Airborne Particulate Matter: An Investigation of Buildings with Passive House Technology in Hungary

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Abstract

In this case study, we investigate the building infiltration rate and indoor aerosol concentration levels in two buildings equipped with passive house technology and one "conventional" house in Ócsa, Hungary. We have aimed to determine the indoor aerosol pollution level and its elemental composition, establish the relationship between the indoor and outdoor concentration levels, and study how the different ventilation rates and modes affect the indoor particulate matter (PM) contamination. Our results indicate that the measured PM concentration levels were well below the recommended limits overall. In particular, the mean PM_{fine} (aerodynamic diameter <2.5 μm) concentration was around 5 μg m^{-3} while the outdoor PM_{fine} level was 20 μg m^{-3}. The mean indoor concentration of the coarse fraction aerosols (aerodynamic diameter >2.5 μm) varied between 2.5 and 7 μg m^{-3}, with higher values corresponding to better airtightness of the house. As assessed by the indoor/outdoor elemental ratios and mass size distribution data, the filtration of the coarse mode particles was adequate in the passive houses. However, the PM_{fine} fraction could get through the filters unhindered, as indicated by PM_{fine} levels independent of the ventilation modes. The coarse mode particles inside the passive houses mainly originated from indoor sources.

***Keywords*:** Passive House; Mechanical ventilation with heat recovery; Indoor air quality; Airborne particulate matter; Elemental composition of PM

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INTRODUCTION

Atmospheric aerosols are one of the biggest environmental problems of urban areas nowadays (Forouzanfar *et al*., 2016; Hänninen *et al.,* 2014), not least because of the observed associations between increased PM (particulate matter) concentration and various health problems (e.g. respirarory and cardiovascular morbidity) (Forouzanfar *et al*., 2016; Pope *et al*., 2002; Reichardt, 1995). A better understanding of aerosols in our indoor environment is at least as important as the outdoors because people spend 30-60% of their time in indoor environment (Jenkins *et al.,* 1992). Yet relatively few studies made indoor air pollution their focus. (Moschandreas *et al.,* 1979; Kulmala *et al.,* 1999; Abt *et al,* 2000; Özkaynak *et al.,* 1996; Koponen *et al.,* 2001; Hussein *et al.,* 2005; Dimitroulopoulou *et al.,* 2006; Martuzevičius *et al.,* 2008; Chen and Zhao, 2011; Hänninen *et al.,* 2011) Aerosol particles in the indoor environment may originate from indoor sources or infiltrate from the outdoor air. (Hänninen *et al.,* 2013) Thus, in order to estimate personal PM exposures in a building of interest, it is important to known what types of connection exist in that building between the indoor and outdoor environment. This connection will depend on the type of construction and ventilation of the building. A trend embraced by architects during the last few decades is to maximize energy efficiency as much as possible, resulting in the growing number of passive houses being built. To date, high construction costs have limited the spread of this type of house to high-income countries. But as the technologies involved become more affordable with time, energy-efficent construction is expected to broaden its popular appeal. Beacause of the relative novelty of this type of buildings, their indoor air quality has not been widely investigated (Wallner *et al*., 2015; Wells *et al*., 2015; Kauneliene *et al.,* 2016; Guyot *et al.,* 2016; Maas *et al.,* 2017). We are aware of only four papers that have focussed on PM concentrations at energy efficient buildings (Derbez *et al.,* 2014a, b; Langer *et al.,* 2015;

Broderick *et al.,* 2017), and ours is the first to report elemental concentration of PM in passive houses.

In an indoor environment the PM concentration level is affected by many mechanisms and processes. As mentioned above, the indoor PM concentration of a house is pimarily depends on the outer aerosol pollution and the connection of the outdoor and indoor environment. For example a strongly polluted outdoor air could profoundly affect the indoor air quality in a poorly shielded house. In addition, several other processes also play an important role in affecting the indoor PM level (Morawska and Salthammer, 2003), including: penetration of outdoor particles (through doors, windows and building envelope); deposition and resuspension of indoor particles; removal of indoor particles by ventilation and exfiltration; chemical reactions leading to particle formations and generations. Several aerosol studies focussed on these effects and their results were well summarized in review papers (Wallace, 1996; Lai, 2002; Holmes and Morawska, 2006; Diapouli, 2013). Because of the above factors, passive houses can differ significantly from conventional ones. In particular, remarkable differences could be expected in indoor air quality and PM concentrations between the two house types.

Passive houses represent a unique indoor environment because of their special construction. A house built with passive house technologies typically include the following features: passive solar gain (through south-facing windows), super glazing (U-value 0.8 W/m^2K), airtight building envelope, and thermal bridge free construction (Passive House Institute, 2016). These reduce the annual demand for space heating to 15 kWh m^{-2} a^{-1}, with the limit for total primary energy use of 120 kWh m^{-2} a^{-1}. To fulfil these requirements balanced mechanical ventilation with heat recovery (MVHR) must be built in. In addition, during the heating season no uncontrollable natural ventilation or considerable infiltration through the leaks of the building shell is allowed.

A recently published paper by Langer *et al.* (2015) reports the quality of the indoor air quality (IAQ) in newly built passive dwellings was comparable to or better than in conventionally built new houses. Derbez *et al.* (2014a, b) evaluated the IAQ and the occupants' comfort in newly built low energy houses during the pre-occupancy stage and during the first and first three years of occupancy. The authors reported that compared to standard French buildings, the concentrations of $PM_{2.5}$, volatile organic compounds and radon were low, whereas the formaldehyds and CO_2 levels were not significally different. However, Hasselaar (2008) displayed that certain health problems occur twice or three times more often in dwellings with mechanical ventilation with heat recovery than the ones equipped with conventional (exhaust) ventilation with natural inlet functions. He found the poor overall ventilation explains these problems. Heidorf (2007) found that the average CO_2 levels in a passive-house school were high but comparable to those in conventionally ventilated ones. In summary, the available data on IAQ in passive houses is scarce, and these studies often contradict each other or commonly held assumptions. An example for the latter is Guyot *et al.* (2016) whose measurements in low-energy homes challenged the assumption that leakage is uniformly distributed. The authors consider the case of a building with substantial ventilation where some rooms can become underventilated if short-circuited. Recognizing that such special circumstances can have a strong impact on IAQ, Guyot *et al.* developed a performance-based approach for ventilation in low-energy buildings that integrates IAQ and health issues.

In this study we have investigated the indoor air quality with a special emphasis on airborne particulate matter pollution, and compared it in buildings with passive house technology and in a conventional house. The effects of the outdoor environment, the ventilation system and human activity on the IAQ were also studied.

METHODS

Environmental character

Due to the very few numbers of passive residential houses in Hungary, it was hard to find neighbouring ones. The buildings involved in this study are in Ócsa, Hungary, a suburban settlement of detached houses with a population of 9064 (in 2010) to the south-east of Budapest. The village is about 8 km outside the city borders of the capital and commuting is served by a highway and a direct railway line. The vehicular traffic intensity in the village is quite low. For these reasons the proportion of the traffic-related aerosol is probably small. Similarly to other rural and suburban regions of Hungary, a wood-burning stove is frequently used for the heating of residential buildings, although heating with natural gas is also common. There is no notable industrial production plant, but the village is surrounded by cultivated areas and in the south - next to the investigated houses - there is a natural reserve.

