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Investigation of formaldehyde sources in French schools using a passive flux sampler

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ABSTRACT

While indoor air quality issues have received increasing attention the past decades, detailed investigations of primary sources of indoor pollution are still difficult to carry out. There is a lack of analytical tools and measurement procedures to identify sources of pollutants and to characterize their emissions. Formaldehyde is a ubiquitous pollutant in indoor environments, which is known to lead to adverse health effects. This study describes a measurement procedure to apportion formaldehyde emissions from building and furnishing materials and presents a source apportionment study performed in French public schools. More than 29 sources of formaldehyde were characterized in each investigated classroom, with higher emissions from building materials compared to furnishing materials. Formaldehyde emission rates measured using passive flux samplers (PFS) range from 1.2 to 252 $\mu g/m^2/h$, highlighting several strong emitters made of wood products and foam. Interestingly, the ceiling was identified as the main source of formaldehyde in most classrooms. Measured emissions and air exchange rates were constrained in a mass balance model to evaluate the impact of formaldehyde reduction strategies. These results indicate that formaldehyde concentrations can be reduced by 87–98% by removing or replacing the main source of emission by a less emissive material and by increasing the air exchange rate to 1 h^{-1}. In addition, an intercomparison of total emissions calculated from (1) PFS measurements and from (2) measured formaldehyde concentrations and air exchange rates indicate that an unidentified sink of formaldehyde may exist in indoor environments.

1. Introduction

People from industrialized countries spend approximately 90% of their time in indoor environments such as housing and office buildings [1–3], being exposed to elevated concentrations of indoor air pollutants. In light of these observations, indoor air quality has received increasing attention over the past decades to characterize both the nature and the concentration levels of indoor pollutants. There is a general agreement within the scientific community that chronic exposures to indoor pollutants can lead to pathologies such as asthma [4] and cancers [5].

Indoor environments are enriched in Volatile Organic Compounds (VOCs) [6], with indoor concentrations being 2–100 times higher than outdoor [7]. Reasons usually highlighted to explain elevated concentrations of indoor VOCs are related to an increasing use of manmade materials for building and furnishing, the use of cleaning supplies, unvented combustion processes such as gas stoves, and low air exchange rates due to recent energy saving politics [8].

Formaldehyde (HCHO) is one of the most frequently detected VOC in indoor environments as well as the most abundant aldehyde. This compound has been classified as carcinogenic in 2004 by the International Agency for Research on Cancer (IARC) [5]. A study conducted from 2003 to 2005 by the French observatory of indoor air quality (OQAI) in 567 accommodations reports a mean concentration of formaldehyde of 19.6 $\mu g/m^3$ [9], which is approximately 10 times higher than outdoor. While this value is lower than the upcoming French regulation threshold of 30 $\mu g/m^3$ for long-term exposure [10], it was highlighted that 22% of these accommodations exhibit concentrations higher than 30 $\mu g/m^3$. Studies performed in other countries indicate similar results in Japan [11]

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and Canada [12] with mean formaldehyde concentrations of 17.6 μg/m^3 and 33.2 μg/m^3 respectively, lower concentrations in Sweden with a mean value of 8.3 μg/m^3 [11], and higher concentrations in the USA with a mean value of 55 μg/m^3 [6].

To reduce indoor concentrations of formaldehyde, as well as other VOCs, there is a need to identify the main sources of emission and to quantify the contribution of each source to the emission budget. Building and furnishing materials are known to emit a large range of VOCs, including formaldehyde, and to significantly enhance their indoor concentrations [13]. In this context, several analytical devices were developed to perform in-situ measurements of VOC emissions from solid materials, including the Field and Laboratory Emission Cell (FLEC) and Passive Flux Samplers (PFS).

While FLEC apparatus can be used to directly measure emission rates in indoor environments, most studies involving a FLEC focused on laboratory measurements to characterize building and furnishing materials and to evaluate the impact of air velocity, temperature, and relative humidity on emission rates [14–20]. On-site studies are not as frequent because this technique requires cumbersome equipment such as cylinders of zero air, pumps, and flow controllers [15,17,21]. In addition, a FLEC device is limited to measuring emission rates from one material at a time while indoor environments are usually built using several tens of different materials. The use of several FLEC to perform an exhaustive characterization of indoor emissions would be too cumbersome, time consuming, and expensive.

