Indoor exposure from building materials: A field study

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A B S T R A C T

The present study has been conducted in the frame of BUMA (Prioritization of Building Materials Emissions as indoor pollution sources), a European funded project, aiming at assessing the exposure to emitted compounds in indoor air. Field campaigns in five (5) European cities (Milan, Copenhagen, Dublin, Athens and Nicosia) were carried out. These campaigns covered weekly winter and summer concentration measurements in two (2) public buildings and two (2) private houses in each city. BTEX, terpenes, and carbonyls were measured using passive sampling in two sites inside the building and one outside. VOC emission measurements on selected building material have also been performed using Field and Laboratory Emission Cell (FLEC). The results on indoor concentrations for compounds such as formaldehyde (1.2–62.6 μg m⁻³), acetaldehyde (0.7–41.6 μg m⁻³), toluene (0.9–163.5 μg m⁻³), xylenes (0.2–177.5 μg m⁻³) and acetone (2.8–308.8 μg m⁻³) have shown diversity and relatively significant indoor sources depending on the building type, age etc. Indoor concentrations of these substances are varied depending on the building age and type. The percentage of approximately 40% of the indoor air quality levels originated from building materials.

1. Introduction

Many studies have demonstrated that building materials can be significant emission sources of Volatile Organic Compounds (VOCs), which may affect and determine the concentration levels in indoor environments (Knudsen et al., 1999). VOCs originate from both indoor and outdoor sources; they are of particular concern due to their potential impact on human health (Marchland et al., 2006). Formaldehyde and benzene, for example, are some of the most studied pollutants since they are classified in Group 1 of human carcinogens by the International Agency for Research on Cancer (IARC, 2004).

For many of these chemicals, the risk on human health and comfort is almost unknown and difficult to be predicted because of the lack of toxicological data. In the frame of the INDEX project (Kotzias et al., 2005) the existing knowledge worldwide has been assessed in terms of type and levels of chemicals in indoor air, as well as the available toxicological information. It was concluded that VOCs such as benzene, formaldehyde, acetaldehyde, toluene and xylenes have to be considered as priority pollutants with respect to their health effects. On the other hand, chemicals such as limonene and α-pinene require further research with regard to human exposure or dose response and effects.

Reported indoor concentration of individual VOCs are generally below 50 μg m⁻³, with most below 5 μg m⁻³ (Wolkoff et al., 2006). Formaldehyde indoor concentrations were varied between 38 and 310 μg m⁻³ (Schleibinger et al., 2001). Formaldehyde mean values could reach 134 ± 93 μg m⁻³ and 86 ± 58 μg m⁻³ in new and old buildings, respectively (Park and Ikeda, 2006). Additionally, typical European indoor exposure concentrations for xylenes, acetaldehyde and terpenes varied from 2–37 μg m⁻³, 10–18 μg m⁻³ and 6–83 μg m⁻³, respectively (Kotzias et al., 2009).

Combined indoor/outdoor air quality measurements have shown that there exist significant VOC sources indoors. For example the aldehyde concentrations are usually 2–10 times higher than outdoors (Marchland et al., 2006). It has been pointed that in renovated or completely new buildings, the VOCs concentration levels are often several orders of magnitude higher (Kim et al., 2006). The main sources of aldehydes in homes include building materials, hardwood, plywood, laminate floorings, adhesives, paints and varnishes and in some cases they are products of ozone-initiated reactions (Marchland et al., 2006; Weschler et al., 1992). For example, interior coatings can increase indoor air pollution due to VOC emissions (Kwok et al., 2003). Some of the major VOCs emitted from an oil-based varnish were ethylbenzene, m,p-xylene, o-xylene and formaldehyde (McCrillis et al., 1999).
Formaldehyde is known to be released by press wood products used in home building construction such as MDFs, and paneling and products made by urea- formaldehyde resins (Kelly et al., 1999). The contribution of building materials and furnishing to indoor air pollution has been demonstrated by a study of VOC emissions in newly built, unoccupied houses at BRE (Yu and Crump, 1999). The sampling of VOCs in indoor air has shown that the contribution of building material emissions was significant during the first six months.