The buildings

All three investigated detached houses are family residences. They were built in the southwest part of Ócsa, about 1 km distance between each. Each house fulfils the requirements of the Hungarian building code of energy efficiency. The conventional house (House A) is a century old, enlarged adobe house, with the loft converted to an attic (two floors). The enlargement is made of modern brickwork, the loft is of timber-frame construction insulated with a plastic air-vapour barrier. The old windows were renovated: an air-tight seal was built into the plank frame windows and the inner layer of the glass pane was replaced with a double-layer heat-insulated glazing. The whole building was insulated outside with special reed boards and plastered with clay (adobe should only be insulated with vapour-permeable materials). The building is heated with a wood gasification boiler which is placed in a nearby outbuilding. It is not equipped with mechanical ventilation.

The other two buildings were new: both of them were inhabited for less than a year in the time of the study. Each of them was planned as a passive house, but failed to fulfil the requirements of the passive house standard in one way or another. That is why we called them "buildings with passive house technology" instead of the term of "passive house". One of them (House B) is made of reinforced concrete with permanent molded polystyrene formwork, functioning at the same time as insulation. It has a slab foundation with polystyrene insulation under it. The wooden ceiling is insulated with mineral wool. Because of the sophisticated geometry of the ceiling and the unskilled contractor, the cracks were not sealed properly; therefore the building does not meet the requirement of airtightness of passive houses (although it fulfils all the other requirements of the passive house standard). The windows and doors are made of P.V.C. frame with three-layer glazing. The house is equipped with a balanced heat recovery mechanical supply and exhaust ventilation system. The building is heated with an electrical floor heating (only in the bathroom) and an electrical wall panel heating (in the living room). During the time we conducted the measurements for this study, a steel plate chimney was built in for a fireplace. There is no heating in the other rooms.

The third building (House C) is made of brickwork and has a reinforced concrete slab ceiling. The walls are insulated with expanded polystyrene foam and the ceiling with blown cellulose. The doors and windows are P.V.C. structures with three-layer glazing. The house is ventilated with a balanced mechanical ventilation with heat recovery and heated via ceiling-mounted electrical radiant panels. The building fulfils the requirements of the passive house standard, except for the annual demand for space heating - it is slightly more than 15 kWh m^{-2} a^{-1}. and the airtightness is a bit worse than acceptable. Both of the investigated houses are equipped with only primary air filters of grade G4.

The new European standard for air filters, EN 779:2012 (2012), the purpose of which is to classify air filters based on their lowest filtration efficiency, defines three filter classes: G1-

G4 Coarse filters; M5-M6 Medium filters; F7-F9 Fine filters. The desired air quality can be achieved economically by two-stage air filters, with the 1st stage a grade G3 or G4 filter and the second stage is a secondary filter of grade F7 or F8 filter. Filters grade G4 perform almost 100% retention of PM larger than 5 μm, while filters F7 performs the same retention of PM larger than 2 μm. The use of finer filters should reduce the volume of the fine particles that get in through the ventilation system.

The summary of the key characteristics of the three buildigs are presented in Table 1.

The correlation between airtightness and annual infiltration rate

In terms of indoor air quality the principal difference between passive and conventional houses is the level of airtightness. Passive houses are airtight; therefore only a relatively small amount of outdoor air gets in uncontrolled through the cracks of the building envelope. In contrast, because of the mechanical ventilation system a significant amount of fresh air gets in under controlled and regulated conditions, cleaned through filters. Window opening is not typical. By comparison, the airtightness of conventional buildings is tipically poor, and air exchange is caused in uncontrolled and unpredictable contributions by leakage and infiltration on the one hand and by airing on the other hand.

For the estimation of the average infiltration rate we used the Blower-door tests, a commonly used method to determine the airtightness of buildings. The equipment measures the airflow at a given building-to-outside reference pressure (ΔP = 50 Pa) and calculates the "air changes at 50 Pa" (former ACH_{50}, nowadays called n_{50}, h^{-1}). According to the passive house standard, the building must leak no more than 0.6 air changes per hour ($n_{50} < 0.6\ h^{-1}$). ACH_{50} or n_{50} should not be confused with an infiltration rate, because it is an air flow at an artificially induced condition. It is an indicator of leakage, and is not equal to infiltration (Sherman, 1987).

185

186 ***Air change rate of infiltration***

187 Several studies have been conducted on the correlation between the leakage of the building
188 shell (n_{50}) and the annual average infiltration rate (Sherman, 1987; Jokisalo *et al.,* 2009). By
189 comparing the leakage-infiltration map of Sherman to the updated world map of the Köppen-
190 Geiger climate classification (Kottek *et al.,* 2006) we obtained the formula

191

192 $$n_{inf,\ winter} = 1.33\ n_{50} / (18\ cf_1\ cf_2\ cf_3) \quad (1)$$

193

194 where $n_{inf,\ winter}$ is the air change per hour in the winter season via infiltration, n_{50} is the air
195 change rate per hour at 50 Pa of pressure difference (h^{-1}) and $cf_{1,2,3}$ are the correction factors
196 shown in Table 2. The correction factors according to Sherman (1987) are: cf_1 – height
197 correction factor (decreasing with the number of stories, e.g. cf_1=1.0 for 1 storey; cf_1=0.8 for
198 2 stories, etc.), cf_2 – shielding correction factor (cf_2=1.2 for well shielded; cf_2=1.0 for normal;
199 cf_2=0.9 for exposed), cf_3 – leakiness correction factor (cf_3=1.4 for small cracks; cf_3=1.0 for
200 normal; cf_3=0.7 for large holes).

201 The PHPP (Passive House Planning Package, a spreadsheet based design tool aimed at
202 architects and designers to assist the design of passive house standard) calculates the
203 infiltration on the basis of the ISO/PDIS 13790 (2007) (Feist, 2007) The PHPP provides the
204 following functional relationship between n_{50} and n_{inf}:

205

206 $$n_{inf} = n_{50}\ e\ V_{n50} / V_L \quad (2)$$

207

208 where V_{n50} is the volume taken into account at the airtightness measurement and V_L is the
209 heated volume (in our cases their ratio is 1.00). The quantity "e" is a coefficient of shielding

(assumes a value of 0.07) (in Table 3). It is important to keep in mind that the software Passive House Planning Package is not feasible for applications in "conventional" buildings.

Air change rate of ventilation

There is no mechanical ventilation system in House A, therefore this building has an air change only through the cracks. The airing and air change due to door opening depends among other factors on the occupants' behaviour, the climate and the season. In this case, considering that the occupants only made short time airing once a day in the morning (since the temperature was constantly below zero during the measuring campaign), the ventilation via open doors or windows was deemed to be insignificant. The other houses are equipped with a balanced heat recovery mechanical supply and exhaust ventilation system. In these houses with passive house technology - owing to the continous mechanical ventilation – airing is not typical and not recommended, either.

