The future of passive techniques for air change rate measurement

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ABSTRACT

Ventilation is critical in interpreting indoor air quality (IAQ), yet few IAQ assessments report ventilation rates; even when they do, the measurement method is often not fully described. Most ventilation assessments use a tracer gas test (TGT) to measure total air change rate. In a TGT, the indoor air is marked with an easily identifiable gas (tracer) so that the air exchange rate can be inferred by monitoring the tracer's injection rate and concentration. Passive sampling (adsorptive/absorptive samplers) is mostly preferred to monitor tracer concentration for its simplicity, practicality and affordability. Such samplers are commercialized by a range of companies and are widely used in IAQ studies to assess pollutants levels. Currently used passive TGTs present some limitations: inadequate tracer gas, disconnection from IAQ analysis (providing ventilation rates in a different time-scale than the pollutant concentrations), lack of verifiability/reproducibility. Thus, this paper proposes a new approach on the passive TGT method, using as tracer a suitable gas (non-reactive, insensitive, unique, measurable and safe) which can be co-captured and co-analysed using commercial passive samplers employed in IAQ studies. A literature review was carried out in pursuit of such a gas. Considering that the most relevant compounds in IAQ studies are volatile organic compounds (VOCs), which are sampled separately from inorganic pollutants, the gases considered as possible tracers were the VOCs capable of being captured by the samplers commercialized by Radiello®, 3M and Gradko. They are composed by activated charcoal, which captures all VOCs in the targeted molar mass range by adsorption. The info-sheets for these samplers were consulted. The option currently under consideration is the solvent 2-butoxyethyl acetate (EGBEA), a low-reactivity glycol ether mentioned by Radiello® in their VOC CS_2-desorption sampler info-sheet. Although EGBEA is present in various household products, several field studies show that its background indoor concentration is usually very low or negligible. Regarding human health, EGBEA has generally low toxicity and has not been linked to any chronic effects. A preliminary field test was carried out in order to check EGBEA's measurability: Radiello® samplers were used to measure its concentration in one room before and after the placement of a beaker containing the solvent. Results showed insignificant background EGBEA concentration (EGBEA mass desorbed from the sampler placed before the beaker placement was lower than from a blank sampler). The relatively low volatility of EGBA (0.23 g evaporated in 4 days) did not hinder its measurability by the Radiello® sampler, which measured a 4-days average EGBEA concentration of 14.1 $\mu g\ m^{-3}$. Further test chamber and field tests will be performed in order to determine the sampler's accuracy in measuring known EGBEA concentrations and the actual applicability of this substance as tracer in TGTs.

KEYWORDS

Ventilation, indoor air quality, tracer gas test, passive sampling.

1 INTRODUCTION

Air pollution found in indoor environments is of great concern, since people, especially in urban areas, tend to spend the majority of their time inside buildings. Within buildings, there are numerous and significant known sources of substances with high potential to cause adverse effects to human health, including by-products of combustion, aldehydes, volatile and semi-volatile organic compounds and biological contaminants (Godish, 2005). This, allied to the typically low air exchange rates in this type of environment, considerably increases the concentration of pollutants in the indoor air. Exposure to air pollution in indoor environments may be responsible for almost 2 million premature deaths in developing countries and for about 4% of the global burden of disease (Bruce et al., 2000; Godoi et al., 2009).

A crucial factor in determining the accumulation of pollutants in indoor environments is ventilation (EPA, 1994). The impacts of ventilation on the health and comfort of building occupants has long been recognized in ventilation standards and regulations, and ventilation rates are also closely related to the energy efficiency of a building (Persily and Levin, 2011; Persily, 2015). Evidence shows that better ventilated buildings tend to present lower concentrations of indoor air contaminants (Flemish Government, 2012; 2014). As the airtightness of a building increases, the total ventilation becomes even more important. Increased airtightness in buildings is becoming more and more common with the rise of the energy-efficient buildings concept, such as the passive building (Foster et al., 2016).

However, despite the fact that ventilation is critical in interpreting indoor air quality (IAQ) measurements, only few IAQ field studies measure ventilation rates or otherwise characterize the ventilation design of the study building (Persily, 2015). Moreover, even when ventilation rates are reported, in many cases the measurement approaches are not described in sufficient detail to evaluate their quality or applicability to the study design (Persily and Levin, 2011).

Given the importance of ventilation in estimating sources strength and determining appropriate remediation actions if needed, it is crucial that IAQ assessments provide information on basic ventilation parameters (e.g. building ventilation type, designed air change rate) as well as on actual air change rate values, by means of a reliable and reproducible measurement method. Moreover, efficient ventilation assessment is even more important when taking into account the high costs to install and maintain most ventilation systems, being therefore highly desirable to effectively monitor such systems in order to ensure that they deliver the required air change rates.