Although, numerous studies have investigated the levels of indoor air pollutants and emission measurements in laboratories, research on systematic in- field studies, linking the VOC concentrations to their indoor sources is rather limited (Jarnstrom et al., 2007). The primary aim of the present work is to characterize building materials as indoor VOC emission sources by conducting indoor concentration and emission measurements at houses and public buildings- including schools, across Europe. The measurements cover mainly carbonyls, BTEX and terpenes with emphasis to the abovementioned priority compounds.

2. Field study and methodology

This study was carried out from 2007 to 2008 in five European cities (Athens, Nicosia, Dublin, Copenhagen and Milan) during summer and winter period. Measurements were conducted in four buildings per city; one public building (in some cases 2), one school and two private houses. Three sites per building were selected, two indoors and one outdoor (e.g. for a public building: office, hall, out). The buildings were selected according to the following criteria: (1) the age (less than two years) (2) the last reconstruction or renovation, (3) the position of the building (urban sites preferred) and (4) the building's access. Indoor and outdoor VOC measurements were carried out using Radiello® passive samplers for one week (charcoal/carbograph type for VOCs and DNPH- covered for carbonyl compounds). At indoor locations, the passive sampling equipment was placed approximately 1.5 m above the ground, either on tables or other furniture. Outdoor sampling locations were chosen in order to avoid significant point sources of pollution, such as building exhaust vents. Selected building material in- field emissions measurements were also performed by using the Field and Laboratory Emission Cell (FLEC). The selection of the building surfaces for FLEC emission measurements was based on criteria such as the material involved is known as “good” emitter, material with high frequency of appearance in the present field campaign time, relative high room concentrations on priority pollutants, as well as, building/surface accessibility. The FLEC measurements have been carried out according to ENV standards (ENV 13419, 2003; ASTM 5116, 1997). Tenax TA for BTEX and its homologues and DNPH cartridges for carbonyls were used as absorption tubes for the FLEC experiments. The supply air flow for BTEX was set to 100 ml min⁻¹ and the sampling rate, in each tube, to 40 ml min⁻¹ for 30 min. The total amount of sampled air through each tube was 1200 ml. Respectively, the supply air flow for aldehydes was set to 400 ml min⁻¹ and the sampling rate to 160 ml min⁻¹ for 30 min, as well.

Samples analysis conducted according to ISOs (ISO 16000-3, 2001; ISO 16000-6, 2004). The analysis of BTEX and its homologues trapped in Radiello samplers was performed by GC/FID after desorption recovery of the analytes with C₅₂ and were confirmed by GC-MS. The analysis of carbonyls was carried out using HPLC-VIS (λ = 360 nm) after desorption of the analytes with acetone.

A quality assurance program was implemented in the framework of which all analytical systems related to the analysis of VOCs have been checked for their performance. Various Quality Control tools were used in order to ensure that adequate laboratory performance was maintained. These included Control Charts for standard solutions, analysis of Control Standards as unknowns and analysis of CRM's (CRM562 Aromatic hydrocarbons sorbed on active charcoal in tubes and CRM551 2,4-DNPH derivatives in Acetoneitrile). The Limits of Detection for seven days exposure for BTEX and terpenes were determined and found to be 0.1 μg m⁻³ for benzene, toluene, ethyl benzene, m,p-xylene and 0.2 μg m⁻³ for o-xylene, α-pinene, 1,2,4 – TMB and D-limonene. The Limits of Detection for seven days exposure for carbonyls were determined and found to be 0.1 μg m⁻³ for formaldehyde, acetaldehyde and acetone and 0.15 μg m⁻³ for propionaldehyde and 0.3 μg m⁻³ for hexanaldehyde.

For better data assessment air exchange rates measurements were also carried out using a tracer gas technique (NORTEST METHOD NT VVS 118). The temperature and relative humidity both indoors and outdoors were also recorded.

