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Predicting the gas-phase concentration of semi-volatile organic compounds from airborne particles: Application to a French nationwide survey

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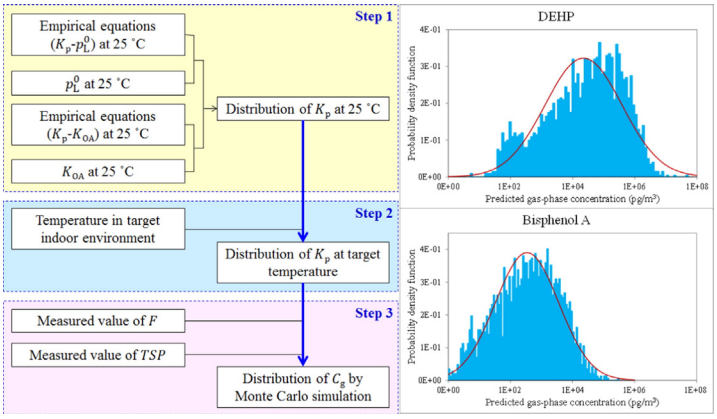
HIGHLIGHTS

- A new approach was developed to predict indoor gas-phase SVOC concentrations (C_g).
- The approach was based on Monte Carlo simulation at the indoor temperature.
- The distributions of the C_g of 66 SVOCs in the French housing stock were predicted.
- The C_g of some rarely studied SVOCs, e.g., DMEP and triclosan, were reported.

GRAPHICAL ABSTRACT



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ABSTRACT

Semi-volatile organic compounds (SVOCs) partition indoors between the gas phase, airborne particles, settled dust, and other surfaces. Unknown concentrations of SVOCs in the gas phase (C_g) can be predicted from their measured concentrations in airborne particles. In previous studies, the prediction of C_g depended largely on choosing a specific equation for the calculation of the particle/gas partition coefficient. Moreover, the prediction of C_g is frequently performed at a reference temperature rather than the real indoor temperature. In this paper, a probabilistic approach based on Monte Carlo simulation was developed to predict the distribution of SVOCs' C_g from their concentrations in airborne particles at the target indoor temperature. Moreover, the distribution of the particle/gas partition coefficient of each SVOC at the target temperature was used. The approach was validated using two measured datasets in the literature: the predicted C_g from concentrations measured in airborne particles and the measured C_g were generally of the same order of magnitude. The distributions of the C_g of 66 SVOCs

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in the French housing stock were then predicted. The SVOCs with the highest median C_g, ranging from 1 ng/m^3 to >100 ng/m^3, included 8 phthalates (DEP, DiBP, DBP, DEHP, BBP, DMP, DiNP, and DMEP), 4 polycyclic aromatic hydrocarbons (fluorene, phenanthrene, fluoranthene, and anthracene), 2 alkylphenols (4-tert-butylphenol and 4-tert-octylphenol), 2 synthetic musks (galaxolide and tonalide), tributyl phosphate, and heptachlor. The nationwide, representative, predicted C_g values of SVOCs are frequently of the same order of magnitude in Europe and North America, whereas these C_g values in Chinese and Indian dwellings and the C_g of polybrominated diphenyl ethers in U.S. dwellings are generally higher.

1. Introduction

Semi-volatile organic compounds (SVOCs), such as phthalates, polybrominated diphenyl ethers (PBDEs), and polycyclic aromatic hydrocarbons (PAHs), are emitted from various sources in living environments (Weschler and Nazaroff, 2008). SVOCs exist in the indoor environment in the gas phase, airborne particles and settled dust (Weschler and Nazaroff, 2008). Therefore, SVOCs can enter the human body through different pathways, namely the ingestion of settled dust, inhalation and dermal exposure. Many studies have suggested that exposure to SVOCs has negative effects on human health (Costa et al., 2014; Kim et al., 2014; Wang et al., 2015).

To assess the domestic exposure to SVOCs on a nationwide scale, the airborne particles (PM_{10}) in 285 dwellings (2003–2005) and the settled dust in 145 dwellings (2008–2009) throughout France were analyzed (Glorennec et al., 2011; Mandin et al., 2016). However, the SVOC concentrations in the gas phase were not measured but are essential from the perspective of assessing inhalation and dermal exposure.

At equilibrium partitioning, unknown SVOC concentrations in the gas phase (C_g) can be predicted from the measured SVOC concentrations in airborne particles (F) using the particle/gas partition coefficient (K_p) (Weschler and Nazaroff, 2008). The K_p values for SVOCs can be measured using an experimental method (Benning et al., 2013) or calculated using empirical equations that address the relationship between K_p and the saturation vapor pressure of the pure SVOC liquid (p_L^0) (He and Balasubramanian, 2009) or that between K_p and the SVOC octanol/air partition coefficient (K_{OA}) (Harner and Bidleman, 1998).