The air change rate of mechanical ventilation is determined by two aspects: CO_2 concentration and humidity. The required air volume is 30 m^3 h^{-1} $person^{-1}$ to keep the CO_2 concentration in the living space at 0.1% or below. However, the excessive air volume getting into the house in winter results in a very low indoor humidity level which can cause sensory irritation. So as to avoid the very low air humidity (recommended rate is 40-60%) the air change per hour must be between 0.3 and 0.5 h^{-1}. In passive houses the lower value is recommended. Thus, in these cases, the requirement is, $n_{vent} = 0.3$ h^{-1}.

Aerosol sampling

The aerosol sampling campaign was carried out during a two-week period starting on January 3 and ending on January 16, 2012. The sampling time was 48 hours, which started at 9 am or 12 pm depending on the residents' activities. The aerosol sampling took place in the

following places: on the veranda of House A (outdoor), in the bedroom of House A, in the bedroom of House B and the living room of House C. The bedrooms of House A and House B were guestrooms with no indoor activitiy. The living room of House C had usual daytime activities of a four-member family. The following sampling devices were used: Nuclepore two-stage samplers and two ten-stage PIXE International cascade impactors. The Nuclepore samplers were loaded with two Nuclepore polycarbonate filters with different pore diameters to collect the aerosol particles separately in two size fractions. One of the filters had 8 μm diameter holes to collect the coarse fraction (PM_{coarse}, particles with aerodynamic diameter larger than 2.5 μm) while the fine particles (PM_{fine}, particles with aerodynamic diameter smaller than 2.5 μm) were deposited on a filter with 0.4 μm pore diameter. (Hopke *et al.,*1997; Maenhaut *et. al.,* 1994). Portable membrane pumps developed at MTA Atomki were used to carry out the samplings. The collection of aerosol samples was carried out with a flow rate of 14-15 1 min^{-1}. Furthermore, ten-stage PIXE International cascade impactors were applied to collect size-resolved samples in the following size fractions <0.06, 0.06-0.12, 0.12-0.25, 0.25-0.5, 0.5-1, 1-2, 2-4, 4-8, 8-16, and >16 μm aerodynamic diameter. These particles were collected on kapton foils coated with paraffin. The samplings were carried out outdoors on the veranda of House A and indoors in the bedroom of House B.

Aerosol analysis

The total mass concentration was determined by gravimetric methods: the filters were weighed before and after on a microbalance. Before weighing the filters were conditioned at least 24 h in the weighing box at 24 °C temperature and approximately 50% relative humidity. Particle induced X-ray emission (PIXE) analytical method (Maenhaut and Malmquist, 2001) was used to determine the elemental composition of the aerosol samples at the PIXE chamber installed on the left 45° beamline of the 5MV Van de Graaff accelerator of the IBA laboratory

of the Institute for Nuclear Research, Hungarian Academy of Sciences (MTA Atomki) (Borbely-Kiss *et al.,* 1985). A proton beam of 2 MeV energy and 40 nA current was applied to irradiate the samples. 40 μC was the accumulated charge on each sample. The PIXEKLM program package (Szabo and Borbely-Kiss, 1993; Szabo, 2009) was utilized to determine the elemental compositions for Z>13. Concentrations of the following elements were determined: Al, Si, P, S, Cl, K, Ca, Ti, V, Sc, Co, Mn, Fe, Ni, Cu, Zn, As, Ba, Cd and Pb. The values were given in ng m^{-3}. Depending on the element the detection limit varied between 0.5 and 20 ng m^{-3} while the uncertainty of the determination of concentration was between 2% and 10%.

RESULTS AND DISCUSSION

Ventilation rates of the investigated houses

The results of the Blower-door tests are presented in Table 1. The n_{50} value in both Houses B and C (1.60 and 0.89 h^{-1}) were unable to meet the requirement of airtightness of passive houses (Feist, 2007). The n_{50} value of House A was extremely high (7.13 h^{-1}).

Table 2 shows that our conventional building has a relatively high air change rate (0.94 h^{-1}), but only through the cracks. According to Tables 2 and 3 the other two houses have far less natural infiltration, but a relatively large air exchange which is provided by artificial ventilation (0.30 h^{-1} each). Comparing these values for the three buildings, it can be seen that the conventional building (House A) possesses the highest air change per hour, approaching 1 (0.94). However, this rate of infiltration is is fully uncontrolled. The total air change rate of House B (0.42 h^{-1}) amounts to approximately half of House A, and infiltration reaches only about 30% of it. The total air change rate is the lowest at House C (0.35 h^{-1}), with a 15% infiltration rate, therefore the mechanical ventilation is the dominant ventilation mechanism of this building.

Using the method of the PHPP, the infiltration values in the heating season of buildings B and C, derived from n_{50} using a coefficient e = 0.07 (slight shielding) are shown in Table 3. There is no result for House A in this table, because this method is not feasible for applications in "conventional" buildings. It is evident that the $n_{inf,winter}$ values of Houses B and C negligibly differ from the ones shown in Table 2 (House B: 0.12 and 0.11; House C: 0.05 and 0.06).

Mass concentration of PM_{fine} and PM_{coarse}

The indoor and outdoor average and min-max PM_{coarse} and PM_{fine} mass concentration (µg m^{-3}) is summarized in Table 4. In all cases, PM_{coarse} and PM_{fine} mass concentration values exceed neither the guidelines of the Environmental Protection Agency (EPA) nor the ones of the Occupational Safety and Health Administration (NAAQS/EPA, 2016; OSHA). Regarding the different buildings, the average indoor PM_{coarse} concentrations were higher in the buildings with passive house technology than in the conventional one. The indoor PM_{fine} concentration levels were about 25% of the outdoor PM_{fine} levels in all three buildings. Despite the very different air change rate values measured in the three houses, the average PM_{fine} indoor mass concentrations was about the same in each. This is taken as evidence that the infiltration of PM_{fine} was independent from ventilation mode. The average of the $PM_{fine}/PM_{fine+coarse}$ ratio (see Table 5) was 0.4 in House C and 0.6 in House B, while it was 0.7, the same as outdoors, in the conventional house (House A). This means that the outdoor air could penetrate into House A without hindrance due to the very high air exchange rate which was observed in House A. These ratios indicated that fine particles comprised a large fraction in the total mass at each sampling site, except for House C where coarse particles were dominant. The Indoor/Outdoor ratio is used to characterise the relation between the indoor and the outdoor aerosol concentration. In this case, it is not easy to draw any conclusion based on this value,

because it is influenced by many factors such as outdoor concentration, penetration factor, deposition factor, indoor particle source emission rate and indoor activities. The average of I/O ratio of the mass concentration in all sampling sites is summarized also in Table 5. The average of I/O ratio for the fine fraction was lower than for the coarse mode particles, indicating weaker indoor sources for PM_{fine} at the houses. In addition the average I/O ratios for the fine fraction were nearly identical in each house, suggesting that the origin of the PM_{fine} indoor pollution was the outside air. In the conventional house the I/O ratio for the fine and coarse fractions were nearly the same indicating that the origin of both fine and coarse fractions of indoor pollutants was most probably the outside air, as suggested by the high exchange rate. Furthermore, the I/O ratio for PM_{coarse} was lowest in the conventional building (0.4 in House A) and much higher in the buildings with passive house technology (0.7 in House B) and 1.2 in House C suggesting that the main PM_{coarse} source was the resuspension of household dust. The differences between Houses B and C can be explained by the much higher level of activities in House C.

