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Non-microbial sources of microbial volatile organic compounds

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ABSTRACT

Background: The question regarding the true sources of the purported microbial volatile organic compounds (MVOCs) remains unanswered.
Objective: To identify microbial, as well as non-microbial sources of 28 compounds, which are commonly accepted as microbial VOCs (i.e. primary outcome of interest is Σ 28 VOCs).
Methods: In a cross-sectional investigation of 390 homes, six building inspectors assessed water/mold damage, took air and dust samples, and measured environmental conditions (*i.e.*, absolute humidity (AH, g/m^3), temperature (°C), ventilation rate (ACH)). The air sample was analyzed for volatile organic compounds (μg/m^3) and; dust samples were analyzed for total viable fungal concentration (CFU/g) and six phthalates (mg/g dust). Four benchmark variables of the underlying sources were defined as highest quartile categories of: 1) the total concentration of 17 propylene glycol and propylene glycol ethers (Σ17 PGEs) in the air sample; 2) 2,2,4-trimethyl-1,3-pentanediol monoisobutyrate (TMPD-MIB) in the air sample; 3) semi-quantitative mold index; and 4) total fungal load (CFU/g).
Results: Within severely damp homes, co-occurrence of the highest quartile concentration of either Σ17 PGEs or TMPD-MIB were respectively associated with a significantly higher median concentration of Σ 28 VOCs (8.05 and 13.38 μg/m^3, respectively) compared to the reference homes (4.30 and 4.86 μg/m^3, respectively, both *Ps* $\leq$ 0.002). Furthermore, the homes within the highest quartile range for Σ fungal load as well as AH were associated with a significantly increased median Σ 28 VOCs compared to the reference group (8.74 *vs.* 4.32 μg/m^3, *P*=0.001). Within the final model of multiple indoor sources on Σ 28 VOCs, one natural log-unit increase in summed concentration of Σ17 PGEs, plus TMPD-MIB (Σ 17 PGEs + TMPD-MIB) was associated with 1.8-times (95% CI, 1.3–2.5), greater likelihood of having a highest quartile of Σ 28 VOCs, after adjusting for absolute humidity, history of repainting at least one room, ventilation rate, and mold index (*P-value* =0.001). Homes deemed severely mold damaged (*i.e.*, mold index =1) were associated with 1.7-times (95% CI, 0.8—3.6), greater likelihood of having a highest quartile of Σ 28 VOCs, even though such likelihood was not significant (*P-value* =0.164). In addition, absolute humidity appeared to positively interact with mold index to significantly elevate the prevalence of the highest quartile category of Σ 28 VOCs.
Conclusion: The indoor concentration of Σ 28 VOCs, which are widely accepted as MVOCs, are significantly associated with the markers of synthetic (*i.e.* Σ17 PGEs and TMPD-MIB), and to less extent, microbial (*i.e.*, mold index) sources.

1. Introduction

The purported microbial volatile compounds (MVOCs) refer to compounds, which are generated during metabolism of nutritional substrate by microbial species, including fungi and bacteria (Korpi et al., 2009; Van Lancker et al., 2008). Within damp indoor environment, mold and bacterial growth could generate a wide array of biological contaminants (Thrasher and Crawley, 2009). Human exposure to some of the purported MVOC$_S$, such as 1-octen-3-ol

Abbreviations: Σ 28 VOCs, summed concentration of 28 volatile organic compounds of interest; aOR, adjusted odds ratio; BBzP, benzylbutyl phthalate; CFU, colony forming units; DBH, dampness in building and health; DEHP, di-(2-ethylhexyl)phthalate; ETS, environmental tobacco smoke; MVOC, microbial volatile organic compound; NILU, norwegian institute for air research; OR, odds ratio; PFT, perfluorocarbon tracer; Σ17 PGE, total concentration of 17 propylene glycol and propylene glycol ethers; PVC, polyvinyl chloride; RERI, relative excess risk due to interaction; TMPD-MIB, 2,2,4-trimethyl-1,3-pentanediol monoisobutyrate; VOC, volatile organic compound; 95% CI, 95% confidence interval

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and 2-pentanol, within indoor venue represents a public health concern. This is not only because they are possible sentinel markers of hidden microbial infestation and associated biocontaminants (Wilkins and Larsen 1995), but also because 1-octen-3-ol and 2-pentanol are significantly associated with irritation of mucosal layers (Araki et al., 2010), irritation of ocular and nasal mucosa (Elke et al., 1999; Wålinder et al., 2008), elevated likelihood of asthma and respiratory symptoms, including nocturnal breathlessness in children (Kim et al., 2007), allergic rhinitis (Araki et al., 2012), and sick building syndrome in adults (Araki et al., 2010; Takeda et al., 2009), respectively. Laboratory-based exposure to 10 mg/m^3 of 1-octen-3-ol could induce increased levels of nasal lavage biomarkers (*i.e.* eosinophil cationic protein, myeloperoxidase, and lysozyme), and respiratory irritation symptoms (Wålinder et al., 2008), reduced pulmonary function in murine models (Korpi et al., 1999), or promote enhanced nasal lavage-myeloperoxidase and nasal patency reduction in a group of house painters (Wieslander and Norback 2010).

