, the PM_{10} and $PM_{2.5}$ fractions, which are typically regulated outdoors, as well as PM_1, can be well estimated indoors (with RI between measured and expected values < 19%) under moderate ambient temperatures (< 22°C in this study). However, RI increases significantly, especially for submicron particles, at higher temperatures, indicating possible transformations in the particulate phase, which are not accounted for by the mass balance model. Therefore caution is recommended regarding the mass balance analysis to estimate PM fractions indoors, especially for PM_1.

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

1. Introduction

The infiltration of outdoor pollution combined with a range of indoor sources emitting volatile 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 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 exposure to harmful species requires a good assessment of pollutant concentrations in the indoor environment, which in turn requires a good understanding of their sources, their sinks and all the physicochemical processes leading to their transformations.

Among atmospheric pollutants, PM has been classified as carcinogenic for humans,[3] and several countries have set air quality standards on ambient mass concentrations of PM with an aerodynamic diameter below 10 µm (PM_{10}) or 2.5 µm ($PM_{2.5}$, also known as fine particles). PM of even smaller sizes, e.g. with a diameter lower than 1 µm (PM_1), is of particular interest since it deposits more easily in the lower respiratory tract where it can penetrate into the circulatory system, impacting other organs.[4,5] Furthermore, PM_1 are observed at high number concentrations compared to larger particles, leading to large surface areas that can carry adsorbed organic pollutants to the circulatory system.[6]

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]

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

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

where C_{in} and C_{out} are the indoor and outdoor concentrations (μg m^{-3}), respectively, V the volume of the test room (m^3), 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 μm. 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

78 mentioned above. The accuracy associated to indoor budgets of PM_x (x = 1, 2.5, 10) derived from
79 Equation 1 depends on how well each term can be characterized, which is extremely challenging
80 given their dependence on the particle size and their co-variability.[33]

81 The mass balance equation (MBE) has been previously used in various studies to (i) estimate
82 indoor concentrations of particles based on observed outdoor concentrations, (ii) quantify some
83 parameters such as particle penetration, deposition, and resuspension rates,[27,30–32,34–55] and (iii)
84 estimate the strength of indoor particle sources resulting from human activities.[46,47,56–62] In most
85 of these studies however, two important simplifications are usually made: (i) particles are inert
86 species and their number concentration and size does not change and (ii) particle resuspension is
87 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
90 conditions. Interestingly, Abt et al.,[47] performed measurements of continuous particle size and
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 μm 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 μm) 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
100 observed in some studies.[43,57,59,63–66] For example Polidori et al., 2006 [59] showed that 71-76% of

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 Nagendra[68] also performed measurements of particulate matter
108 indoors and outdoors ($PM_{2.5}$ in homes, and PM_1, $PM_{2.5}$, PM_{10} 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 $PM_{2.5}$
114 concentrations indoors when the human activities are well identified and characterized. However,
115 the latter showed thatPM_1 and PM_{10} concentrations can be wrongly estimated by not including
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.

119 The objective of this work is to investigate whether the mass balance model described in
120 Equation 1, assuming in a first step unreactive species (no particle processing) and negligible
121 resuspension under unoccupied conditions (anthropogenic influence is negligible), could
122 accurately predict indoor concentrations of PM_x (x = 1, 2.5, 10) if the parameters in this equation
123 were accurately quantified for each PM size fraction, or whether physicochemical

transformations and resuspension need to be included in the model, unlike to what has been mainly considered until recently. In this work, we first characterized each term of the mass balance model (Eq. 1) for an empty room of a low-energy building (described in Sections 2.2 and 3.1), and we then used the MBE to investigate how indoor measurements of PM_x compare to values inferred from the model constrained by particulate outdoor measurements (Sections 2.3 and 4). Subsequently, we investigate whether this approach can highlight physical and chemical processes occurring in the room, and therefore if the equation can be applied directly in more complex situations, as usually assumed.