Additionally, a questionnaire was filled in, giving valuable information regarding sampling sites and activities which were taken during the sampling period. More specifically, it consisted of 3 sections. The first section gives general information about the sampling event (identification number, sampling period and the sampling team). The second section concerns the building information and is divided in the background information (building type, address, contact details of the involved participant), building characteristics and other relative information (location, dimensions, number of floors, year of construction, indication of any renovation, type of equipment/furniture/appliances placed indoors, type and frequency of cleaning, type of heating). Finally, the third section contains data on the sampling sites (location, room's dimensions, room’s materials, type (if any) of room's reconstruction/renovation, the presence of any mechanical ventilation, occupants’ smoking habits, consumer products use and frequency).

2.1. The sites

Table 1 summarizes the characteristics of the sites in terms of their use, the type of building material present inside, and the other expected emission sources.

Wooden varnished floors are found to be used in houses both in north and south Europe. Plaster walls and plastic water based paint are used practically everywhere. Plaster ceiling seems to be common both in the north and the south, since ceiling coverings (ceiling panels containing raff wool) are met only in the public buildings. Linoleum flooring is mainly used in Northern and Central European cities. Linoleum is commonly used in public buildings and schools. On the other hand, ceramic tiles are often used in Southern countries and especially in public buildings (including schools), as well as, wooden floor, carpets and rugs in houses. Furthermore, gypsum boards are met only in countries of Southern Europe and usually in houses. Additionally, furniture in all cases is constructed from similar wood based panels (MDF, covered particleboards or wood). Printing equipment is present in public buildings, as expected. In schools, the use of markers for painting was also observed. In this table, one can be see that the building materials existed in each measuring site can be identified as potential emission sources for all priority pollutants.

Table 2 summarizes the indoor activities and ventilation types in each tested building according to the recorded data in the questionnaires. Due to the differences in outdoor temperatures and climate between the northern and southern cities, mechanical ventilation systems are commonly used in southern countries (Greece, Nicosia) while natural ventilation is a common way in northern countries (Denmark, Ireland). In some cases, automatic ventilation is also utilized. The use of fans is met in kitchens and bathrooms, as well as, schools due to the vulnerability.
of the children. Additionally, smoking is prohibited in public building (including schools), however there are two reported cases of smoking habits in residences in Milan and Athens (Tables 1 and 2). Indoor activities, such as cleaning, cooking and use of disinfectants reported everywhere. Burning of incense and use of air fresheners are reported in houses. Finally, printers usually allocated in all public buildings. The tested of office in Milan public building (both campaigns), and in Nicosia (winter campaign), were not occupied during the sampling period.

2.2. Ventilation, humidity and temperature

Table 3 presents the range of the indoor temperature, relative humidity and air exchange rate, as were recorded during the field campaigns. It can be observed that ventilation rates show diversity and this trend is followed for humidity, as well. Temperature ranges from 11 to 25 °C in winter and 17 to 30 °C in summer, with an average value of 20 and 22 °C, respectively.

The air exchange rates were not expected to be the same in summer and winter. The air exchange rates in public buildings with natural ventilation ranged between 0.47 to 1.28 h⁻¹, close to the range for houses. However, air exchange rates for natural ventilation in schools may be much lower especially during summer period. Fig. 1 shows the air exchange rates which were measured in Athens, Dublin and Milan. As it can be observed, with the exception of school buildings, ventilation rates are higher during summer period especially in public buildings, as expected.

3. Results and discussion

3.1. VOCs concentration and emissions

Fig. 2 shows the indoor concentrations ranges of the measured compounds in all sites. The data show a considerable diversity due to the different indoor emission sources, ventilation rates, building type, and outdoor environment. Most of average concentrations remain below 50 μg m⁻³. Elevated concentrations have been observed for in houses and in public buildings. Houses seems to have the maximum concentrations for toluene (163.5 μg m⁻³), m,p-xylenes (177.4 μg m⁻³), d-limonene (159.4 μg m⁻³), acetone (308.8 μg m⁻³) and hexanaldehyde (113.3 μg m⁻³). On the other hand, it appears that schools have the lowest average concentrations for all measured compounds. Formaldehyde average concentrations are almost equal in each one of the three types of buildings. Indoor air pollutants such as benzene, formaldehyde and acetaldehyde show indoor concentrations ranges of 0.1–10.2, 5.8–62.6 and 0.7–41.6 μg m⁻³ respectively. The levels of formaldehyde remain below the WHO guideline of 100 μg m⁻³ for 30-minute average exposure (WHO, 2000). It is noticed that the levels of formaldehyde concentrations are comparable with the ones reported in the AIRMEX study (Kotzias et al., 2009).
The maximum concentration observed in Milan houses ($62.6 \mu g \text{m}^{-3}$) are greater than the values reported in Paris dwellings (Clarisse et al., 2003) and lower than the most recently obtained values for France (OQAI, 2006). Benzene concentrations were lower than the average annual limit of $5 \mu g \text{m}^{-3}$, which was