Methods currently used for measuring ventilation rates either involve direct flow rate measurements at vent holes combined with a pressurization test, or a tracer gas dilution/dispersion test (TGT). The first type is used only under very specific circumstances, such as in extremely airtight buildings where all airflows occur mechanically in ductwork (Persily, 2015). In these cases, ventilation is restricted to that generated by the mechanical system, making its measurement very straightforward.

Therefore, most ventilation assessments use TGT as a method to measure the total (i.e. intentional plus unintentional) outdoor air change rate of a building/indoor space (Persily, 2015). The fact that this type of measurement can be carried out in occupied buildings, during normal occupants' activity, grants the TGTs not only convenience but also a greater accuracy, since it takes into account the large effect occupancy has on a building's air change rate.

However, most times the ventilation measurement methods provide instantaneous results (due to the use of online monitors), which are in essence very variable over time. The concentrations of indoor pollutants (also variable over time and significantly influenced by air change rate) are usually measured over a longer period using long-term sampling techniques that report time-averaged values, i.e. the link between the IAQ and the ventilation data is not direct, hindering their comparability.

Thus, an inexpensive TGT conceived to be executed in conjunction with common IAQ analysis, i.e. which provides ventilation rates in the same time-scale as the pollutant concentration measurements, would be more appropriate than the ones currently used. Furthermore, considering that the presence of occupants highly impacts the IAQ of any indoor environment, it is also important that the new method is suitable for use during normal occupancy. Thus, the TGT must not cause any type of disturbance and the employed tracer gas must be scientifically proven harmless to the occupants' health, respecting susceptible populations. Also, a highly desirable characteristic for this new method is the non-dependence on electricity, which lowers the utilization costs and broadens the range of buildings where it can be applied.

Taking all these characteristics into account, the best suitable option is to develop a TGT which relies entirely on passive techniques, both in the tracer gas emission and sampling. Therefore, this project aims to develop a reliable and reproducible TGT using as tracer a substance that can be co-captured and co-analyzed with standard passive samplers used for common IAQ assessments (e.g. Radiello, Gradko, or 3M). This new TGT will be temporally and analytically in line with IAQ measurements, affordable, accurate and safe for use during occupancy. Also, the tracer gas emission and behavior must be as independent as possible from the room conditions (e.g. temperature and humidity), since in field experiments such conditions are essentially uncontrollable.

An additional challenge in IAQ assessments is measuring internal airflows in the building. Most studies include only one room, often ignoring the most intensively occupied rooms and rooms where most pollutant sources are located. Capturing the interactions between the sources in these different rooms and their dispersion/interaction in buildings is still a challenge for indoor air research. Therefore, a second objective of this research is to diversify the tracer source so it can be used to capture internal and external airflows.

2 MATERIALS AND METHODS

From an IAQ point of view, volatile organic compounds (VOCs) are the most relevant contaminants, since they are ubiquitous in indoor spaces, specially occupied ones, and several of them are knowingly harmful to human health (Uhde and Salthammer, 2009). This group of compounds is truly extensive and its constituents vary greatly in characteristics and properties. In IAQ assessments, such gases are sampled separately from inorganic pollutants due to their different physicochemical characteristics. The vast majority of researchers utilize passive sampling to measure average VOCs concentration in common IAQ studies, for its simplicity, accuracy, practicality and suitability for use during normal occupancy.

Considering that a main characteristic for the new TGT is that the time scale for the ventilation measurements matches that of the concentration measurements, the best option would be to select as tracer a gas which can be captured simultaneously by the commercial passive VOC samplers. This approach would simplify the process of ventilation measurement in IAQ studies as well as represent an economy of time and resources for the required lab analysis, since only one sample would provide all the information needed to infer both the air change rates and the indoor VOCs levels simultaneously. Thus, the gases considered as possibilities for use as tracer were the VOCs capable of being captured by the passive VOC samplers commercialized by Radiello®, 3M and Gradko, the most widely employed in common IAQ assessments. These samplers are composed by activated charcoal, in which organic gases are captured by adsorption. This process is essentially non-specific, meaning that any VOC in the targeted molar mass range may be captured and later analyzed in lab if the adequate extraction process is used.

2.1 Selecting a VOC for use as tracer gas

Considering that a good tracer gas must be suitable for use during normal occupancy (as occupancy greatly affects IAQ), the substance must be safe for occupants. As a first step, the information sheets for the commercial passive VOC samplers were consulted. Radiello® and 3M samplers capture VOCs in the range C2-C12, and Gradko samplers capture compounds up to C28 (3M, 2017; Gradko, 2012; Radiello, 2007). Among the numerous VOCs in this range, paraffins and isoparaffins initially stood out as possible candidates for use as tracer gas.