2. Materials and methods

2.1. Building and campaign description

The measurement facility is located at the Institute Mines Telecom Lille Douai, on the Douai Campus, in northern France. This facility is a wooden building, a so-called energy-efficient building with the BBC label ("Bâtiment Basse Consommation"), which according to the French 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 constructions aiming at significantly reducing the carbon footprint from heating and hot water production, and therefore represents a trend for future constructions.[70] The building is composed of two similar rooms of approximately 12 m^2 of surface area and 2.4 m height, leading to a surface to volume ratio of 0.4 m^2/m^3. Only one room (hereafter called "test room") was used to perform all the characterization experiments (quantification of the parameters shown in Eq. 1) and the simultaneous indoor/outdoor particle measurements, with no air exchange between the 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 m^3 h^{-1}. This system includes a G4 filter for the efficient removal of coarse particles. Ventilation was kept on during the whole measurement period. In order to achieve the standards of energy-efficient buildings, the building is equipped with thermally efficient windows (triple-glazed with Argon filling and an aluminum frame) and the walls and roof are composed of wood, and insulated using wood wool and fibers. The floor is covered by linoleum on top of wood wool and an OSB board. The external door is made of 33-mm thick aluminum profiles and a triple-glazed window. Given the negligible air leakage, each term of the MBE can be accurately characterized.

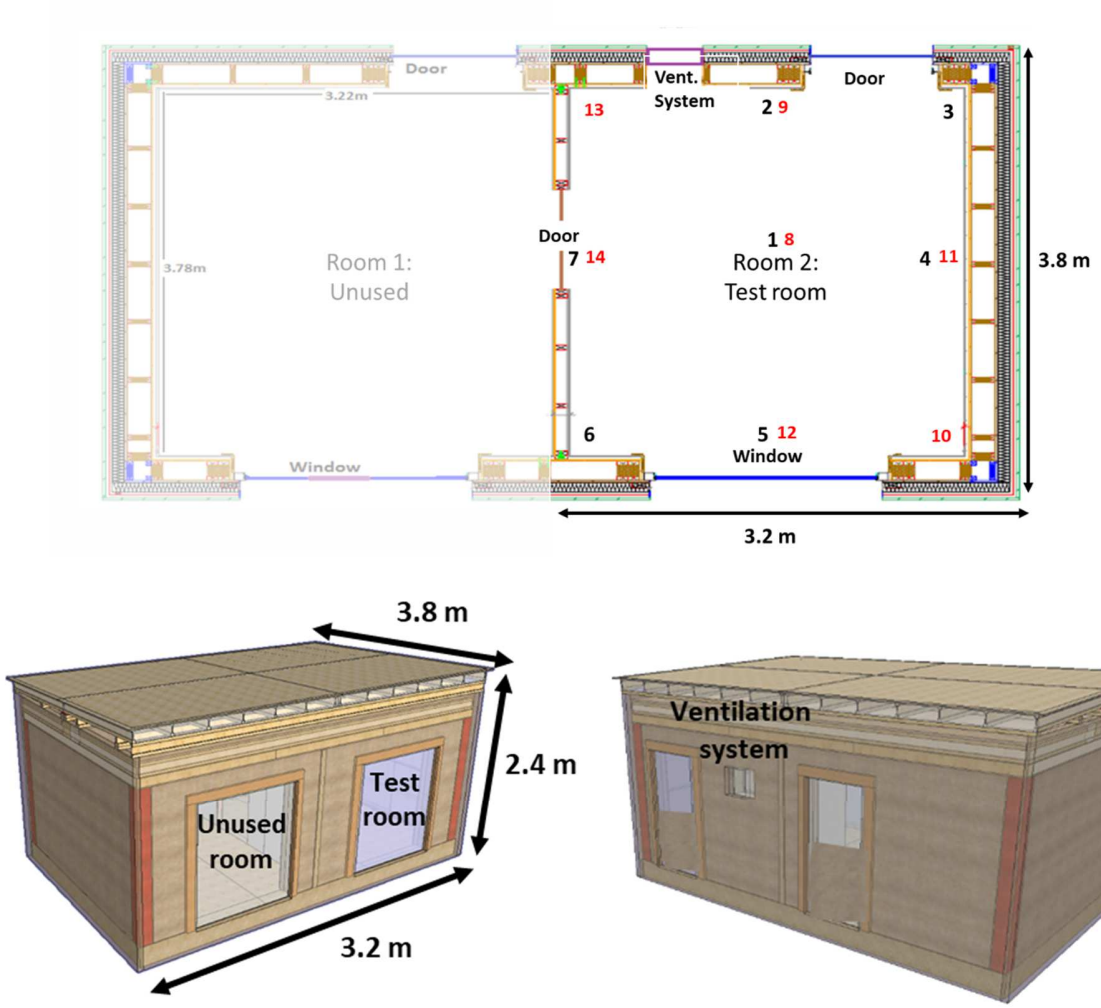
The experiments – including the characterization of P, α and K from Eq. 1 and indoor/outdoor measurements of size-resolved particles – were conducted over a long time period from June 2016 to October 2017 to encompass a complete 1-year weather cycle. The test room was kept unoccupied and unfurnished during the experiments to minimize resuspension and emissions. Particulate matter measurements were performed using two optical particle counters (OPC, AEROTRAK 8220, Table 1), sampling air at a flow rate of 2.8 L/min. Particle number concentrations were acquired for five different size bins (0.3-0.5, 0.5-1.0, 1.0-3.0, 3.0-5.0, 5.0-10.0 μm) at a time resolution of 10 s. A larger size bin (for particles larger than 10 μm) has been discarded due to very low counts. The two OPCs were cross-calibrated before, during, and after the measurement campaign (see section 1 in the Supplementary Information). Collocated measurements from the two OPC indicate deviations lower than 14% for all size bins. For indoor/outdoor measurement periods, the two OPC were used simultaneously to record size-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% ± 10% at 0.3µm; 100% by 0.45µm; 50% ± 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 C_{in}. It was assessed by keeping one OPC at a fixed reference point (#1 in
173 Figure 1) and moving the other to 13 different locations within the test room (#2-13 in Figure 1).
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**

