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The Effects of Cement-Based and Cement-Ash-Based Mortar Slabs on Indoor Air Quality

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Declaration of interest: none

Abstract

The effects of emissions from cement-based and cement-ash-based mortar slabs were studied. In the latter, 30 % of the cement content had been replaced by sewage sludge ash. They were tested singly and together with either carpet or linoleum. The air exhausted from the chambers was assessed by means of odour intensity and chemical characterisation of emissions. Odour intensity increased with the increased exposed area of the slabs. It did not differ significantly between cement-based or cement-ash-based mortar and neither did the chemical composition of the exhaust air. A significant sink effect was observed when linoleum was added to any of the two slabs examined. The sink effect increased as the exposed area of the slabs was increased. The odour intensity of the mixture of the slab and linoleum was lower than the intensity of odour produced by any of the two materials when tested singly. A plausible explanation for this effect was that the mortar slabs adsorbed the organic acids that were emitted at a high rate from linoleum, mortar being strong base. The same sink effect was also observed when the mortar slabs were exposed together with carpet but it was much smaller because the carpet emitted smaller quantities of acids. The total concentration of organic compounds measured was not appreciably different when the slabs were tested alone or together with linoleum or carpet. Considerable differences in the concentration of organic compounds were however observed when the total concentration of each functional group was calculated and compared.

Key Words

Cement-based mortar; Cement-ash-based mortar; Perceived air quality; Chemical measurements; Emissions; Sink effect

1. Introduction

The development of building new technologies is currently being driven by the demands of energy conservation, waste management and economic prosperity. In these areas, building development seeks to maintain high living standards and positive socio-economic trends while also seeking to minimize the irreversible destruction of nature. As a result, in recent decades the development of innovative and high-performing building materials has been one of the major drivers for the construction industry. The Industrial Technologies arm of the Research and Innovation Department of the European Commission estimated that 70 % of all new product innovation today is based on materials with improved properties ("Promotional material - Research & Innovation - Key Enabling Technologies - European Commission," n.d.). The new materials offer potentially huge benefits for the building industry in terms of cost and performance. At the same time, it must be ensured that the new materials do not create new risks in the form of negative effects on indoor air quality (IAQ) that results in discomfort and a higher risk of health problems. IAQ is one of the major concerns in buildings because it can impact the health, comfort, well-being, and cognitive performance of building occupants (US EPA, n.d.; Wargocki et al., 1999). Short and long-term exposures of building occupants to pollutants emitted from building materials or pollutants produced due to chemical transformation of the emitted pollutants can produce sensory nuisance (e.g. Wargocki et al., 2004) and can lead to the development of building related health symptoms such as fatigue, eye irritation, itchiness, nose and throat irritation, nausea, headaches, dizziness, skin irritation, rashes (Fucic, 2012). In particular cases, chronic and severe diseases can be caused or exacerbated. These include asthma, reproductive impairment, disruption of the endocrine system, cancer, impaired child development, birth defects, and immune system suppression (Fucic, 2012). Consequently, new and/or alternative materials should always undergo a comprehensive assessment process, including an examination of the emissions of potentially toxic compounds. These aspects are not typically the concern of the building industry and are not considered exhaustively when new materials are being developed.

Among many new alternative building materials there is an ash mortar. In ash mortar, fly ash is used to partially replace cement. Mortar that is a structural material itself, or is used in concrete production, usually contains cement, sand and water. Cement is a crucial ingredient of the mortar and acts as a binder. Cement manufacturing is economically and energetically a very demanding process. About 5 % to 7 % of all global anthropogenic carbon dioxide (CO₂) emissions are released from plants manufacturing cement and 900 kg CO₂ is emitted to the atmosphere during production of one ton of cement; this is a nearly 1:1 ratio (Chen et al., 2011). Any improvements in cement production will thereby bring tangible effects and there have been significant steps towards improving the sustainability of concrete production by finding cement substitutes (Carp et al., 2004). Cement is additionally the second most consumed substance in the world by weight, second only to water, with global cement production at 4.2 billion tons in 2016 (“USGS - Minerals Information: Cement,” n.d.). It is estimated that annual concrete consumption is at 3.8 billion m³ worldwide (as stated by Cement Association of Canada). Thus even a few percent of cement substitution by an alternative material would bring considerable benefits. This substitution will additionally reduce the need to mine the virgin raw material. Among other benefits there is also a reduction in the need for landfilling.

For decades the industry incorporated fly ash from coal combustion power stations as a supplementary cementing material (Chandra, 1997). However, very limited information was available on emissions from mortar containing fly ash. A Danish study (Bødker, 2006a) compared the emissions from cement-based concrete and cement-ash-based concrete. The results showed that concrete that was cast in a mould that had been sprayed with mineral-oil emitted hydrocarbons which could be eliminated by using vegetable-based oils, while concrete containing fly ash from coal combustion power plants emitted ammonia. The emission rates of ammonia were rather low, and they were not thought to create any considerable risk of reduced indoor air quality. Actually, it was also shown that the cement-based concrete emitted ammonia. Neither of the two concrete types were sources of amines, alcohols or aldehydes. The report from the project mentioned that sensory assessments were made of the emissions of cement-based concrete with an area of 0.4 m². They showed that they produced moderate to weak odour intensity that decreased over time.

Besides the benefit of cement replacement, fly ash acquired from coal combustion power stations might possess some air cleaning properties similar to those of activated carbon. Peloso et al. (1983) and Rovatti et al. (1988) investigated whether fly ash from coal combustion could be used as an adsorbent material for toluene vapour. The experiments showed that coal fly ash may indeed act as a possible adsorbent of toluene and more generally, of other organic gases and vapours, but only

after suitable activation treatment, which specifically included particle aggregation and thermal activation. These processes resulted in the coal ash acquiring adsorbing properties that were comparable with the usual but more expensive activated carbon. Whether adding fly ash that has undergone activation treatment to cement mortar can result in the product having air cleaning properties has not yet been investigated.