The variation in time of PM_{fine} and PM_{coarse} inside the houses and outdoors is presented in Fig. 1. The huge increase of the PM_{coarse} in House B on 10-11th January 2012 could be attributed to the drilling of the reinforced concrete during the modification of the heating system. As we mentioned earlier, an external steel chimney for the fireplace was built into House B on those days. Therefore these results were excluded as outliers from further analysis of the data.

Regarding the fine fraction, the indoor PM_{fine} concentration levels closely followed the variation in the external values, with an approximately 50% attenuation in levels. In the case of the coarse fraction, no lawful dependence could be identified between the internal and external concentration values.

335 ***Elemental composition***

336 The average concentrations of 15 elements in both size fractions at each sampling site are
337 presented in Table 6. We found in all sampling sites, that the concentration of anthropogenic
338 related elements such as sulphur, potassium, zinc and lead was always the highest in the fine
339 fraction. Nevertheless, the temporal variation of the concentration of these elements was
340 similar during the campaign in all selected buildings (Fig. 2). Similar to the variation of the
341 PM_{fine} mass, the indoor alteration of the concentrations followed the outdoor change of these
342 elements, a strong evidence that this phenomenon was likely the result of outdoor particles
343 being transported to the indoors. The S and Zn PM_{coarse} concentrations inside all houses also
344 followed the external changes. However, in the case of the mineral dust elements like Al, Si,
345 Ca, Ti, Mn, Fe, no correlation was found between the external and internal values.

346 The I/O ratios calculated for the measured elements can be found in Table 7. As mentioned
347 before, the I/O ratio is strongly affected by many factors like indoor particle sources,
348 penetration factor, air exchange rate and outdoor particle concentration. For example, if there
349 is no indoor particle emission source, the I/O ratio will increase with the increase of the air
350 exchange rate, while if the indoor particle emission rate is very large and the outdoor particle
351 concentration is low, the I/O ratio will decrease with the increase of the air exchange rate
352 (Chen and Zhao, 2011).

353 In our study, the average of the I/O ratio values were below 1 in the coarse fraction except
354 for copper at all houses. Moreover, in House C the I/O ratio for some elements (S, K, Zn) was
355 higher than 1.

356 In the fine fraction the average I/O ratios for elements S, K, Pb, Zn in all houses were
357 around 0.4, similar to the PM mass. For mineral dust elements the mean I/O ratio was around
358 1 for House A and C, and for House B it was twice as much. For Cl and Cu, I/O ratios higher
359 than 1 were found in the houses.

In the coarse fraction smallest I/O ratios were measured in House A both for the anthropogenic and the mineral dust elements. I/O ratios for S, K and Pb was highest in House C while for mineral dust elements the average I/O ratios were similar in the two passive houses (0.5). The highest I/O ratios were found for Cu in all houses.

The lower I/O ratios suggest external sources of the given element. We discuss the possible origin in paragraph *Emission sources.*

Size distribution

The mass size distribution of some elements was measured both indoors (House B) and outdoors (House A) from 3rd to 6th January 2012. These results are shown on Fig. 3. According to similarities in the size distribution, the elements were classified into two different groups in both cases. The first group contains soil mineral compounds such as Si, Ca, Fe and Mn. In case of these elements two prevalent peaks were found in the coarse fraction at the outdoor sampling site: one at 2-4 μm aerodynamic diameter size range and the other higher peak at 8-16 μm size range. The indoor concentration of Si and Ca were elevated towards the coarse mode with the dominant peak at the 2-4 μm size range. Moreover, Fe and Mn size distributions were shifted: one lower dominant peak was observed at the 1-2 μm size range. This means that the high amount of soil-derived elements disappeared in indoor environment, confirming that the applied G4 filters are adequate for EU standard. The second group consists of elements of anthropogenic origin: S, K, Zn and Pb. These elements could be derived from combustion processes such as biomass burning or oil combustion. In their size distribution there was an increase at 0.25-0.5 μm and at 1-2 μm aerodynamic diameter size range at the outdoor sampling site. Nevertheless, only one predominant peak could be found at the 0.25-0.5 μm size range at the indoor sampling site. Furthermore, the indoor concentrations were approximately the third of the outdoor aerosol concentrations in all cases.

As inferred from the elemental I/O ratio and the size distribution, fine particles (containing S, K, Zn and Pb) could get through the filters.

Emission sources

Enrichment factor (EFs) analysis provides a picture of whether the origin of the indoor or outdoor particulate matter is the crust or some anthropogenic activities. Furthermore, if the element has indoor sources, the EFs of this element are higher indoors than outdoors. In this work EFs were calculated relative to the average crustal rock composition (Mason and Moore, 1982) using Si as the reference element, as shown in the following equation

$$EF = (X/Si)_{PM}/(X/Si)_{crustel} \quad (3)$$

However, these ratios are not able to describe the relative strength of outdoor sources. Thus the indoor/outdoor EF ratios (in the following: indoor EF) were calculated (Salma *et al.,* 2013). This relative enrichment to the outdoor aerosol composition is displayed in Table 8. If the value of the indoor EF is equal or less than 1 the element usually has an outdoor source. Significantly higher indoor EF values indicate indoor origin. For all elements the PM_{fine} indoor EFs ranged from 0.1 to 6.3 and their average levels were less than 1, indicating that these aerosol components could be of outdoor origin in each house. The maximum EF was higher than 1 for some elements (Ca, Ti, Cu). These larger ratios should be attributed to indoor sources. The maximum indoor EF of Ca was higher in House A (2.0) and in House C (2.2) than in the House B, where weaker indoor activity was observed. Furthermore, the maximum indoor EF of Cu was also larger in House A (6.3) and in House C (3.3).