183 The air exchange rate was measured according to the ASTM E741-00 method[71] using CO_2 (Air
184 Liquide) as a tracer. Approximately 2,500 ppm (at the peak) of CO_2 was injected in the test room
185 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 CO_2 temporal decay, using the slope of a linear regression between the natural logarithm of indoor CO_2 and time as shown in Equation 2:

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

where C_{CO2} and $C_{CO2,bkg}$ are the indoor air and background CO_2 mixing ratios (ppm), respectively, 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.

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

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,

uncertainties from all other constrained parameters would propagate into the derived penetration factor.

Measurements of penetration factors were conducted twice for a duration of minimum 12 hours each, at the beginning (October 2016) and toward the end (June 2017) of the measurement campaign (see Supplementary Information, Table S2). For a given measurement period and despite sometimes large variations of the average outdoor concentrations (from 9 to 85% depending on the size fraction, likely linked to changing ambient conditions of temperature, 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%, except for the larger size bin). When comparing the two periods, additional clogging of the air filter as well as lower number concentrations led to higher relative standard deviations for the larger size fractions. The size-dependent uncertainty associated to this parameter (variability due to changes in ambient conditions and clogging of the air filters) was propagated during the calculation of relative imbalance values. It is worth noting however that the penetration factor showed stability over time for the smaller size bins corresponding to PM_1, which nonetheless 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]

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

where C_{in} and $C_{in,bkg}$ are the particle indoor and background concentrations (# m^{-3}), respectively, α the air exchange rate (h^{-1}) and t the time (h). Given the complexity of generating large amounts of particles within the whole volume of the room in all the relevant size bins, several criteria were taken into consideration to ensure an accurate quantification of size-resolved PM deposition rates: (i) a peak of particle number concentration during resuspension at least twice higher than the background concentration for each size bin; (ii) a steady background, with a peak-to-peak 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 analysis (Eq. 4). Details on the deposition rate experiments are given in the SI.