Only a few experiments have examined the interaction between mortar and air pollutants. Most of them examined the possibility of air cleaning by adding photocatalytic additives, specifically TiO_2 , to concrete (e.g. Calia et al., 2016; Carp et al., 2004; Chen et al., 2011; Kolarik and Toftum, 2012; Nath et al., 2016; Sugrañez et al., 2013). Some studies focused on the interaction between cement-based concrete with various pollutants such NO_x , CO_2 , and SO_2 (e.g.-Bødker, 2006b; Decio et al., 2009; Iizuka et al., 2011; Martinez-Ramirez et al., 2002; Moghtaderi et al., 2012; Ramakrishnan and Orlov, 2014). Decio et al. (2009) examined the interaction of three mortar mixtures with volatile organic compounds (VOCs). Samples included common cement mortar, a dehumidifying salt resistant mortar and a cement-free mortar containing a natural binder (pozzolana). The sorption ability of the mortar was examined by injecting a mixture of VOCs (methanol solution composed of several VOCs: benzene, heptene, heptane, o-m-p xylene, toluene, ethyl benzene, cumene, ethylene glycol monobutyl ether and diethylene glycol monobutyl ether acetate, carene, isolongifolene) into the steel chamber, where a specimen of cement mortar was present. The results revealed that all three mortar types reduced the total concentration of VOCs (TVOC) during a 24-hour test; during this period the chambers were flushed with nitrogen at the rate of $20 \text{ cm}^3/\text{h}$. Specifically, in the case of common mortar, the TVOC concentration decreased from an initial concentration of approximately $4,500 \mu\text{g}/\text{m}^3$ to $1,000 \mu\text{g}/\text{m}^3$. In addition, the mortar containing natural pozzolana caused the largest reduction in TVOC concentration compared to the other two mortars. This effect could be due to the different composition of the mortars and the use of a natural material that affects the final structure of the mortar, as the final product has higher porosity: the pictures from the scanning electron microscope revealed that a greater superficial area was available for adsorption. Decio et al. (2009) postulated that various reductions of TVOC in the presence of mortars can be caused not only by differences in porosity and in the physical characteristic of the mortars but also by chemical reactions. This postulation was based on results that showed that the VOCs detected in the presence of mortar containing natural pozzolana differed from those detected in the presence of other mortars. However, no details of the examinations and procedures were provided in the conference paper by Decio et al. (2009). There are also studies that investigated interaction of mortar with outdoor pollutants under conditions typical for outdoor air such as Bødker (2006a, 2006b); Johannesson and Utgenannt

(2001); Ramakrishnan and Orlov (2014b). As they are outside the scope of the present work they are not described here though it is worth mentioning that they showed that mortar has the potential to reduce levels of NO_x .

The overall objective of the present work was to compare mortar samples with standard composition (cement-based mortar) with alternative mortar samples in which cement had been partly replaced by sewage sludge ash (cement-ash-based mortar). The study formed a part of the ZeroWaste initiative at the Department of Civil Engineering at the Technical University of Denmark (DTU) ("ZeroWaste Byg," n.d. http://www.byg.dtu.dk/innovation_og_myndighedsbetjening/zerowaste-byg). The purpose of this initiative was to examine and potentially develop building materials in which natural raw materials are replaced by secondary resources, including waste materials. Besides comparing the quality of air polluted by emissions from two types of mortar samples, it was additionally examined whether indoor air quality would change if mortar slabs were present together with either linoleum or carpet, two typical indoor floor surface materials. The intention was to determine whether such concrete slabs exhibit any air cleaning properties. The two specific research questions investigated in the present experiments were thus as follows: (1) Do emissions from mortar with and without ash have the same impact on perceived odour intensity? and (2) Does the odour intensity change when the mortar slabs are present together with samples of carpet or linoleum? Sensory assessments and chemical measurements were used to answer these questions. The former is supplementary to the latter especially as it is difficult to predict the impact of pollutants emitted by materials indoors on air quality as it is perceived by occupants of buildings (Knudsen et al., 1997, 1999).

2. Methods

2.1 Materials

Two mortar mixtures were cast according to the standard DS/EN 196-1 (2005); a standard cement-based mortar made of a cement-sand mixture (further abbreviated as CM) and a cement-ash-based mortar (abbreviated as AM) where 30 % of the cement content had been replaced by sewage sludge ash originating from the incineration power plant located nearby in Copenhagen, Denmark. Newly cast mortar panels were air-dried at 45°C after hardening for 28 days in a water bath, and afterwards stored in airtight foil; they were unpacked prior to being loaded into test chambers made of glass.

Linoleum (abbreviated as Li) and carpet (abbreviated as Ca) were chosen as two typical materials representing indoor wall-to-wall floor coverings. Both of them are typical materials used indoors and both emit elevated concentration of VOCs into

the air. Their emissions can also participate in chemical transformations that result in new indoor air pollutants (Bako-Biro, 2004; Gunnarsen et al., 1994; Jensen et al., 1995a, 1995b; Johnsen et al., 1991; Sakr et al., 2006; Wargocki, 1998; Wargocki et al., 1999; Wolkoff et al., 1993). The linoleum (with coloured patterns) used in the present experiment was bought in the early 2000, cut into pieces and used in many experiments (Bako-Biro, 2004; Clausen and Wyon, 2008; Kolarik and Toftum, 2012; Kolarik and Wargocki, 2010; Melikov et al., 2013; Sakr et al., 2006; Wargocki et al., 2010). It had never been installed on the floor in a building. Between experiments, the samples of linoleum were stored on shelves in a well-ventilated laboratory, unpacked. The carpet used in the present experiments was a tufted bouclé (100% polyamide fibres and latex backing) that had been installed on a floor and used for about 20 years in an office building, removed in the mid-1990s in connection with other experiment (Pejtersen et al., 2001) and later used as a source of indoor air pollution in many laboratory experiments (e.g. Wargocki et al., 1999, 2002). When not used in the experiments, the samples were stored on shelves in the laboratory, unpacked, as were the samples of linoleum. During experiments, the samples were stapled back to back so that only the front side was exposed to the air.

Immediately prior to each experiment, all the material samples were conditioned in the preparation room located in front of the test chambers by exposing them to typical indoor conditions for a week in order to bring the samples into equilibrium with the indoor temperature and relative humidity.