In spite of such growing body of evidence, one of the most critical gaps in knowledge regards their true sources (Fischer and Dott 2003; Korpi et al., 2006). That is, a growing line of controlled chamber experiments and epidemiologic investigations has failed to observe a consistent association between fungal and/or bacterial contamination and the purported MVOC$_S$ (Kim et al., 2007; Korpi et al., 2006; Malysheva et al., 2014; Matysik et al., 2008; Sahlberg et al., 2013; Schuchardt and Strube, 2013). In contrast, multiple synthetic sources (*e.g.*, building materials, cleaning agents, automobile emissions) (Nalli et al., 2006; Singer et al., 2006), anthropogenic (*e.g.*, cigarette smoke, cooking processes, foods) (Korpi et al., 2009; Newsome et al., 1965; Schleibinger et al., 2008), or atmospheric sources (Korpi et al., 2009) have been shown for the purported MVOCs. For example, 1-octen-3-ol, which is widely accepted as an MVOC, was observed in high concentration in the personal air samples for the house painters (Wieslander and Norback 2010). 1-Octen-3-ol was also significantly correlated with glycol ethers and other plasticizers, which could be emitted from water-based paints (Wieslander and Norback 2010). A number of other purported MVOCs have been observed in moderate to high correlation with building materials (Bornehag et al., 2005b; Matysik et al., 2008; Polizzi et al., 2012) or under humid conditions (Polizzi et al., 2011, 2009). Within our earlier investigations, we observed significant association between summed propylene glycol and glycol ether concentration (Σ17 PGEs) with commercial cleaning fluids, paint, and history of repainting at least one room (Choi et al., 2010a, 2010b). Other investigations have noted moderate to high correlation between 2,2,4-trimethyl-1,3-pentanediol monoisobutyrate (TMPD-MIB) with latex paint and floor polish (Kim et al., 2007; Wieslander et al., 2010; Wieslander and Norback 2010). In addition, di-(2-ethylhexyl)phthalate (DEHP) and benzyl butyl phthalate (BBzP), are common plasticizers added to the polyvinyl chloride material (Bornehag et al., 2005b).

Within the present investigation, we posit that identification of the most plausible sources of the purported MVOCs is critical for better understanding their health risks (Araki et al., 2010). This is particularly important in light of earlier observation of synergistic increase in health risks from exposure to the MVOC mixture (Cometto-Muñiz et al., 1997; Korpi et al., 1999), even if the individual risk is not significant (Schleibinger et al., 2008). To date, direct search for non-microbial sources of MVOCs has not been systematically conducted. Therefore, the primary goal of the present investigation is to identify specific microbial and/or synthetic sources of the MVOCs using both quantitative and qualitative markers of the sources. We define Σ17 PGEs, DEHP, and BBzP as two distinct groups of markers of multiple synthetic sources. In contrast, TMPD-MIB is considered a marker for latex paint and/or coating-related materials. The secondary goal is to clarify how the indoor environmental conditions, including temperature, ventilation rate, and absolute indoor humidity modify the relationship between purported MVOC sources and their concentration.

2. Methods

Detailed descriptions of the study methods are provided elsewhere (Bornehag et al., 2005c; Holme et al., 2010). Briefly, this study is part of the on-going Dampness in Buildings and Health (DBH) study focusing on the impact of indoor environmental factors on asthma and allergy among children in Sweden. The present analysis has been conducted as part of exposure assessment.

2.1. Questionnaire

A cross-sectional questionnaire was answered by all parentsof all children included in the present analysis (Bornehag et al., 2005a, 2005b, 2003, 2005d).

2.2. Home exposure assessment by professional inspectors

Six inspectors conducted a visual and olfactory exam of mold and water damage, on building construction, materials, ventilation type, and mold and moisture problems. They also collected air and dust samples in the homes during the heating season (October 2001-April 2002). The inspectors graded each home regarding: 1) first impression of indoor air quality; 2) unpleasant smell along the skirting board; 3) visual signs of water damage (*i.e.*, stains) on walls or ceiling; and 4) damaged structural material (*e.g.*, blackened, bubbly, or loosening flooring material). Each outcome was graded zero (no issue); 1–2 (mild issue); or 3 (severe mold issue). They measured average ventilation rates using validated perfluorocarbon tracer (PFT) technique (Nordtest 1997; Stymne et al., 1994). Temperature (°C) and relative humidity (%) were measured instantaneously during the home visit (VL2000 Temperature & Humidity Sensor, Vaisala, Helsinki, Finland) and continuously at every hour for a week (Mitec Satelite-TH, Mitec Instrument AB, Säffle, Sweden).

2.2.1. Air sampling and mold index

The same team also took air samples from the child's bedroom, kitchen, as well as the living room for analyses of viable microorganisms (*i.e.*, fungi and bacteria) under usual living conditions (Holme et al., 2010). Reference samples were also taken from outdoors of every home. Back within the Mycoteam AS (Oslo, Norway), the concentration of microbial colony forming unit per cubic meter of air (CFU/m^3) was converted to mold index by a team of four mycologists, considering both diversity and concentration of the indoor sample relative to the outdoor reference sample (Norwegian standard for building survey, Tilstandsanalyse for byggverk, NS 3424) (Holme et al., 2010). Briefly, the mold index for given building was coded as 0 (no or weak indication of microbial infestation) or 1 (clear or severe signs of microbial infestation) (Holme et al., 2010).

2.2.2. Air sampling for volatile organic compounds

As described in Choi et al. (2010a, 2010b), SKC pocket pumps (model 210-1002, SKC Blandford, Dorset, UK) collected a sample of air in child's bedroom at 80 ml/min for 60–90 min 5–8 l) through Perkin Elmer adsorption tubes (glass, 300 mg Tenax TA). The samplers were placed 1 m above the floor in the room. The air samples were sealed with PTFE stoppers, shipped, and analyzed in Norway Institute for Air Research Norway within two weeks of sample collection. Use of adsorbent, preparation of adsorbent