2.3. Size-resolved investigation of the aerosol budget indoors

Simultaneous indoor and outdoor particle measurements were conducted on 24-hour periods which were repeated 25 times during the campaign to investigate the aerosol budget indoors, that is to say to characterize both sources and sinks and investigate mass closure under steady state conditions. The dataset was screened for short-time windows (at least 1.5 h), characterized by steady outdoor particle concentrations. A time window was considered suitable when the relative standard deviation (RSD) of the measurements was less than 35%. This threshold was chosen to ensure a statistically significant number of time windows for a robust analysis. Calculations using other RSD thresholds (< 35%) do not present improved results (Figure S3). Those short-time windows allowed the application of the MBE, which otherwise would require to integrate the 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 parameter C_{out} was derived by averaging the outdoor concentration over the time windows while C_{in} was averaged from the subsequent half hour (due to outdoor-indoor lag time discussed in

250 section 4 of the SI). All the other parameters required in Equation 1 were taken from the
251 characterization results described previously (α, P, K) and the geometry of the room (volume V).
252 Negligible resuspension (unoccupied room) and no direct emission or formation (empty room, no
253 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 CO_2 time decays, see an example in section 5 of the SI). The values reported in the literature for
258 different types of buildings typically range from 0.12-3.5 h^{-1} under mechanically-ventilated
259 conditions, reaching up to 7.9 h^{-1} when air exchange is forced by the opening of windows and
260 doors[23,76,77]. Our results show that air exchange is reasonably low, which is a feature of energy-
261 efficient buildings where heat loss is also minimized through reduced air exchange. In addition,
262 the air exchange rate calculated from the ventilation unit flow rate of 17 m^3 h^{-1} (value provided
263 by the manufacturer) and the room volume of 29.2 m^3 leads to a value of 0.58 h^{-1} (see section 5
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.

267 Results for the penetration factor are shown in Figure 2, depicting a significant decrease with
268 increasing size (> 0.8 for PM_1 down to less than 0.2 for the coarse fraction). This trend, in
269 agreement with the literature,[34,36,37,78,79] is expected since penetration depends on the particle size,
270 with fine particles (< 2.5 µm) typically penetrating more easily than coarse (> 2.5 µm) ones.
271 Other parameters influencing penetration factors are the chemical composition, phase state and

mixing state of particles,[36,72,73] as well as the nature, thickness and shape of the filter, which may explain some of the variability observed in the literature for each size bin (grey area in Figure 2).

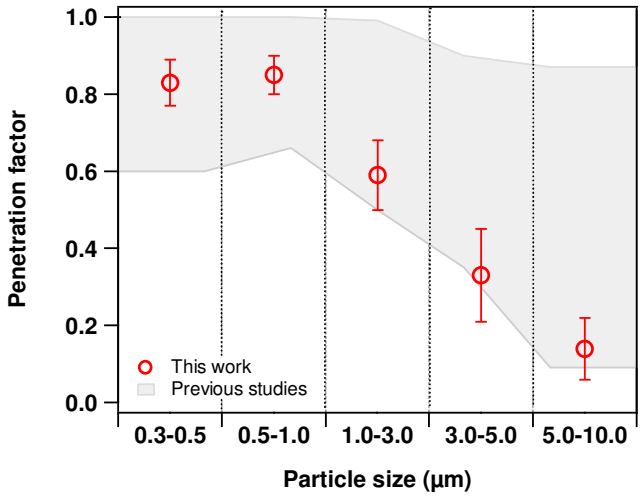


Figure 2: Average values of penetration factors (open markers) as a function of particle size. Error bars represent ± 1σ (n = 235). The shaded area represents the range of values obtained in previous studies.[72]

Deposition rates were found to range from 0.2 h^{-1} up to 1.2 h^{-1}, as shown in Figure 3 and section 6 of the SI. Concentration levels reached upper values of 2.0×10^8 and 2.9×10^5 particles m^{-3} for 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 size bin in this study are in agreement with previously reported deposition rates, although large 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 represents approximately one third, half and the total sink for particles smaller than 3.0 µm, ranging from 3.0-5.0 µm, and larger than 5 µm, respectively.