2.2 Experimental facilities

The experiments were carried out in the twin stainless steel climate chambers at DTU (Albrechtsen, 1988), Figure 1. Each chamber had a floor area of 9 m², a height of 2.5 meters and a volume of 22.5 m³. They were ventilated using underfloor air distribution that supplied 100 % outdoor air, with exhaust outlets in the ceiling. The chambers were primarily designed for air quality experiments and could provide temperatures from 10°C to 40°C and relative humidity from 10 % to 90 %. A high air exchange rate could be achieved by underfloor ventilation, up to 50 h⁻¹.

Seven ventilated 200L glass chambers described in detail by Andersen et al. (1996) (40 x 50 x 100 cm) were placed in the stainless steel chambers, 3 in one chamber and 4 in the other one. These chambers had been used in many other experiments to characterize the emissions from materials, using a so-called small-scale setup (Strøm-Tejse et al., 2003; Wargocki, 2004). The samples of the materials tested in the present experiments were placed inside these chambers. The chambers

were made of glass glued together on metal support racks. Two ventilators were attached on the top of each glass cover to ensure that the chambers were well ventilated. The air was extracted via a diffuser allowing for sensory and chemical measurements of the air exhausted from the chamber. The samples of materials were placed in the inner part of the chamber on a stainless steel rack (Fig. 1b). Mortar slabs were placed with the flange on the bottom of each glass chambers while linoleum or carpet was attached to the metal rack placed inside the chamber. The samples were placed with some distance between each so that the air could flow freely around them. A smoke test was performed to confirm that the air was well mixed in the fully loaded glass chambers.

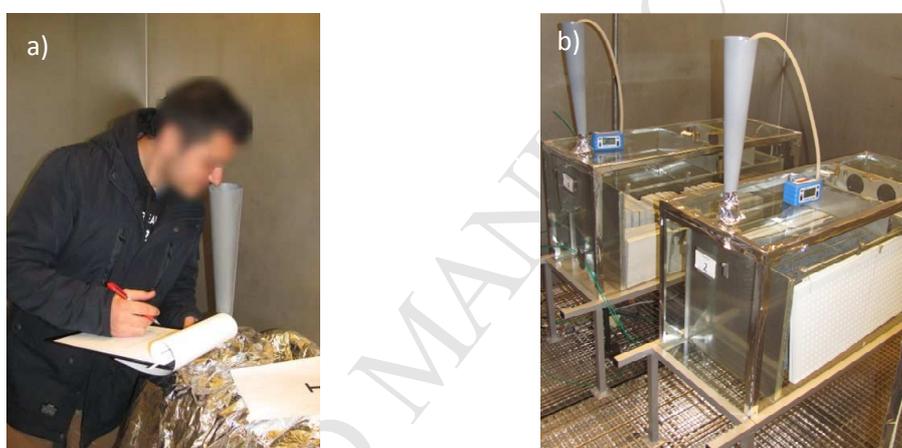


Figure 1 *A subject inhaling the air from a diffuser above the glass chamber, which was covered with aluminium foil to prevent subjects from identifying the pollution source (1a) and experimental set up and air sampling for the chemical measurements (1b)*

2.3 Experimental conditions

The steel chambers were ventilated with a maximum outdoor airflow resulting in the outdoor air change rate of 50 h^{-1} . The air temperature was set to 23°C . Relative humidity was not controlled and varied during experiments between 13 % and 32 %. The glass chambers were ventilated with the air supplied to the steel chambers. The inlet airflow was set to be about 0.9 L/s and the air velocity at the top of diffuser in the glass chamber where the sensory and chemical measurements were performed was about 0.2 m/s .

The loading of materials in the glass chambers were designed in accordance with the Nordtest method described in NT Build 482 Report (Nordtest, 1998). The method defines loading factors for specific surface areas with respect to the dimensions of a model room. Based on the loading factors for a model room (Table 1) and the airflow in the chamber, the total surface areas of the materials exposed were calculated to be 1.4 m², 1.9 m² and 2.8 m² for the mortar slabs. The area of 1.4 m² was considered as an initial loading of the mortar slab that represents a floor or ceiling area in the model room. The maximum exposed surface area of 2.8 m² was determined with respect to the capacity of the glass chambers – there was no room for more material. Mortar was tested with carpet under two mortar loadings, compared to three loadings in the case with linoleum, because of limited number of the available experimental days being the consequence of limited resources to perform this study. For the carpet and linoleum a loading of 1.4 m² was used, corresponding to the floor area in the model room, as indicated above. All 18 conditions tested in the present experiments are shown in Table 2.

Table 1 *Loading of materials in the present experiments; CM=cement mortar; AM=cement-ash-mortar; Li=linoleum; Ca=carpet*

Material loading in glass chamber [m ²]	Area specific air flow rate q [m ³ /h m ²]	Model room		Material
		Loading factor [m ² /m ³]	Surface area [m ²]	
1.4	2.4	0.420	7.1*	CM, AM, Li, Ca
1.9	1.8	0.571	9.6	CM, AM
2.8	1.2	0.841	14.2	CM, AM

*Surface area of 7 m² represents floor/ceiling area in the model room based on the Nordtest method NT Build 482

Table 2 *Experimental plan including material set up and loadings; CM=cement mortar; AM=cement-ash-mortar; Li=linoleum; Ca=carpet*

Experimental matrix								
Mortar loading [m ²]	CM	CM + Li	CM + Ca	AM	AM + Li	AM + Ca	Li	Ca
1.4	•	•	•	•	•	•	•	•
1.9	•	•		•	•			
2.8	•	•	•	•	•	•		

In one of the glass chambers, 2-propanone was evaporated passively so that its concentration was constant at about 80 ppm; the concentration was measured using an Innova 1312 gas monitor. According to Wargocki et al. (2009), this concentration is predicted to produce a strong odour (as shown in the Supplementary Material (SM2), subjects in this experiment also assessed that this concentration is producing strong odour). 2-propanone exposure was used as a reference condition to examine whether any changes in the performance of the sensory panel performing sensory assessments occurred during the course of experiment.