Passive sampling is more suitable for in-situ measurements of emissions. This technique uses inexpensive samplers and allows multiple samplings at a time by multiplying the number of samplers. Recent studies showed the feasibility of measuring emission rates of a few VOCs using passive samplers [22], including formaldehyde [23–25], toluene and pinenes [26], and semi-volatile organic compound such as phthalates [27].

A PFS was developed at Mines Douai to quantify formaldehyde emissions from building and furnishing materials [23]. This PFS was deployed in 24 unoccupied student rooms [28], showing that most emissions were in the range 1.2–12.1 μg/m^2/h. A few strong emitters were identified, with emissions as high as 21.3–131.3 μg/m^2/h. This study highlighted the potential of this new analytical tool to apportion the main sources of formaldehyde in indoor environments, and therefore to improve indoor air quality.

A national campaign was conducted in France from 2009 to 2011 in 316 day-care centers and primary schools to provide a comprehensive picture of formaldehyde and benzene concentration levels in French buildings receiving children [29,30]. This study was mandated by the French ministry of ecology, sustainable development, transport and housing and included the technical support from the French national institute for industrial environment and risks (INERIS) and the French scientific and technical center for building (CSTB). An average formaldehyde concentration of 15.7 μg/m^3 was inferred from two measurement periods of 4.5 days (summer and winter). This study showed that formaldehyde concentrations are acceptable for 85% of the buildings considering the upcoming French regulation value of 30 μg/m^3 for long-term exposure [10]. However, this study also highlighted that eight buildings were exhibiting formaldehyde concentrations close to 50 μg/m^3, requiring further measurements to identify emission sources.

The French central laboratory for air quality monitoring (LCSQA) and Mines Douai carried out a diagnostic of formaldehyde sources in these eight polluted buildings. This publication describes the basics of a methodology designed to identify formaldehyde emitters and to quantify their contribution to the formaldehyde budget, based on the use of the PFS mentioned above. Emissive materials were categorized into building and furnishing materials to differentiate these two classes of emission. This study also makes use of chemical mass balance equations to evaluate the relevance of strategies of formaldehyde reduction.

2. Measurements

2.1. Description of the sites

Measurements were carried out from June–July 2011 inside four buildings located in Picardie (school 1), Provence-Alpes-Côte d'Azur (schools 2–3), and Pays de la Loire (school 4), and during May 2012 inside four additional buildings in Limousin (school 5), Centre region (school 6), and Franche-Comté (schools 7–8). Only one of the most polluted rooms was investigated for each site.

Details about the sites are given in Tables 1 and 2, including formaldehyde concentrations measured during the 2009–2011 national campaign. As shown in Table 1, more than 29 potential sources of formaldehyde were identified in each room. In order to correlate measured emission rates to total emission rates calculated from measured concentrations of formaldehyde and air exchange rates (Section 2.2.3), it was decided to remove school consumables such as books and drawing equipment, whose emissions could not be measured in this study. The rooms were left closed for at least 12 h before the measurements to make sure that formaldehyde concentrations had reached a steady-state concentration.

2.2. Methodology designed to apportion emission sources of formaldehyde

2.2.1. Material

The methodology designed to investigate the formaldehyde budget involves five steps:

(i) Visual identification of different materials & measurement of covered surface areas
(ii) Measurement of air exchange rates
(iii) Measurement of indoor and outdoor formaldehyde concentrations
(iv) Measurement of formaldehyde emission rates for each identified material
(v) Evaluation of strategies of formaldehyde reduction through the use of chemical mass balance equations

Measurements of air exchange rates were performed according to ASTM E 741-00 [31]. Carbon dioxide (CO_2) was injected at the center of the room using a gas cylinder of pure CO_2 (Air liquid) and a fan was used to speed-up the mixing. Initial mixing ratios of CO_2 were in the range 3000–5000 ppm. Temporal decays of CO_2 were

Table 1
Description of the measurement sites.

School#	Sampling date	Type of school	Type of area	Formaldehyde concentration[a] (μg/m^3)	Number of identified sources
1	June–July 2011	Primary	Periurban	54.5	36
2			Rural	59.4	39
3			Periurban	52.7	45
4			Industrial	42.6	45
5	May 2012		Urban	46.4	39
6			Rural	52.4	42
7		Day-care	Urban	46.6	29
8		Primary	Urban	43.3	31

[a] Concentrations of formaldehyde measured during the 2009–2011 national campaign.