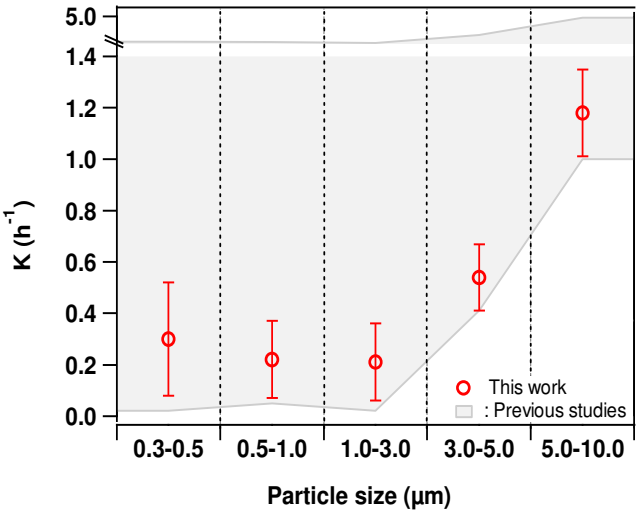


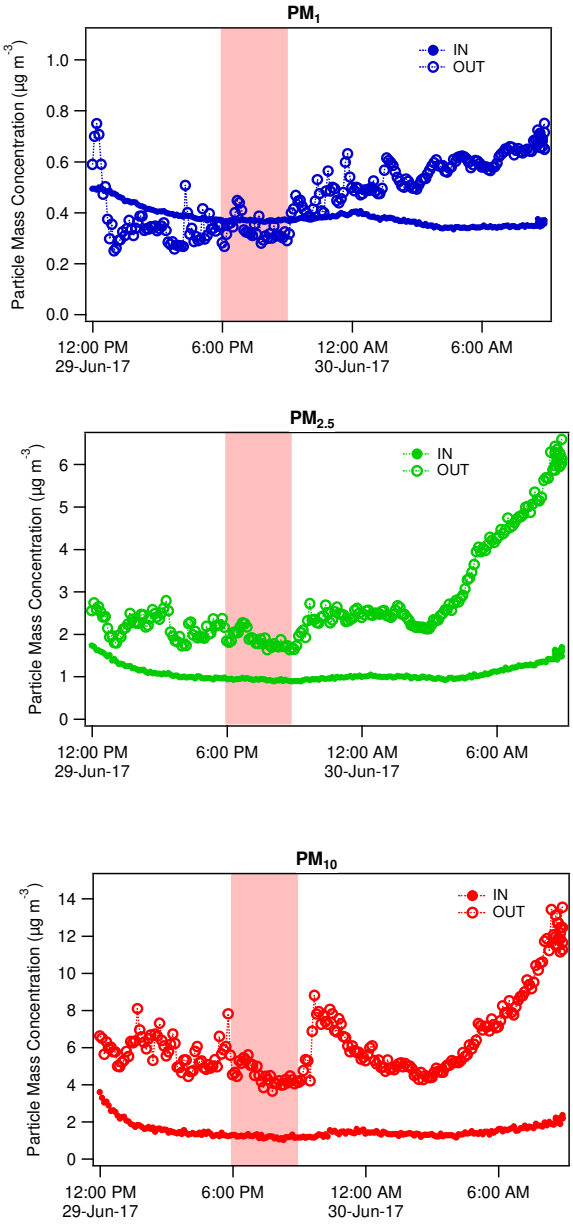
Figure 3: Average values of deposition rates (open markers) as a function of particle size. Error bars represent ± 1σ (9-15 measurements). The shaded area represents the range of values obtained in previous studies.[37,78]

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

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

288 In Europe, the European Environmental Agency (EEA) and the World Health Organization
289 (WHO)[81,82] have regulated the PM_{10} and $PM_{2.5}$ fractions in ambient air, thus the mass
290 concentrations of PM_{10}, $PM_{2.5}$ (considered as particles with optical diameter below 3.0 µm for
291 this study) and PM_1 were calculated assuming a spherical shape and an aerosol density of 1.4 g
292 cm^{-3}.[83] An example of indoor and outdoor particle concentrations is given in Figure 4 together
293 with a time window selected for the mass budget analysis. The time series show that outdoor
294 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.