All the glass chambers were covered with aluminium foil during the experiments so that their content was invisible for the subjects performing the sensory assessments. The location of the materials in the chambers was randomly changed during the course of the experiments to avoid any systematic errors.

2.4 Sensory panel

The sensory panel consisted of twenty-five subjects (13 males and 12 females). They were recruited through advertisements at the university campus. They were 22 to 38 years old, all university students, non-smokers, and not suffering from asthma, allergies, or other chronic diseases. The subjects judged themselves to be fit and healthy. They participated voluntarily in the experiment. The subjects were asked not to eat spicy food and not to use perfumes during on any of the days of the experiment.

2.5 Sensory measurements

The subjects assessed odour intensity, the acceptability of the air quality, air freshness, pleasantness and nose irritation. Only the ratings of odour intensity are presented in the present paper and the reason for this is provided later. The ratings were made using scales printed on separate sheets of paper. The odour intensity scale (Yaglou et al., 1936) shown in Fig. 2 was used to obtain sensory measurements of odour intensity. The subjects were instructed on how to perform sensory evaluations and had an opportunity to practice the assessments before the actual experiments.



Figure 2 *Odour intensity scale. Once the ratings were performed the scale was coded as follows: 0 – No odour, 1 – Slight odour, 2 – Moderate odour, 3 – Strong odour, 4 – Very strong odour, 5 – Overwhelming odour*

2.6 Chemical measurements

The air used in the chemical analysis was withdrawn from the glass chambers through sorbent tubes inserted into a diffuser perpendicular to the direction of air flow; steel tubes containing Tenax TA sorbent were used (Fig. 1b). Double sampling was made; the volumes sampled were 3 L and 4 L. The airflow rates through the tubes were 0.1 L/min and 0.08 L/min respectively; they were controlled by calibrated pumps. A 3-L sampling was additionally made of the background air in the chamber. Blanks were made and analysed, too.

The samples were analysed by an external certified laboratory. The sampling tubes were desorbed in an automated thermal desorption/purge and trap injector. The airborne substances adsorbed on Tenax TA were analysed by TDS-GC-MS

according to ISO 16000-6. After transferring to a non-polar capillary column, the trapped compounds were separated in a gas chromatograph and detected in a mass spectrometer. Identification of compounds was achieved using standard mass spectra libraries (NIST, Wiley). The compounds identified were quantified against pure reference compounds covering volatile organic compounds (VOCs) from C5 to C22. Substances in the range of C6 to C16 were reported as VOCs, the more volatile ones as very volatile organic compounds (VVOCs), and those eluting after C16 as semi-volatile organic compounds (SVOCs). The concentrations were calculated as toluene equivalents with a lower reporting limit of $1 \mu\text{g}/\text{m}^3$.

In the conditions in which cement-ash-based mortar samples were present there could have been ammonia emissions due to the ash processing (“Selective catalytic reduction,” 2017; “Selective non-catalytic reduction,” 2016; Bødker, 2006a). Two conditions with such samples at the highest mortar loading of 2.8 m^2 were therefore examined for ammonia according to the guidelines relevant for organic recycled materials and ash-made concrete as defined by the National Institute for Occupational Safety and Health (NIOSH). The method is based on a solid sorbent tube, sulfuric acid-treated silica gel, and spectrophotometry. The accuracy of the method is not available.

2.7 Procedures

The sensory evaluations of the air extracted from the chambers with samples of mortar, flooring materials and their combinations present, an empty chamber and a chamber containing 2-Propanone were carried out on eight non-consecutive days in March and April 2015. The glass chambers in the stainless steel chambers were loaded with the tested materials on the day prior to each experimental session and sealed with an aluminium emission-free tape. Material samples were conditioned for about 20 hours prior to the assessments or to chemical sampling. During sensory assessments, the subjects assembled in the ventilated hall adjacent to the stainless steel chambers. They were then called one-by-one by name and asked to perform an evaluation of the air extracted through the diffuser from one of the seven chambers. After entering the chamber, they approached the diffuser and inhaled the air exhausted from the chamber through it. Only one inhalation was made, after which they rated odour intensity, the acceptability of the air quality, irritation, freshness and pleasantness of the air. Then they left the chamber and took a 1-3 minute break in the ventilated space before the next evaluation. The subjects performed 9 evaluations on one day and a total of 72 evaluations during the eight-day experiments. Besides assessments of the air extracted from the chambers they also assessed the air in each steel chamber. The order in which the evaluations were made was randomized for all subjects.

Chemical sampling was performed on the same samples and with an identical chamber setup one year after the sensory experiment. In the meantime, the material samples had been wrapped and stored. Identical procedures for conditioning the samples as in case of the sensory assessments were applied.

2.8 Statistical analyses

Statistical software R version 3.3.1 (R Core Team, 2016) was used to analyse the results of the sensory assessments. A Linear Mixed Effects (LME) model (Laird, 1982) was used to analyse the influence of the different types of mortar, the amount of mortar present (loading) and the presence/absence of linoleum/carpet on the odour intensity assessed by the subjects. Type and loading of mortar and presence of pollution source were treated as fixed effects (with interactions) in the model. Subjects were treated as a random factor to account for a repeated-measures design. Inspection of Quantile-Quantile plots (QQ-plots) was used to evaluate the Normality of the model residuals. As the model residuals turned out to deviate from a Normal distribution, the data were log-transformed before analysis. In addition, pair-wise comparisons were made using the Wilcoxon matched-pairs signed-ranks test to compare linoleum and carpet alone and in combination with mortar, and to investigate differences between mortars at a specific loading. The level for rejection of the Null Hypothesis was set at $P=0.05$ (2 tail).

3. Results

3.1 Sensory assessments

The odour intensity increased linearly as a function of the logarithm of the material loading, following the function established by Cain (1969) (Fig. 3).