Moreover, the PM_{coarse} indoor EF were higher than 1 for some elements (Al, Ti, Ca, S, K, P, Cu, Zn) in the coarse fraction, displaying that these aerosol constituents were extensively

410 enriched and could be derived from indoor sources. At the conventional house (House A) the
411 greatest indoor EF were observed for K, P and Cu. The indoor EF of the potassium was varied
412 from 1.2 to 2.6 and their average was 1.8. In addition, the observed range of the phosphorus
413 was between 0.9 and 2.7 with the average of 1.6. As mentioned above, this building is heated
414 with a wood gasification boiler. Due to this, K and P could be enriched indoors. Furthermore,
415 it is possible that the elevated value of these elements has biological origin from indoor plants.
416 Moreover, the copper indoor EF ranged from 1.4 to 15.8 with average of 5.7, which was the
417 highest value compared to the average of the buildings with passive house technology.
418 The probable source of Cu might be electric devices that apply copper commutators for motor
419 rotation such as vacuum cleaners and electric fans (Zhao *et al.,* 2006). A vacuum cleaner was
420 used in the lobby every day.
421 In House B, the maximum indoor EF of some elements (Al, S, K, Ti, Mn, Cu, Zn) was
422 above 1, suggesting that these elements should be enriched only a few days. On the last
423 sampling date, the indoor EF of Al and Zn was significantly high. The reason for this
424 phenomenon was unknown.
425 Furthermore, the highest indoor EF values were observed in House C in the coarse fraction,
426 where the human activity was the highest at the sampling site. Here the indoor EFs for S, K,
427 Ca, Ti, Cu, Zn were higher than 1. The indoor EF of S ranged from 1.8 to 3.4 and the average
428 level was 2.6. The range of K was between 1.2 and 2.2, with an average at 1.8. Potassium and
429 sulphur could be related to indoor sources such as cooking, smoking, emissions of wood fires
430 and human activity (Moschandreas *et al.,* 1979). In addition, greater indoor EFs were noticed
431 for Cu, and Zn too.
432 In both buildings with passive house technology, indoor EFs for Zn were higher than in the
433 conventional house. Nevertheless, the origin of Zn could not be identified and no relationship
434 between the enriched Zn and the passive house were found either. There is no detectable

source of Zn in these houses, and the ventilation system as a possible Zn source has not been examined. The latter possibility requires further investigation.

Removing particles of both indoor and outdoor sources is quite slow through the cracks of the building shell (Thatcher and Layton, 1995), the ventilation system is more effective at it. But in these cases the air change rate through the building envelope of House A (n_{total} = 0.94 h^{-1}) is about two times larger than the air change rate originated from the mechanical ventilation system of Houses B and C (n_{total} = 0.42 and 0.35 h^{-1}) - so the effectiveness of removing contamination through infiltration and through ventilation are comparable to each other. It is quite obvious that using a ventilation system that operates below its filtration efficiency specifications (or potential?), the indoor air quality may not be adequately improved even in passive houses. In these two specific cases, the coarse aerosol particles remained indoors and accumulated despite ventillation.

CONCLUSIONS

~~In this work indoor aerosol mass concentration, mass size distribution and elemental composition were studied in a conventional house and in two energy efficient buildings in a winter period.~~

~~Contrary to expectations, our observation did not support the assumption that the indoor air quality with regard to PM contamination of buildings with passive house technology would be better than that of conventional houses. Although neither of the investigated energy efficient buildings complies with the requirements of airtightness of the passive house standard, their pressure test air flows (n_{50}) were 4.5-8 times lower than those of the conventional house. The measured particulate matter concentration levels were well below the recommended limit values.~~

We found that the average PM concentration was higher in the buildings with passive house technology than in the conventional one. The difference was due to a much higher concentrations of coarse particles found in the passive houses. The mean of the coarse fraction aerosols indoor concentration varied between 2.5 and 7 μg m^{-3}, with higher values corresponding to better airtightness of the house. In the fine fraction no differences were found between the houses, and we showed that origin of the fine fraction aerosol was the outdoor air. The PM_{fine} concentration was in average around 5 μg m^{-3} in all houses while the outdoor PM_{fine} level was 20 μg m^{-3}. From the indoor/outdoor elemental ratios and mass size distribution data we could conclude that the filtration of the PM_{coarse} was solved in the passive houses while the PM_{fine} fraction could get through the filters without hindrance. The PM_{coarse} inside the passive houses mainly originated from indoor sources and the clearance of this PM_{coarse} was not solved. The PM_{fine} levels were independent of the ventilation modes.

It seems to be a problem that buildings B and C equipped with a balanced mechanical ventilation system with heat recovery have only primary air filters of grade G4 without the recommended secondary ones of grade F7. Unfortunately this is a designing error, which can be repared later only at a high cost, since the filtering system is supposed to be set up gradually, so that the elements are not overloaded (1st stage G1-4, 2nd stage F7-8, the 3rd stage - HEPA, ULPA - are optional). Based on Fig. 3 we can see that the coarse fraction is filtered from the air getting in, while the fine particles are present at a greater extent. This undoubtedly shows that the secondary filter grade is missing. Without realizing the effective filtering possibilities of the ventilation system, the indoor air quality cannot be improved even in passive houses.

It would be worth making further investigations in buildings equipped with better filters (even with HEPA filters) if they indeed perform PM filtering closer to assumptions.

~~Unfortunately we were only able to investigate a very few number of dwellings, due to the limited number of passive houses in Hungary and the unsuccessful application for further grants. Additional sampling of airtightness and indoor/outdoor PM in a greater number of homes that control for key variables (e.g., building location, occupancy status during sampling, interior finishing, heating system, etc.) would be necessary to gain an empirically well-supported understanding of indoor-outdoor air quality relationships in passive house dwellings.~~

We have characterized and compared indoor aerosol pollution in two energy efficient buildings and in a conventional house in Hungary during the winter of 2012. Indoor pollution level, composition and possible sources of PM were determined via measuring the mass concentration, mass size distribution and elemental composition indoor and outdoor aerosols at the same time. We also measured airtightness.

Although pressure test air flows (n_{50}) in the two energy efficient buildings in the study were 4.5-8 times lower than in the conventional house, neither complied with the requirements of airtightness of the passive house standard.

In all three houses, the measured particulate matter concentration levels were well below the WHO-recommended 24-h limit. Nevertheless, the average PM concentration was higher in the two buildings with passive house technology than in the conventional one, with most of the excess PM concentration accounted for by coarse particles (particles with aerodynamic diameter > 2.5 μm) of indoor sources that got trapped indoors by superior airtightness and also insufficient clearance.

In contrast, there was no difference in the concentration and composition of the fine fraction ($PM_{2.5}$) in the three houses. The origin of this fine fraction aerosol was the outdoor air. Supporting this claim, the indoor/outdoor elemental ratios and the mass size distribution data indicated that the PM_{coarse} was sufficiently filtrated in the passive houses while the PM_{fine}

fraction could get through the filters without hindrance. Furthermore, the PM_{fine} levels were independent of the ventilation modes. Both energy efficient houses were equipped only with primary air filters of G4 without the recommended secondary filters. The presence of the fine particles of outdoor sources showed that this filter was not effective at all in removing this size fraction. In order to reach lower $PM_{2.5}$ levels the houses should have been equipped with better filters (even with HEPA filters). The clearance of the coarse particles originating from indoor sources should be also solved.