Weschler et al. (2008) proposed a model to calculate the C_g and F of phthalates from the total concentrations of each compound in air. The K_p value for each compound was calculated from the p_L^0 and K_{OA} values using the empirical equations proposed by Naumova et al. (2003) and Finizio et al. (1997), respectively. The K_p values calculated from the p_L^0 or K_{OA} values vary, resulting in a difference in the prediction of F. A suggested conclusion was that the accuracy of the prediction of the F or C_g of phthalates is limited by the accuracy of the calculation of K_p from either p_L^0 or K_{OA}. Salthammer and Schripp (2015) used a Monte Carlo method to calculate the distribution of K_p from the p_L^0 or K_{OA} values for some SVOCs. Comparison of the K_p values calculated using p_L^0 or K_{OA} showed satisfactory correlation and a trend toward higher estimation of K_p using p_L^0. In both studies, the prediction was performed at a reference temperature. However, the indoor temperature varies among dwellings and must be taken into account for the calculation.

To summarize, two problems remain unsolved in the previous studies regarding the prediction of the SVOC C_g. (1) A number of the empirical equations proposed in the literature result in different calculations. Therefore, the calculation of K_p and the prediction of the SVOC C_g depend largely on choosing a specific equation. (2) The calculations were frequently performed at a reference temperature, e.g., 25 °C, but the indoor temperature varies among dwellings. Wei et al. (2016a) found that an increasing indoor temperature from 15 °C to 30 °C can affect the K_p and predicted C_g by 750%.

Predictions of C_g are useful when they cannot be measured directly. The objectives of the present study were (1) to develop a probabilistic approach to predict the equilibrium concentrations of SVOCs in the gas phase from their concentrations measured in airborne particles for a large dataset while accounting for the uncertainties in the K_p and indoor temperature, (2) to apply this approach to predict the gas-phase concentrations of 66 SVOCs in a representative sample of French dwellings, and (3) to compare the gas-phase SVOC concentrations in French dwellings with those in other studies worldwide.

2. Material and methods

2.1. Framework for the prediction of the gas-phase concentration C_g

The framework of the probabilistic approach for the prediction of SVOC concentrations in the gas phase from their concentrations in airborne particles is presented in Fig. 1. The C_g in a given dwelling was predicted in three steps. Firstly, to prevent the bias of choosing a specific equation for the calculation of K_p, 38 available equations proposed in the literature to calculate K_p from two parameters (p_L^0 and K_{OA}) were retrieved. The empirical equations were all originally developed at a reference temperature (25 °C). Thus, these equations should be applied in an environment of 25 °C. Moreover, the available p_L^0 and K_{OA} values (an average of 10 values for each SVOC) were also retrieved. The distribution of K_p for each SVOC at 25 °C was calculated using the retrieved equations and parameters and was reported by Wei et al. (2016b). The median values of $\log_{10}K_p$ ranged between −5.69 (dichlorvos) and 1.90 (BDE 209).

Secondly, the distribution of K_p at 25 °C was adjusted to the distribution of K_p at the indoor temperature measured in each dwelling using

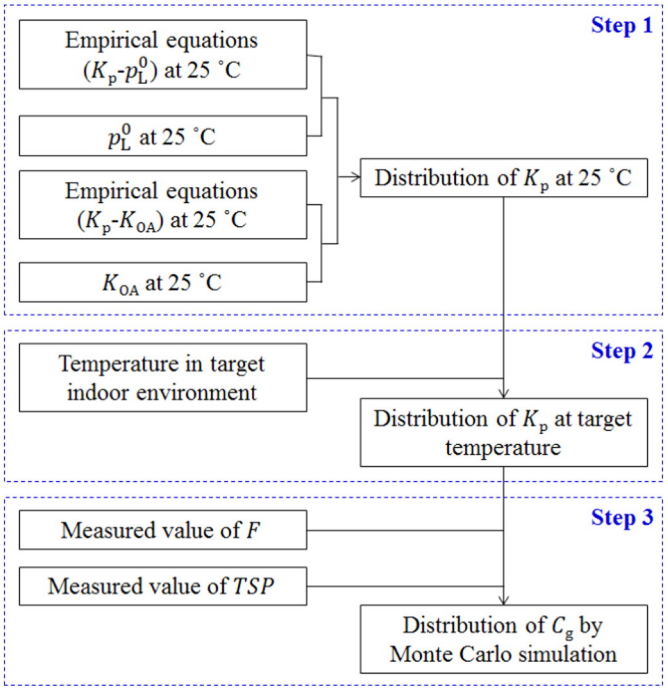

Fig. 1. Framework for predicting the gas-phase SVOC concentration.