Paraffins are linear acyclic saturated hydrocarbons (alkanes), and isoparaffins are the branched products of paraffinic isomerization (iso-alkanes). First four alkanes (methane to butane) are colorless, odorless gases at ambient conditions, C3 to C17 are colorless, nearly odorless, volatile liquids and higher alkanes are wax-like solids. Alkanes are the simplest class of organic compounds, extremely stable and inert, because they contain only carbon and hydrogen atoms (small electro negativity difference) and have no functional groups (Bano, 2007). According to an extensive review of toxicity studies on alkanes carried out by HSPA (2015), C5-C15 alkanes present no significant toxicological effects to humans, with the exception of n-pentanes, trimethylpentanes and n-hexanes. Alkanes of >C15 have lower vapor pressures, hindering their application as passive tracer gases (HSPA, 2015). Inhalation studies in rats indicate that C8-C15 isoparaffins can cause kidney effects. However, such effects are species-specific and not a concern for human health (Carrillo et al., 2013). Nevertheless, adding that to the fact that isoparaffins are somewhat more reactive than paraffins due to their branched nature, the best options for tracers are considered to be among the paraffins group.

There is however a major problem regarding the applicability of paraffins as tracers: since these compounds are largely present in the composition of a range of household products, their typical background concentration is considerably high in most indoor environments. An ideal tracer should be able to be easily recognized from all other constituents of air, i.e. it should not be a normal constituent of air in the ambient it is being placed. Ideally, the background concentration should be negligible, but a tracer with non-zero background may be used provided that such background is stable and that the additional tracer concentration is significantly larger than it. The background concentration should also be as low as possible in order not to oversaturate the passive sampler, risking a backdiffusion effect (when the adsorbed mass of analyte reaches the maximum amount allowed by the adsorbing medium capacity, leading to an underestimation of the actual environmental concentration).

A second option for a tracer gas is the solvent 2-butoxyethyl acetate (also called ethylene glycol monobutyl ether acetate or EGBEA, CAS 112-07-2), a compound from the group of glycol ethers mentioned by Radiello® in the VOC CS_2-desorption sampler info-sheet. Glycol ethers comprise a large family of more than 80 chemical compounds. The two most common series of glycol ethers are E-series ($ROCH_2CH_2OH$), which are the reaction products of ethylene oxide with various alcohols, and the P-series ($ROCH_2CH_2CH_2OH$), which are the products of propylene oxide with various alcohols (Zhu et al., 2001). Due to their excellent solvency, chemical stability and water compatibility, glycol ethers are good solvents for many applications (Archer, 1996). These chemical compounds are amphiphilic, completely miscible with water and with a large number of organic solvents and present relatively low volatility. Around 30 different derivatives of glycol ethers have been synthesized and extensively used as solvents by different sectors of the industry (Plaisance et al., 2008). With an increased number of so-called water-based consumer products (e.g. in paints, lacquers, resins, oils and fat) in the last four decades, they have gained more and more importance. In common households, they are mainly found in liquid cleaning products, paints, inks, varnishes, polish removers, surface coatings, cosmetics, etc. (Fromme et al., 2013).

During recent years, it became readily apparent that four E-series glycol ethers (2-methoxy ethanol, 2-ethoxy ethanol and their acetates) are potent reproductive and developmental toxicants and they were subject to French regulations to forbid their presence in cosmetics and drugs, and to limit their concentrations above 0.5% in consumer goods (Plaisance et al., 2008). Their industrial use is still permitted, as long as they carry a label stating their potential health effects. Consecutively they have been progressively replaced by other glycol ether derivatives (e.g. methoxy propanol, butoxy ethanol) which have seen a growth of production in the last years, so nowadays many research activities are performed in order to assess their potential toxicity on human health.

Amongst the currently used glycol ethers, EGBEA, which is the acetate of 2-butoxyethanol (EGBE), is a relatively high production volume compound. Its estimated production volume in the European Union is about 1.2 x 104 tons y^{-1}, a value about 10 times lower than EGBE (SCHER, 2006). Although EGBEA is present in various household products (ATSDR, 1999), its background indoor concentration is usually very low. A national survey for IAQ carried out in 490 French dwellings found EGBEA concentrations below the detection limit in 97% of the assessed houses (Billionnet et al, 2011). A similar assessment, carried out as part of the HABIT'AIR Nord – Pas de Calais program, monitored the presence of glycol ethers in 60 homes located in northern France, finding no trace of EGBEA in any of the assessed houses (Plaisance et al., 2008). More recently, Derbez et al. (2014) performed a field survey in 7 newly built energy-efficient houses, also in France, measuring a range of IAQ indicators and environmental parameters both before and during the first year of occupancy; EGBEA was not detected in any sample from any of the assessed houses. Regarding human health, EGBEA has generally low toxicity and has not been linked to any chronic effects (ATSDR, 1999; ECETOC, 2005; SCHER, 2006).