Our study, by highlighting several factors that bear influence on the indoor air quality of passive houses, is only a first step on the path to a better understanding of indoor-outdoor air quality relationships in such buildings. Future studies taking the necessary next steps on that path must engage in the systematic investigation of airtightness and indoor/outdoor PM in a greater number of homes while controlling for key variables (e.g., building location, occupancy status during sampling, interior finishing, heating system, etc.). The knowledge thus gained will be indispensable to developing more efficient, more dependable, and healthier future building technologies.

ACKNOWLEDGEMENTS

This research was supported by the TÁMOP 4.2.4. A/2-11-1-2012-0001, "National Excellence Program - Elaborating and operating an inland student and researcher personal support system convergence program". The project was carried out as part of a doctoral studies program of the János Bolyai Research Scholarship of the Hungarian Academy of Sciences and received funds from the European Union and was co-financed by the European Social Fund, and the National Research, Development and Innovation Office – NKFIH (contract number PD 125086).

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675

676 **Table Titles**

677 **Table 1.** Building characteristics

678 **Table 2.** Air change per hour, originated from infiltration and ventilation

679 **Table 3.** Air change per hour, originated from infiltration, using the method of the PHPP.

680 **Table 4**. Average, minimum and maximum values of PM_{coarse} and PM_{fine} mass concentrations

681 in $\mu g\ m^{-3}$

682 **Table 5.** $PM_{fine}/PM_{fine+coarse}$ and Indoor/Outdoor ratios for PM_{coarse} and PM_{fine} fractions

683 **Table 6.** The average concentrations (ng m^{-3}) of elements obtained for fine and coarse

684 samples collected in each sampling sites

685 **Table 7**. Minimum, maximum and average indoor/outdoor ratio for PM_{fine} and PM_{coarse}

686 fractions

687 **Table 8**. Minimum, maximum and average indoor/outdoor (indoor EF) ratio for PM_{fine} and

688 PM_{coarse} fractions

689

690 **Table 1.** Building characteristics

	House A	**House B**	**House C**
character	detached house	detached house	detached house
storeys	2	1	1
occupants	1 adult + 2 children	2 adults	2 adults + 2 children
occupation during measuring campaign	~ 85%	~ 50%	~ 90%
wall	adobe/brickwork + reed boards	reinforced concrete + polystyrene formwork	brickwork + exp. polystyrene foam
roof/ceiling	timber-frame construction roof + blown cellulose	wooden ceiling + mineral wool	reinforced concrete slab + blown cellulose
windows	renovated old plank frame windows	P.V.C. frames, 3 layers glazing	P.V.C. frames, 3 layers glazing
area [m^2]	199	121	135
heating	wood gasification boiler	electrical wall panel (living room) and electrical floor heating (bathroom)	ceiling mounted electrical radiant panels
cooking	electric ceramic cooker	electric ceramic cooker	electric ceramic cooker
balanced heat recovery supply and exhaust ventilation	no	yes	yes
filter grade	-	G4	G4
window opening	short time airing once a day in the morning	almost never	almost never
annual heat demand [kWh m^{-2} a^{-1}]	105.2	14.9	15.3
n_{50} [h^{-1}]	7.13	1.60	0.89

691

692 **Table 2.** Air change per hour, originated from infiltration and ventilation. Determination
693 of the leakage-infiltration ratio and the correction factors: Sherman (1987)

house	n_{50} [h^{-1}]	leakage-infiltration ratio [dimensionless]	correction factors [dimensionless]			$n_{inf, winter}$ [h^{-1}]	n_{vent} [h^{-1}]	n_{total} [h^{-1}]
			cf_1 building height	cf_2 shielding	cf_3 leakiness			
House A	7.13	18	0.8	1.0	0.7	0.94	0	0.94
House B	1.60	18	1.0	1.0	1.0	0.12	0.30	0.42
House C	0.89	18	1.0	1.0	1.4	0.05	0.30	0.35

694

695 **Table 3.** Air change per hour, originated from infiltration, using the method of the PHPP.

house	n_{50} [h^{-1}]	$n_{inf,\ winter}$ [h^{-1}] according to PHPP $n_{inf} = n_{50} \cdot 0.07$
House A	7.13	- [a]
House B	1.60	0.11
House C	0.89	0.06

696
697 [a] PHPP is not feasible for applications in "conventional" buildings

698

699 **Table 4.** Average, minimum and maximum values of PM_{coarse} and PM_{fine} mass
700 concentrations in μg m^{-3}

	PM_{coarse}			PM_{fine}		
	Min.	Average	Max.	Min.	Average.	Max.
House A	0.3	2.5	5.1	2.9	4.8	7.3
House B	1.5	3.5	4.8	3.7	5.0	8.3
House C	4.2	7.1	11.6	1.9	4.9	7.4
Outdoor	3.6	5.7	17.6	6.9	19.9	44.7

701

Table 5. $PM_{fine}/PM_{fine+coarse}$ and Indoor/Outdoor ratios for PM_{coarse} and PM_{fine} fractions

				I/O					
	$PM_{fine}/PM_{fine+coarse}$			PM_{coarse}			PM_{fine}		
	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.
House A	0.6	0.7	0.9	0.1	0.4	0.8	0.1	0.3	0.5
House B	0.5	0.6	0.7	0.1	0.7	1.4	0.2	0.4	0.5
House C	0.2	0.4	0.5	0.4	1.2	2.8	0.1	0.3	0.8
Outdoor	0.3	0.7	0.9	-	-	-	-	-	-

704 **Table 6.** The average concentrations (ng m^{-3}) of elements obtained for fine and coarse
705 samples collected in each sampling sites

	PM_{fine}				PM_{coarse}			
	House A	House B	House C	Outdoor	House A	House B	House C	Outdoor
Al	73.4	132.1	73.1	230.2	105.8	205.8	122.3	351.1
Si	47.6	86.8	34.9	32.0	172.6	238.0	186.6	591.4
P	<DL	<DL	<DL	<DL	10.7	3.5	2.1	20.0
S	213.5	257.3	266.1	737.9	43.5	51.5	118.3	130.5
Cl	26.5	16.0	11.7	38.1	33.2	25.4	58.8	410.0
K	384.5	129.4	227.0	516.5	105.0	53.8	103.9	173.0
Ca	97.6	102.4	79.9	61.6	256.4	269.8	403.0	809.2
Ti	1.5	2.1	2.1	2.0	4.3	5.2	8.2	21.2
Cr	<DL	<DL	<DL	<DL	3.0	4.5	4.4	13.2
Mn	1.1	1.7	1.1	2.9	1.7	2.5	2.1	8.0
Fe	20.4	28.7	17.1	53.5	51.4	67.9	64.1	258.1
Cu	5.1	6.6	6.7	7.0	10.3	4.8	11.8	13.8
Zn	12.6	10.8	14.9	40.0	4.2	5.1	10.7	10.4
Pb	5.2	3.7	5.1	18.6	<DL	<DL	<DL	<DL

706

707 **Table 7.** Minimum, maximum and average indoor/outdoor ratio for PM_{fine} and PM_{coarse}
708 fractions