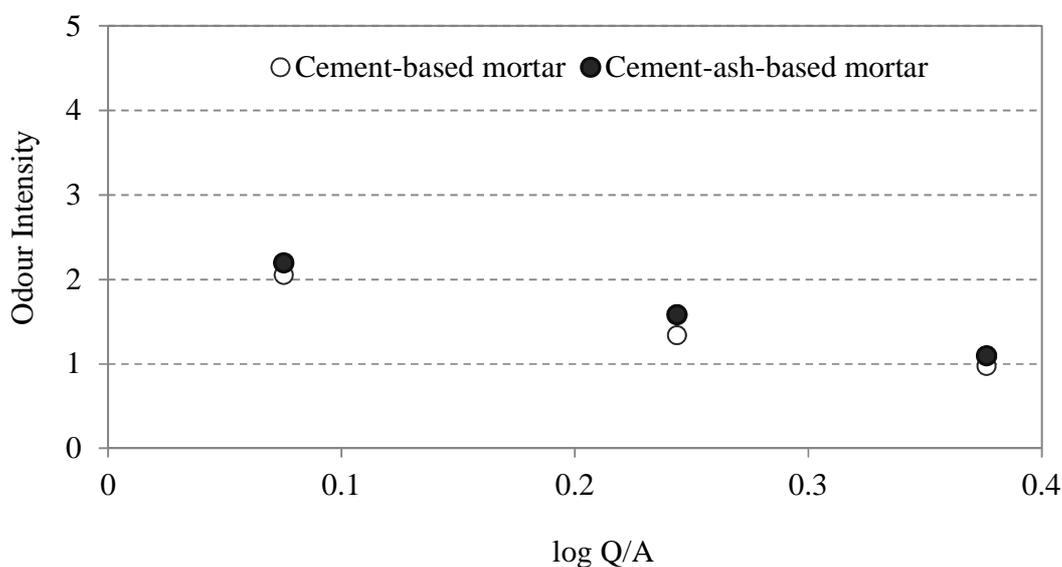


Figure 3 *Exposure – response relationship between the logarithm of the ventilation rate to loading of cement-based mortar (CM) and cement-ash-based mortar (AM) ($\log Q/A$) and the perceived odour intensity*

The results of the analysis of the Linear Mixed Effects (LME) model are shown in Figs. 4 and 5. They show that when mortar slabs were present alone in the chambers there was no significant difference in the mean odour intensity produced by either cement-based mortar or cement-ash-based mortar at the same loading. As the loading increased, the odour intensity increased and the increase of odour intensity with the mortar loading was statistically significant ($p < 0.001$). Addition of linoleum or carpet to mortars at the lowest loading level of 1.4 m^2 significantly increased the odour intensity ($p < 0.05$). The odour intensity of cement-ash-based mortar with linoleum was significantly higher ($p < 0.05$) at the loading levels of 1.9 m^2 and 2.8 m^2 compared to cement-based mortar with linoleum. The similar was observed for the combination of carpet with cement-based mortar and cement-ash-based mortar, significant differences were observed for both studied loadings, specifically significant difference ($p < 0.05$) for 1.4 m^2 loading of mortars and significant difference ($p < 0.001$) for 2.8 m^2 loading of mortars. Increasing the loading of mortars with linoleum significantly decreased the perceived odour intensity ($p < 0.05$). Increasing the loading of mortars with carpet did not change the odour intensity in the case of the combination of carpet and the cement-ash-based mortar but significantly decreased the odour intensity when carpet and cement-based mortar were both present. A detailed summary of the results of the LME model analyses and on the acceptability of the

perceived air quality are presented in the Supplementary Material (SM1), which also includes the results of the assessments made on the acceptability scale and show the same trends as for the ratings of odour intensity.

The Supplementary Material (SM2) also presents the sensory assessments of 2-Propanone (Acetone), which showed no changes in these ratings during the eight-day long experiments.

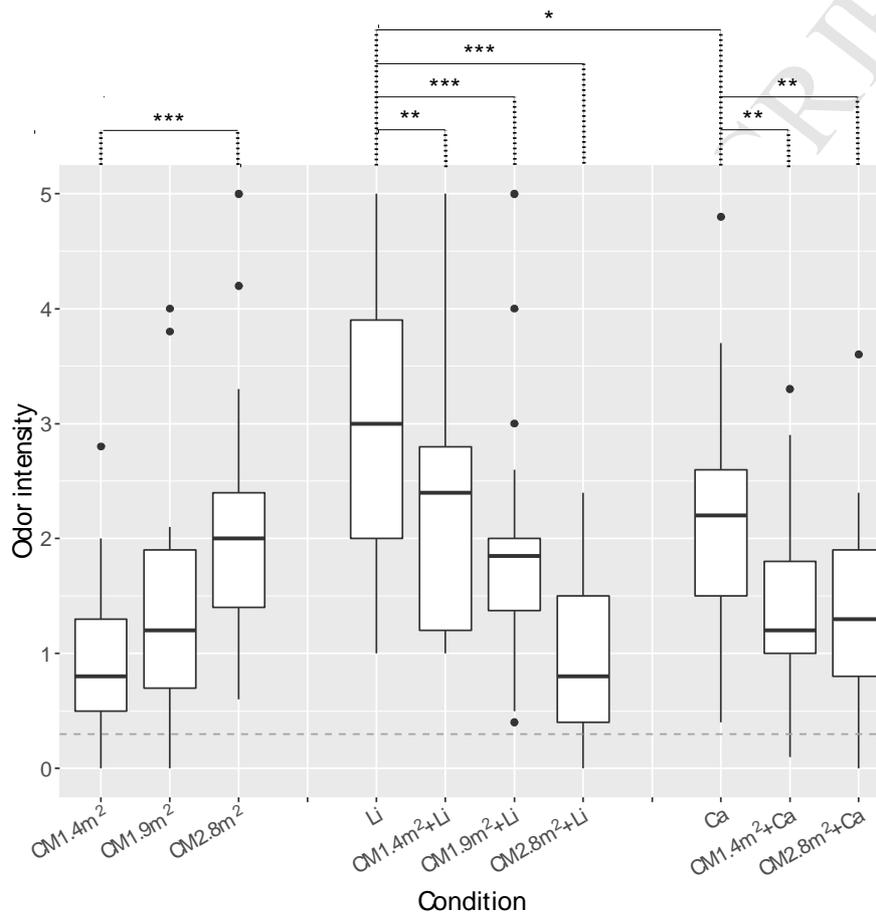


Figure 4 *Boxplots presenting assessment of odour intensity on the air extracted from the chambers containing cement-based mortar slabs (CM), linoleum (Li), carpet (Ca) and their mixtures. Black dots show outliers while dashed line presents the median of odour intensity of the air extracted from the empty glass chamber (OI = 0.3). Asterisks indicate the results of pairwise comparisons examined using Wilcoxon signed-rank test, where * indicates $0.01 < p < 0.05$, ** indicates $0.001 > p < 0.01$ and *** shows that $p < 0.001$. It should be noted that the box plots present raw data while the statistical analyses were made on the log-transformed data*