<table>
<thead>
<tr>
<th>Location</th>
<th>Public building</th>
<th>School/kindergarten</th>
<th>House 1</th>
<th>House 2</th>
</tr>
</thead>
</table>

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Table 3

<table>
<thead>
<tr>
<th>Location</th>
<th>Public buildings</th>
<th>School</th>
<th>House</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Air exchange rate ($h^{-1}$)</td>
<td>Relative humidity (%)</td>
<td>Temperature ($^\circ$C)</td>
</tr>
<tr>
<td>Winter</td>
<td>Summer</td>
<td>Winter</td>
<td>Summer</td>
</tr>
<tr>
<td>Public buildings</td>
<td>0.12–1.28</td>
<td>0.24–0.72</td>
<td>28–66</td>
</tr>
<tr>
<td>School</td>
<td>0.22–0.62</td>
<td>0.13–0.2</td>
<td>25–58</td>
</tr>
<tr>
<td>Houses</td>
<td>0.16–1.1</td>
<td>0.2–0.97</td>
<td>23–60</td>
</tr>
</tbody>
</table>

Air exchange rates ($h^{-1}$)

<table>
<thead>
<tr>
<th>Public buildings</th>
<th>School</th>
<th>House</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mechanical ventilation</td>
<td>0.12–0.66</td>
<td>0.62 ± 13%</td>
</tr>
<tr>
<td>Natural ventilation</td>
<td>0.47–1.28</td>
<td>0.2–0.4</td>
</tr>
</tbody>
</table>

Fig. 1. Winter and summer ventilation air exchange rates during summer and winter period in Athens, Dublin and Milan.
set by EU for outdoor air (EC, 2000), except for one house in Athens. The location of this house is a suburban area near heavy traffic (500 m) and smoking activities where taken place indoors. A fireplace and the gas appliances for cooking in operation, probably have led to the observed elevated concentration levels. The acetaldehyde and hexanaldehyde levels in most of the houses were similar to those found for in indoor environments in Strasbourg (Marchland et al., 2006). Acetone levels in Milan’s public building (hospital) seem to be slightly higher than those found in hospitals in China (Huixiog et al., 2006).

Fig. 3 gives the chemical classes that contribute mainly to indoor concentration levels. The chemical classes that have the highest contribution to the total measured VOC indoors vary from building to building and also among cities. For example, in Nicosia, the public building that was tested, presented values of total measured VOC of the order of 276.6 μg m⁻³. 46% corresponds to aromatic hydrocarbons and 45% to carbonyls and ketones. On the other hand, the tests that were carried out in the school showed 122.1 μg m⁻³ of total measured VOC and in contrast to the previous example exhibit 60% of carbonyls and ketones and 33% of aromatic hydrocarbons.