	PM_{fine}									PM_{coarse}								
	House A			House B			House C			House A			House B			House C		
	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.	Min.	Average	Max.
Al	0.2	0.4	0.9	0.3	0.9	1.8	0.1	0.5	1.2	0.2	0.4	0.6	0.2	0.9	1.8	0.1	0.6	1.6
Si	0.8	2.3	7.2	0.5	5.5	14.1	0.4	1.6	3.0	0.1	0.4	0.9	0.1	0.8	1.4	0.1	0.6	1.9
P	-	-	-	-	-	-	-	-	-	0.2	0.7	1.6	0.3	0.2	0.5	0.1	0.1	0.2
S	0.1	0.3	0.6	0.2	0.5	0.7	0.1	0.4	0.9	0.2	0.4	0.7	0.1	0.6	1.0	0.3	1.3	3.5
K	0.5	0.8	1.6	0.1	0.3	0.6	0.2	0.5	1.1	0.2	0.7	1.4	0.1	0.6	1.0	0.2	1.0	3.0
Cl	0.4	1.5	2.8	0.3	1.1	2.5	0.1	0.8	2.7	0.1	0.2	0.5	0.0	0.2	0.3	0.1	0.5	1.2
Ca	1.0	1.8	3.1	0.6	2.2	3.5	0.5	1.6	3.3	0.2	0.4	0.9	0.1	0.6	1.2	0.2	0.8	2.1
Ti	0.5	0.8	1.4	0.3	1.4	2.4	0.5	1.2	2.0	0.1	0.3	0.6	0.1	0.6	1.1	0.1	0.8	2.2
Cr	-	-	-	-	-	-	-	-	-	0.1	0.1	0.4	0.03	0.4	1.0	0.05	0.6	1.5
Mn	0.2	0.5	0.7	1.0	1.7	2.4	0.1	0.5	1.1	0.1	0.3	0.6	0.1	0.6	1.5	0.1	0.4	0.7
Fe	0.3	0.4	0.7	0.4	0.8	1.3	0.1	0.4	0.7	0.1	0.2	0.4	0.1	0.4	0.9	0.1	0.3	0.8
Cu	0.4	2.5	8.8	0.4	1.8	4.6	0.3	1.3	2.3	0.3	2.9	10.8	0.1	1.4	4.5	0.2	3.5	16.4
Zn	0.2	0.4	0.7	0.2	0.4	0.6	0.2	0.5	0.9	0.2	0.4	0.6	0.3	0.7	0.9	0.7	1.2	2.4
Pb	0.1	0.3	0.6	0.1	0.3	0.5	0.1	0.3	0.8	-	-	-	-	-	-	-	-	-

709

710 **Table 8.** Minimum, maximum and average indoor/outdoor (indoor EF) ratio for PM_{fine}
711 and PM_{coarse} fractions

	PM_{fine}									PM_{coarse}								
	House A			House B			House C			House A			House B			House C		
	Min	Average	Max.	Min	Average	Max.	Min	Average	Max.	Min	Average	Max.	Min	Average	Max.	Min	Average	Max.
Al	0.1	0.3	0.5	0.1	0.3	0.8	0.2	0.3	0.4	0.6	1.1	2.0	0.8	2.3	7.8	0.8	1.1	1.4
P	-	-	-	-	-	-	-	-	-	0.9	1.6	2.7	0.0	0.2	0.4	0.1	0.3	0.5
S	0.0	0.2	0.4	0.0	0.2	0.7	0.2	0.3	0.4	0.6	1.0	1.5	0.6	1.0	1.7	1.8	2.6	3.4
Cl	0.3	0.9	2.0	0.1	0.3	0.6	0.2	0.5	0.9	0.2	0.4	0.6	0.2	0.2	0.3	0.4	0.6	0.7
K	0.1	0.6	1.1	0.0	0.2	0.4	0.2	0.4	0.5	1.2	1.8	2.6	0.5	0.8	1.1	1.2	1.8	2.2
Ca	0.4	1.1	2.0	0.2	0.6	1.2	0.5	1.2	2.3	0.8	1.1	1.3	0.6	0.8	1.0	1.1	1.6	2.1
Ti	0.2	0.5	0.8	0.1	0.4	0.7	0.5	1.0	2.2	0.4	0.8	1.1	0.3	0.7	1.3	0.7	1.4	2.2
Mn	0.1	0.3	0.7	0.1	0.4	1.0	0.2	0.4	0.6	0.4	0.8	1.2	0.3	0.9	1.5	0.4	0.8	1.3
Fe	0.1	0.3	0.7	0.1	0.3	0.7	0.1	0.3	0.6	0.4	0.6	0.9	0.4	0.6	1.0	0.4	0.7	1.0
Cu	0.2	1.7	6.3	0.1	0.5	1.7	0.4	1.2	3.3	1.4	5.7	15.8	0.4	1.4	3.5	1.1	2.6	4.7
Zn	0.1	0.3	0.5	0.0	0.2	0.6	0.2	0.3	0.4	0.7	1.3	2.0	0.5	2.8	11.0	1.3	3.2	6.5
Pb	0.1	0.2	0.4	0.0	0.2	0.7	0.1	0.2	0.3	-	-	-	-	-	-	-	-	-

712

713 **Figure Captions**

714 **Fig. 1.** Variation of PM_{fine} (a) and PM_{coarse} (b) mass concentration between 3.1.2012 and
715 14.1.2012

716 **Fig.** 2. Temporal variation of elemental concentration for S, K, Zn, Pb in the fine fraction
717 between 3.1.2012 and 14.1.2012

718 **Fig.** 3. Mass size distribution of elemental concentration indoors (House B) and outdoors
719 (House A)