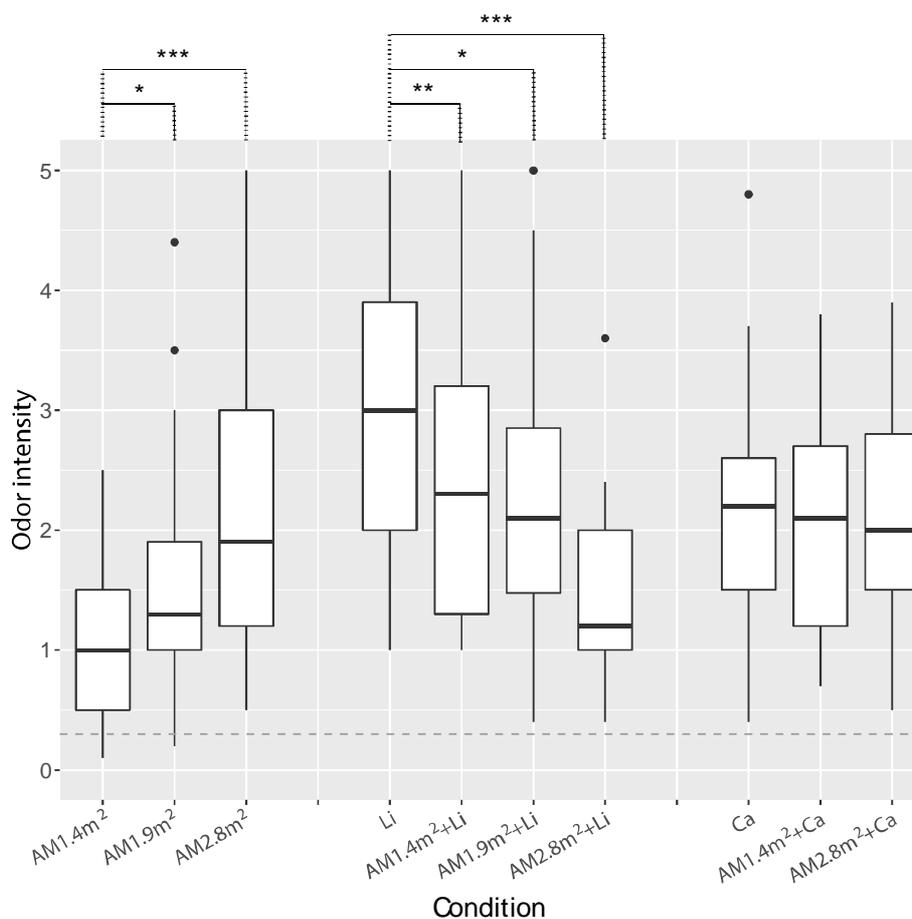


Figure 5 *Boxplots presenting assessment of odour intensity on the air extracted from the chambers containing cement—ash-based mortar slabs (AM), linoleum (Li), carpet (Ca) and their mixtures. Black dots show outliers while the dashed line presents the median of odour intensity of the air extracted from the empty glass chamber ($OI = 0.3$). Asterisks indicate the results of pairwise comparisons examined using Wilcoxon signed-rank test, where * indicates $0.01 < p < 0.05$, ** indicates $0.001 > p < 0.01$ and *** shows that $p < 0.001$. It should be noted that the box plots present raw data while the statistical analyses were made on the log-transformed data*

3.2 Chemical measurements

Table 3 shows the results of the GC-MS analyses, listing the compounds detected with a toluene equivalent concentration higher than $1 \mu\text{g}/\text{m}^3$. The results presented in Table 3 are from the 3-L samples; results based on 4-L sampling (presented in

the Supplementary Material (SM3)) revealed that the compounds detected were the same but they showed lower concentrations for most of the compounds compared with the 3-L samples, suggesting that break-through had occurred. Table 3 shows that any differences in types and concentration of pollutants emitted by cement-based and cement-ash-based mortars were negligible. In the case of the linoleum, the measurements showed that it was a strong source of organic acids, many of which occurred at fairly high concentrations, often close to their odour thresholds. The results presented in the table show that the concentrations of these acids decreased when linoleum was present in combination with cement-based and cement-ash-based mortars. In the case of the carpet, Table 3 shows that it was a moderate source of acids, alcohols and aldehydes. When it was present in combination with the mortars, the concentration of acids decreased as in the case of linoleum and the concentrations of other compounds remained unchanged. Table 3 shows no marked differences in the concentrations of VVOCs, TVOCs and SVOCs for the conditions analysed. However, differences were found when the concentrations of compounds constituting different functional groups were aggregated and compared, especially as regards the organic acids. The graph in the Supplementary Material (SM4) shows these differences.

Table 3 Concentrations of compounds emitted from the cement-based mortar (CM), cement-ash-based mortar (AM), linoleum (Li), carpet (Ca) and their mixtures; the results are from the 3 L samples and the concentrations are toluene equivalents in $\mu\text{g}/\text{m}^3$. Odour thresholds (OT) compiled by Devos (1990) or determined by Nagata (2003), (marked with ‘*’) are presented as well.