Table 4 presents the derived Indoor to Outdoor concentration ratios (I/O) for the measured substances. The I/O ratios which have found >10 are indicated with bold. For most VOCs there is a clear indication of strong indoor sources. For example, for formaldehyde we can observe values up to 16.4 whereas for acetaldehyde this value rises up to 31. Formaldehyde is expected to come from activities rather than building materials (Marchland et al., 2006). The maximum I/O ratio values for many substances seem to be associated with the presence of new building materials inside or with the use of certain consumer products during sampling period (i.e. nail polishes, painting material in schools etc.). An example of this situation is reported in Nicosia’s houses where acetone levels were elevated due to the extensive use of nail polishing (as recorded in the questionnaire). Benzene ratios do not seem to be substantially different than one, indicating no or weak indoor sources. The ban in the use of benzene which was set in 1978 by US consumer Product Safety Commission (CPSC) and the identification of benzene as a known human carcinogen by IARC (1982), have led to the elimination of its use as an ingredient in consumer products and materials, and currently is not intentionally added at all. Toluene indoor levels have a clear connection with carpet emissions; however furnishing could be also a source (Alevantis, 2003; Katsoyiannis et al., 2008). Furthermore, xylenes high indoor levels could be explained by considering their primary indoor source which is plaster on walls and secondly the use of adhesives (Katsoyiannis et al., 2008; Horn et al., 2007).

Fig. 4 presents the comparison between winter and summer indoor concentration levels according to each building type. Indoor concentrations of all substances were higher during winter period except for formaldehyde, acetaldehyde and hexanaldehyde, a trend which was also observed by Rehwagen et al (2003). This observation could be associated with the limited ventilation during winter period and/or other processes that may occur indoors (e.g. fireplaces in operation, use of personal care products). In the case of carbonyls, the seasonality may depend on living and meteorological conditions which are different between cities. On the other hand aromatics and terpenes seem to be reduced substantially during summer period, especially in public buildings. For xylenes in schools, seasonality does not play any key role, no significant differences in concentration levels have been observed.

Although there are numerous studies in which VOCs indoor concentrations were assessed in newly or renovated buildings, little is known concerning the behavior of these compounds in the same environment.
buildings after a certain period (Jarnstrom et al., 2006). Fig. 5 demonstrates the building construction time effect in a newly constructed building. This building was selected because of its age and the fact that it was not occupied during the first campaign (2 months). The second campaign was carried out after 21 months of construction. It is observed that the concentrations of aromatics are elevated during the first campaign, indicating that probably came from the building materials, as no other human activity occurred. In the contrary, carbonyls have shown an increasing trend, between the two measurements, probably due to the existence of other indoor sources apart from building materials contribution.

FLEC measurement results are comparable with other European and International emission data which are included in the BUMA Project Database (www.buma-project.eu). Fig. 6 shows the contribution of building material emissions to indoor air quality, as they were derived by the FLEC on site measurements. In figure the y-axis parameter is defined as \( \frac{1}{1 + 100} \) which is the measure of the excess concentration due to the emission rate \( E (\mu g m^{-2} h^{-1}) \) of the compound under consideration; \( A \) the area of the tested material (m\(^2\)); \( \lambda \) the air exchange rate (h\(^{-1}\)); \( V \) = room volume (m\(^3\)); \( C_{in} \) = the room measured concentration for the specific compound. It can be observed that the building material emissions contribution in many cases was significant and have reached...
40–50%. It is worth mentioning that benzene emissions were lower than 0.5 μg m⁻³ h⁻¹ and taking into account the indoor levels, no significant contribution came from building materials.

4. Conclusions

From the above discussion the following conclusions can be drawn:

1. The frequently used materials in buildings selected for the present study were found to be water based paint, plaster and particleboards.
2. The concentrations of VOCs show a considerable diversity due to the different indoor emission sources and outdoor environmental concentrations.
3. The chemical classes that contribute mainly in the indoor air concentrations are carbonyls and ketones followed by aromatics.
4. Winter indoor concentrations are, in general, higher than those reported in summer period probably due to the tightness of the buildings. However, the findings for carbonyls need further investigation.
5. The indoor excess concentrations of formaldehyde, acetaldehyde, acetone and d-limonene, indicate relatively significant indoor emission sources of these substances in all buildings. These emission sources in some buildings were exclusively coming from building material.
6. Present data indicate that emissions of hydrocarbons such as BTEX and terpenes from building materials could be insignificant after sometime. These findings need further investigation.

Acknowledgement

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ISO 16000-6, 2004. Indoor air- Part 6: determination of volatile organic compounds in indoor and test chamber air by active sampling on Tenax TA sorbent, thermal desorption and gas chromatography using MS/MS.