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

299 time window selected for mass budget analysis following the criteria described in subsection 2.3 is
300 highlighted 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:

306 $$F_{inf} = \frac{\alpha P}{\alpha + K} \quad \{5\}$$

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

313 Actually, given that short-time windows were chosen under steady-state conditions, indoor
314 particle concentrations are always expected to be smaller than outdoor ones. While this was
315 observed to be true for particles larger than 1.0 μm, indoor PM_1 concentrations were sometimes
316 similar to or even higher than outdoor ones, suggesting that processes other than penetration and
317 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 PM_{10}, $PM_{2.5}$ and PM_1**

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 (αPVC_{out}) and their loss through a combined effect of
324 air exchange and deposition ($(\alpha+K)VC_{in}$). 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 $$RI = \frac{\alpha PVC_{out} - (\alpha+K)VC_{in}}{\alpha PVC_{out}} \quad \{6\}$$

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

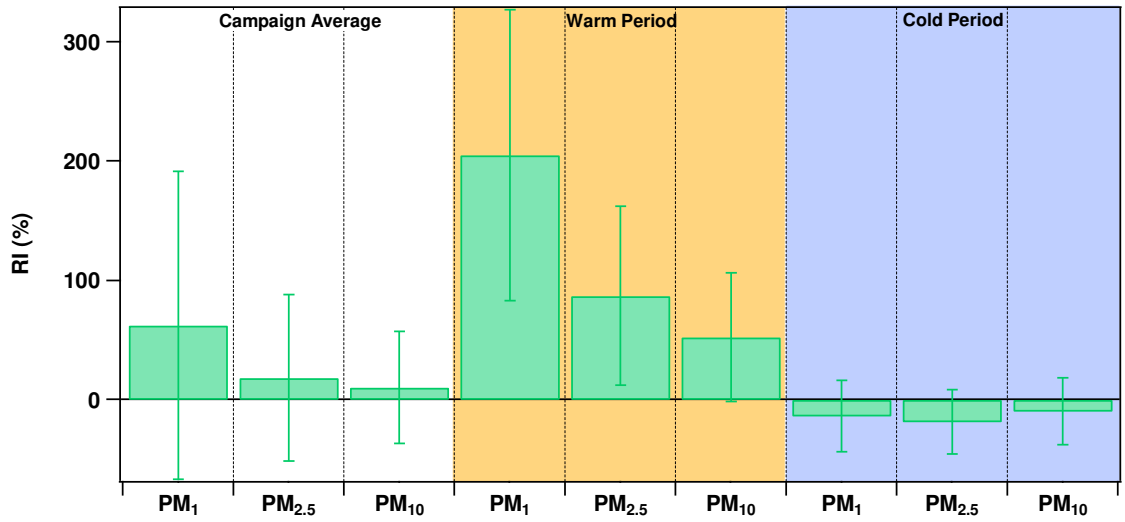


Figure 5: Averaged relative imbalance (RI; see equation 6) for the PM_1, $PM_{2.5}$ and PM_{10} size fractions (from left to right) considering the campaign average, warm periods only (T_{in} > 28 °C) and cold periods only (T_{in} < 22 °C). Error bars represent 1σ.

This figure shows that indoor PM_{10} and $PM_{2.5}$ concentrations are reasonably well estimated on a campaign average when both size-resolved penetration factors and deposition rates are correctly characterized (mean RI < 18%), whereas a larger variability is observed for RI on PM_1. A close look at the RI temporal variability for the three PM fractions indicates an important impact of temperature (Figures 5 and S8), with larger positive RI values at higher temperatures. This is also clearly depicted in Figure S8 (panel b), where RI values observed for each PM fraction during the warm period are larger than the means+1σ observed during the cold period (shaded area). These positive RI values indicate additional sources of PM in the building during the warm period, which are not observed during the cold period. It is interesting to note that the only parameter of the MBE that showed some temporal (or temperature) dependence was the deposition rate (more details in the SI), which was considered in this analysis. Not accounting for the larger deposition rates of PM_1 at higher temperatures would lead to even larger RI values for this PM fraction.

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 analysis, similarly to other studies,[42,60] is the negligible impact of resuspension on PM loading in an unoccupied room. When temperature increases and relative humidity decreases, adhesive forces between particles and surfaces become looser, thus an increase of resuspension rates can impact the ambient PM concentrations. While this assumption may hold for the cold period, 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 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 PM_1 are orders of magnitude lower than for $PM_{2.5}$ and PM_{10}. As a consequence, while an increase of resuspension rates with temperature could explain the RI observed for $PM_{2.5}$ and PM_{10}, resuspension alone is unlikely to explain the larger RI observed for PM_1.