Materials	Concentration range $\mu\text{g}/\text{m}^3$ (3-litre sampling)									
	CM 2.8 m ²	AM 2.8 m ²	Linoleum (Li)	CM 1.4 m ² + Li	CM 2.8 m ² + Li	AM 2.8 m ² + Li	Carpet (Ca)	CM 2.8 m ² + Ca	AM 2.8 m ² + Ca	OT
ALCOHOLS										
1-Dodecanol							2			100
1-Ethoxy-2-propanol								3		
2-Ethyl-1-hexanol							1	3		

Benzyl alcohol							2			115*
n-Butanol	4	3	8	8	4	4	8	7	4	
Phenol				1			1			427/21*
Triethylene glycol	270	14					3			
Trimethylsilanol	1						<1			
Sum Alcohols	275	17	8	9	4	4	17	13	4	
ALDEHYDES										
Benzaldehyde		2	2	2	2	2	5	2	2	186
trans-2-Decenal			3							2
Dodecanal							2			
Pentanal								3		22
n-Decanal		1	3	3	2	7	3	3	3	6
n-Hexanal	2	1					2	5		58
n-Nonanal		2	7	5	3	5	7	4	4	13
Octanal			2		1		3	2		7
Sum Aldehydes	2	6	17	10	8	14	22	19	9	
ORGANIC ACIDS										
Acetic acid	15	8	63	16	21	13	119	20	31	363/16*
Butanoic acid			11		2					14
Decanoic acid (Nonanoic acid)			2	1	2	1	3		1	63
Formic acid			76						15	54954
Heptanoic acid			11		2					148
Hexanoic acid			41	4	5	3	5	2	1	60/3
Isobutyric acid			2							
Lactic acid		1					1	4	2	
Nonanoic acid			5		1		2			13
Octanoic acid			15		3	1	2			24
Pentanoic acid			13							20
Propanoic acid			51		5					110
Sum organic acids	15	9	290	21	41	19	131	26	50	
ALKANES										
2-Methylpentane (3-Methylpentane) (Isohexane)										154
Methylcyclopentane										36
Cyclohexane									5	77625

C6 (Hexane)							36		79433/5690*
C15 (Pentadecane)		4		4	3	3		2	2
C16 (Hexadecane)		3		3	2	3	3	2	3
Hexamethylcyclotrisiloxane		2	2	2	1	2	1	2	1
Sum alkanes	0	9	2	9	6	8	4	237	6
Sum other iso/cyclo-alkanes:	129	152	4	138	94	110	38	84	95
OTHER COMPOUNDS									
Acetone	3	3				3	4	23	34674/11300*
N-Methyl-2-pyrrolidone			2						
gamma-Nonalactone			2		1				
n-Butyl acetate							2		933/82*
Benzothiazole									
Dicarboxylic acid ester			2				3		
Dibutyl eter			2	2		1	1		1
Toluene	2	1	<1	2	1		1	2	5888/1340*
Sum other terpenes:			1				2		
Sum VVOC (< C6):	4	4	76	<1	4	<1	5	179	15
TVOC (C6-C16):	461	194	251	191	154	156	222	225	150
Sum SVOC (> C16):	< 1	< 1	<1	<1	<1	<1	<1	< 1	< 1
TVOC_{Toluene equivalents (ISO 16000-6):}	243	90	42	120	36	62	199	81	46

Chemical analyses of the cement-ash-based mortar samples examining ammonia emissions were performed at the highest loading of the mortar (2.8 m²). They were performed when mortar was present alone and in combination with linoleum. In both cases the concentration was below the limit of detection for ammonia, which was 0.030 mg/m³ in 40 litres of sampled air.

4. Discussion

Sensory assessments of odour intensity did not suggest differences in emissions between cement-based mortar and cement-ash-based mortar (Figs. 3-5). However, chemical measurements (Table 3) showed that the TVOC concentration was higher for the cement-based mortar than for the cement-ash-based mortar at the highest loading (2.8 m²). The reason for the

difference was the high concentration of triethylene glycol. Neither of the mortars is expected to emit this compound. This was confirmed by the results of measurements performed at other loadings and the mixtures of mortars with linoleum and carpet where triethylene glycol could not be detected. It is possible that the presence of triethylene glycol was due to the cleaning of the glass chambers that took place before new loading was placed in the glass chamber, triethylene glycol being a residue of the detergent used for cleaning the glass surfaces. It would then be fair to conclude that adding sewage sludge fly ash to mortar does not have a measurable effect on the VOC emissions from a cement-based mortar slab.

The odour intensity increased when linoleum was placed together with the cement-based and the cement-ash-based mortar. But then it decreased when the loading of the mortars was increased and was even lower than when the mortars alone were present. One possible explanation for the observed phenomenon could be that the mortars adsorb the pollutants emitted from linoleum. Table 3 shows that the mortars are a particularly strong sink for organic acids, which are emitted in fairly large quantities from linoleum. The sink effect can be explained by the fact that the mortars represent a strong base, which attracts acids. They have a particularly high pH when freshly cast (Pham and Prince, 2014) though with time, as the mortar matures, its alkalinity decreases and pH neutralizes due to a carbonation process that forms calcium carbonate. This suggests that the potential for adsorbing organic acids could decrease with time. However, this was not observed in the present experiments. The sensory measurements were performed a few weeks after the mortars were cast, while the chemical measurements were carried out one year later, and both indicated a strong sink effect. In future experiments, it would be interesting to examine how long mortars maintain their ability to adsorb acids.

The aging of mortars produces other changes in their structure due to chemical reactions and drying. Pham (2014a, 2014b) showed that these processes result in micropores being clogged while mesopore structure likely to expand due to the cracks that form during carbonation. These processes may increase the potential area for adsorption even though the pH has decreased. It may thus be postulated that the sink effect observed directly after casting the mortars was due to their basicity and the sink effect observed one year later, when the chemical measurements were made, was due to both their porosity and basicity, but this hypothesis cannot be verified by using only the present result. In the future experiments, it would be useful to study further the mechanisms causing the sink effects and how long they last. The observed sink effects may also change depending on the way the indoor surfaces are finished. Information on these sink effects and their mechanisms may have considerable practical implications.