720

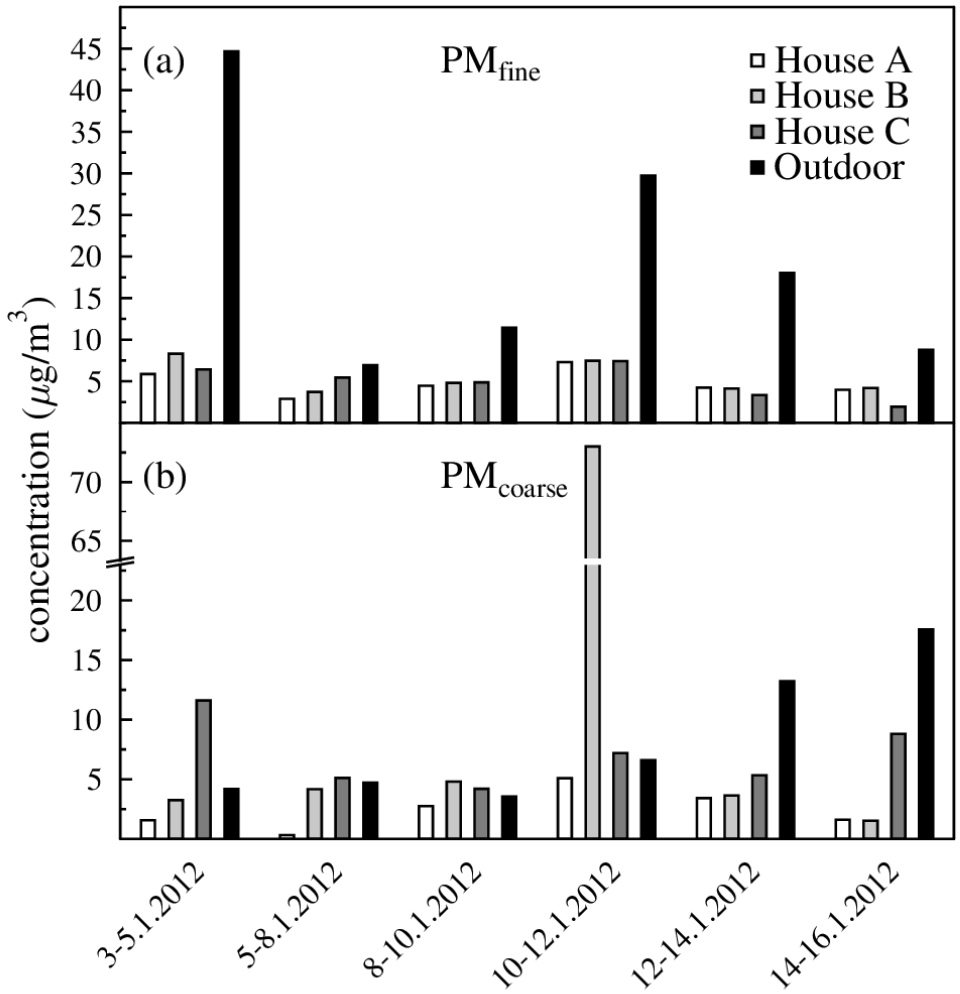

Fig. 1.

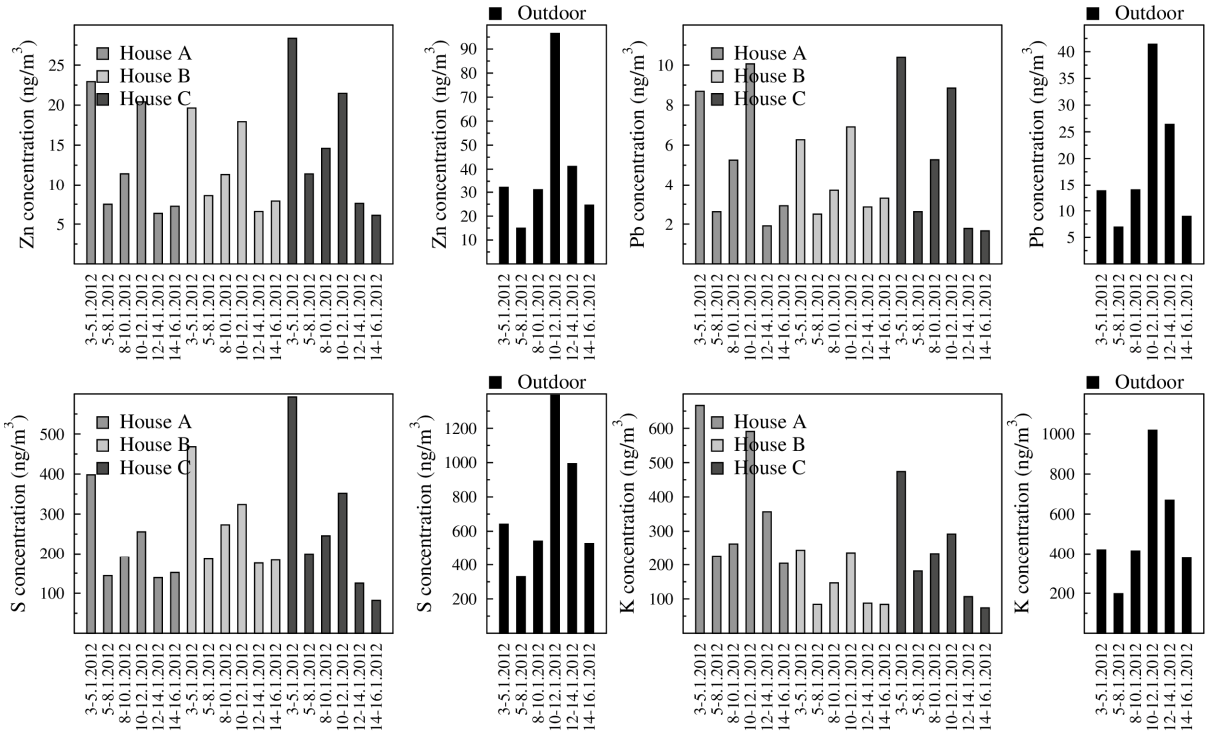

Fig. 2.

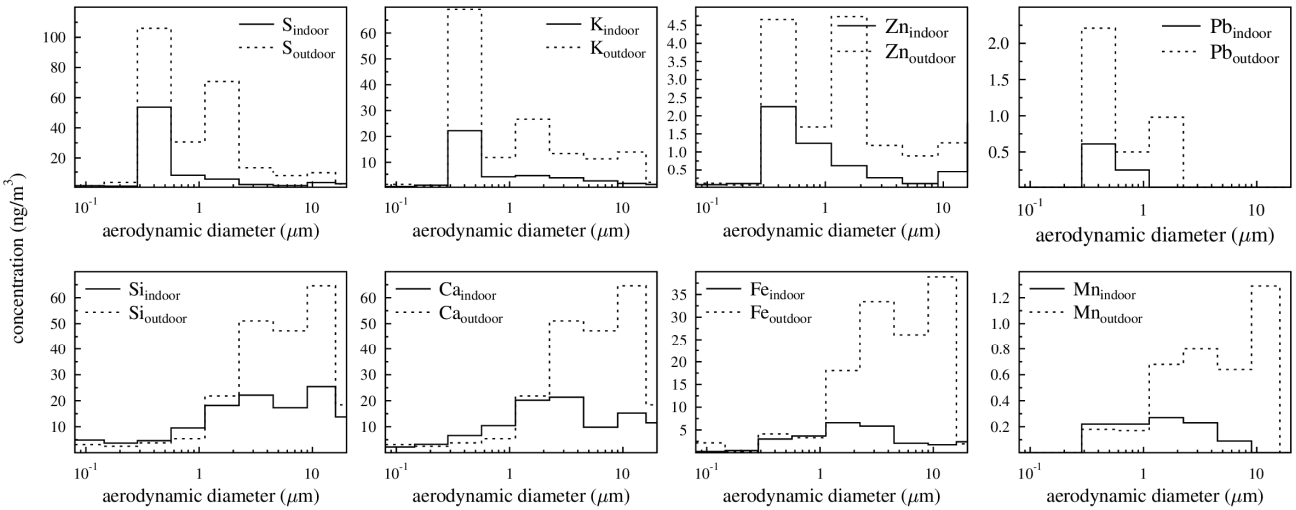

Fig. 3.