2.2 Preliminary test for measurability

A preliminary test was carried out in order to test EGBEA's measurability by Radiello® samplers in simplified field conditions. The Radiello® samplers were used to measure this compound's concentration in one room before and after the placement of a recipient containing the solvent. The room used was an office in VITO which was at that time not being used (unoccupied), although fully furnished (two desks with two chairs, two PCs, one wall cabinet and plastic blinds in the windows). Three sampling cartridges were used in total for this test: one was used as a lab blank, i.e. analyzed right out of its package, no exposition; a second cartridge was used to measure the background EGBEA concentration in the office; the third and last cartridge was used to measure the air concentration of EGBEA after the placement of a source of this solvent in liquid phase. This so-called source consisted of a simple glass beaker filled with approximately 11 ml of EGBEA (9.536 g) left uncapped so the solvent could freely volatilize into the room air, acting therefore as a completely passive source. Both these samplers were exposed in the room for a period of consecutive 4 days. Figure 2 shows the setup of the experiment.

3 RESULTS AND DISCUSSION

Results from the preliminary experiment are presented in Table 1.

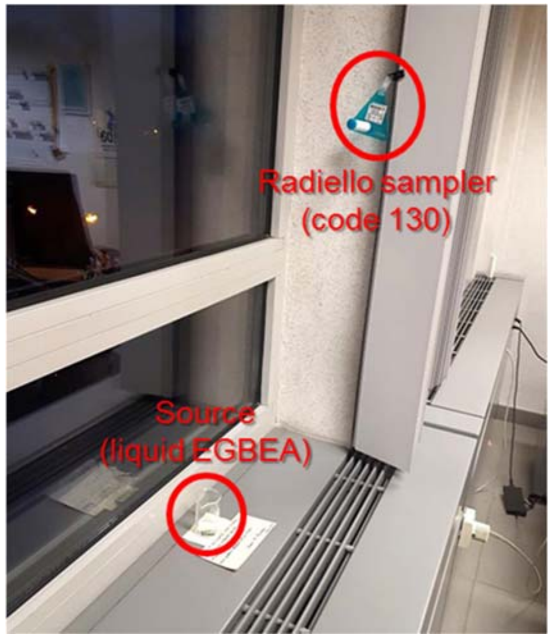

Figure 1: Preliminary test setup

Table 1: Preliminary field test results

Sample	Sampling start	Sampling finish	EGBEA mass
Lab blank	-	-	0,067
Background	15/01 – 17h25	19/01 – 17h39	0,059
With EGBEA source	19/01 – 17h40	23/01 – 17h34	3,324

Based on the values presented in Table 1, the background EGBEA concentration in the assessed room can be considered insignificant: the EGBEA mass desorbed from the cartridge placed before the recipient placement (background) was very similar to the mass desorbed from the blank cartridge, even slightly lower, meaning that virtually no EGBEA was adsorbed onto the cartridge. Furthermore, the relatively low volatility of EGBA (0.23 g evaporated in 4 days) did not hinder its measurability by the Radiello® sampler: the cartridge placed in the room after the addition of the EGBEA source adsorbed an EGBEA mass two orders of magnitude higher than the blank and background cartridges. The calculated average concentration in air corresponding to the adsorbed mass after 4 days exposure to the EGBEA source was 14.1 µg m^{-3}. The results observed in this primary experiment can be interpreted as clear indications of the potential suitability of EGBEA as tracer for the purposes of a new TGT approach: its background concentration in the assessed room was negligible and it was successfully captured by a commercial VOC sample at a measurable level.

4 CONCLUSIONS

A TGT employing as tracer a harmless gas which is not commonly present in background indoor air, can be passively captured by commonly used commercial passive samplers and then analysed along with common IAQ pollutants, using the same analytical lab procedure, is close to an ideal method to measure total ventilation. After several considerations, EGBEA solvent stands out as possible candidate: it can be captured by commercial VOC passive samplers commonly used in IAQ studies, presents no relevant effects on human health and has a typical very low presence in indoor environments. A preliminary test indicated a good measurability of EGBEA by a commercial passive sampler in simplified field conditions. Further test chamber and field tests will be carried out in order to determine the actual applicability of this substance as tracer in TGTs.

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