When the magnitude by which odour intensity was reduced in the combined exposure of mortars with linoleum is examined, it can be concluded that it is rather unlikely that the adsorption of pollutants was the only mechanism that was responsible. For example, it could also be that the odour produced by the mortars was masked by that of linoleum or that the adsorption of compounds on the mortar surface could have reduced emissions by blocking or clogging the mortar pores. A possible explanation for the reduced odour intensity in the combined exposure of mortars with linoleum could also be the compensation or subtraction of odour intensities, which is a specific case of hypo-addition of two odour intensities (Cain and Drexler, 1974). Future experiments should closely examine and describe these processes to further elucidate the results observed in the present study.

Based on chemical measurements, linoleum is a strong source of organic acids while carpet emits fewer acids (Table 3). This explains why the sink effect was much smaller in the case of the combined exposure of mortars with carpet. It should be noticed that the sink effect was seen both for the cement-based and the cement-ash-based mortar, which suggests that adding fly ash did not change the sorption properties. The above comparisons provide an independent verification and reproducibility of the observed sink effect for the cement-based mortar.

The sensory assessments showed that linoleum produced the strongest odour among all conditions tested (Figs. 4 and 5). It was higher than the intensity of odour produced by the carpet, cement-based and cement-ash-based mortars. However, the TVOC concentration did not validate the presence of such considerable differences and showed that the emissions from the cement-based mortar were actually the highest. Taking these results into account it can be concluded that pure chemical measurements may not always provide information on the potential effects of material emissions on air quality as it is perceived by humans. In addition, the results presented in Table 3 imply that there are considerable limitations on aggregating the concentrations of VOCs measured or integrating the signal from gas chromatography and expressing the concentration of all pollutants as the Toluene equivalent concentration. The result of these procedures, the TVOC concentration, may not be able to predict the effects on humans, as was pointed out by Andersen et al. (1996).

It is worth noting that the present results show the benefits of chemical measurements and support the need for them. They show that detailed analysis of pollutants is worthwhile because a clear difference between the concentrations of individual pollutants and the sum of concentrations of functional groups can be seen (Table 3). For example, in the case of linoleum, high levels of organic acids were measured and in the case of the carpet, aldehydes and alcohols were measured at levels that were similar to those reported in other studies (Gunnarsen et al., 1994; Jensen et al., 1995c, 1995b; Johnsen et al., 1991;

Wargocki, 1998; Wargocki et al., 1999; Wolkoff et al., 1993). These compounds have low odour thresholds as compared to the emissions from cement-based and cement-ash-based mortars (“CDC - NIOSH,” n.d.). This can explain why the emissions from linoleum and carpet produced the highest odour intensity (Figs. 4 and 5). Mortar samples, which are composed of mainly inorganic components, which were not measured in the present experiments, were mostly sources of iso/cyclo-alkanes, which can be considered to be without perceptible odour. The origin of the iso/cyclo-alkanes was not identified, but based on the Green Concrete study (Bødker, 2006a), the mineral oil sprayed into the mould used to cast each slab may be a source of hydrocarbons, a functional group of compounds containing alkanes, cycloalkanes and others. Table 3 shows differences between the aggregated concentrations of pollutants from different functional groups, suggesting that if integrated indexes are based on chemical measurements they should differentiate between functional groups of pollutants rather than integrate all pollutants “across the board” as is conventional when calculating TVOC concentration (see e.g. (Salis et al., 2017)). This approach was proposed by Seifert et al. (1999) when they defined the limits for TVOC concentration to avoid discomfort based on the measured VOC levels in existing buildings and proposed limits on indicators created by aggregating the concentrations of VOCs for each specific group of compounds.

As described in the Methods section, the 3 L and 4 L samples were taken during the chemical measurements. The analyses of both samples show that the concentration of pollutants in 4 L samples was in general lower, suggesting breakthrough. This is why only the results for the 3 L samples were presented in Table 3. Despite the breakthrough, the trends of the results of the chemical measurements in the different conditions were the same when the analyses of 3 L and 4 L samples were compared (see Supplementary Material (SM3)), both showing that organic acids were removed by the mortar slabs.

The relative humidity was not controlled in the present experiments. This could bias the assessments of acceptability of air quality (Fang et al., 1998). Odour intensity was therefore used as a major sensory outcome, as the results of Fang et al. (1998) show that perception of odour intensity is not affected by varying temperature and relative humidity of the inhaled air; similar approach was used by Tsushima et al. (2017). Although the ratings of acceptability were not used as the major sensory outcome, they generally corresponded to the ratings of odour intensity (see Supplementary Material (SM1)).

The present results show that building materials should not only be considered as pollution sources (Wargocki, 2004), but also in some cases as potential air cleaners. A similar conclusion was put forward by Sakr et al. (2004), who showed that unpainted gypsum walls can considerably improve perceived air quality, while Darling et al. (2011) showed that clay plaster can act as a buffer for ozone.

Even though the present results do not show any large difference between cement-based and cement-ash-based mortars the potential risks for the environment and for health of the ash content, e.g. leakage of heavy metals, should also be investigated and considered before mortars with fly ash can be considered harmless.

5. Conclusions

- There were no differences in sensory emissions or chemical emissions between cement-based mortar and cement-ash-based mortar at the same loading.
- Odour intensity increased as the loading of cement-based mortar or cement-ash-based mortar increased.
- Both cement-based mortar and cement-ash-based mortar were shown to be significant sinks for organic acids.
- Cement-ash-based mortar is not a source of ammonia emissions at a detectable level.
- TVOC is not a good indicator of the sensory effects of emissions from materials, but the sum of VOCs of functional groups can be considered as a crude indicator of potential sensory effects.
- Future experiments should investigate the long-term adsorption capacity of the mortars investigated in the present experiments for acids and other pollutants and should validate the present laboratory findings in real scenarios.

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Highlights

- Characterization of emissions from cement-based mortar and cement-ash-based mortar
- Odour intensity increased as the slab area exposed was increased
- Emissions and odour intensity did not differ for mortars
- Cement-based and cement-ash-based mortars being strong sinks for organic acids
- Strong basicity of the cement mortar slabs as the main reason for the sink effects