Additional physicochemical processes such as gas-to-particle conversion (condensation of low-volatility gases) for particles smaller than 0.3 µm (lowest size measurable with the OPC) or the reverse (volatilization of NH_4NO_3 or water soluble and semi-volatile organics)[28,87–91] for particles larger than 1 µm could also impact the PM_1 fraction. On one side, building materials covering 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 ozone, which in turn lead to the formation of oxygenated VOCs (OVOCs) of lower volatility. A 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 concentration of the 60 measured OVOCs was at least eight times higher indoors (average C_{in} =

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

While the above discussion remains speculative, the results from this study indicate that assuming no resuspension in an empty room seems to be a valid assumption for the use of the mass balance model as long as ambient temperature remains under moderate values (< 22°C in this study). Under these conditions, PM_x (x = 1, 2.5, 10) concentrations indoors can be accurately assessed from outdoor measurements, providing that air exchange and deposition are well characterized. However, this assumption may break down for higher temperatures, especially for $PM_{2.5}$ and PM_{10}. In addition, the larger imbalance observed for PM_1 indicates that this PM fraction is significantly more impacted by aerosol dynamics and thus requires a more detailed knowledge of the complete aerosol size distribution, its physicochemical characteristics, and physicochemical processes occurring indoors.

5. Conclusions

Particulate matter (PM) concentration indoors is the result of a balance between sources and sinks, including potential physicochemical transformations, and a good understanding of these processes is needed to correctly assess PM budget indoors. During this study the air exchange rate, the penetration factor and deposition rates of size-resolved PM were measured in an 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 was found that indoor concentrations of PM_x (x = 1, 2.5, 10) can be well estimated by the MBE based on measured outdoor concentrations, as long as temperature does not exceed a certain threshold of ambient temperatures (around 22°C in this study). In contrast, the indoor PM budget was significantly less accurate, with measured indoor concentrations being up to four times higher than values calculated from the mass balance approach for lower temperatures. This disagreement is attributed on the one hand to the co-variability between processes and transformations (gas-to-particle conversion, condensation, evaporation) taking place indoors for the PM_1 fraction, and on the other hand to the assumed negligible resuspension mainly of coarse particles ($PM_{2.5}$ and PM_{10}) 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

species as inert and (ii) the assumption of negligible resuspension are not valid depending on the meteorological conditions, and an accurate budget analysis requires significantly more sophisticated methodologies including a detailed chemical composition characterization of the gas and particulate phases, even in a well-characterized and unoccupied environment.

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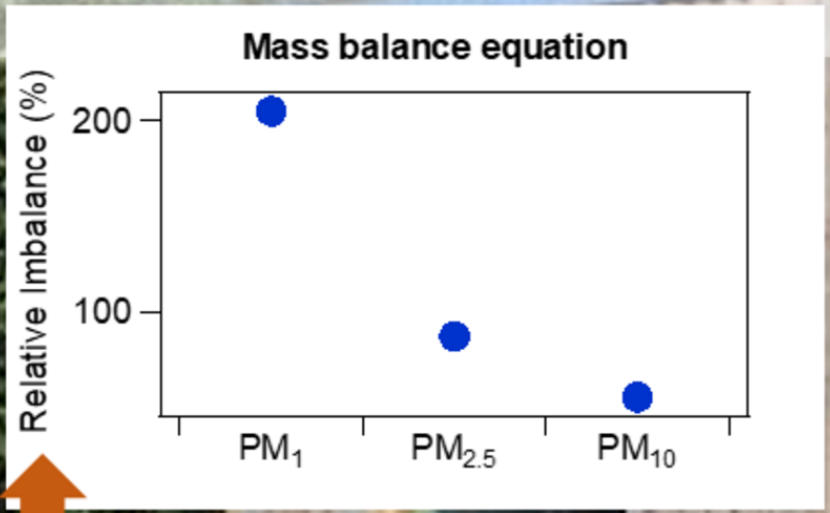
Energy Efficient Building



Deposition / Resuspension

Air exchange

Penetration