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Using long-term air monitoring of semi-volatile organic compounds to evaluate the uncertainty in polyurethane-disk passive sampler-derived air concentrations[☆]

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ABSTRACT

Much effort has been made to standardise sampling procedures, laboratory analysis, data analysis, etc. for semi volatile organic contaminants (SVOCs). Yet there are some unresolved issues in regards to comparing measurements from one of the most commonly used passive samplers (PAS), the polyurethane foam (PUF) disk PAS (PUF-PAS), between monitoring networks or different studies. One such issue is that there is no universal means to derive a sampling rate (R_s) or to calculate air concentrations (C_{air}) from PUF-PAS measurements for SVOCs. C_{air} was calculated from PUF-PAS measurements from a long-term monitoring program at a site in central Europe applying current understanding of passive sampling theory coupled with a consideration for the sampling of particle associated compounds. C_{air} were assessed against concurrent active air sampler (AAS) measurements. Use of “site-based/sampler-specific” variables: R_s , calculated using a site calibration, provided similar results for most gas-phase SVOCs to air concentrations derived using “default” values (commonly accepted R_s). Individual monthly PUF-PAS-derived air concentrations for the majority of the target compounds were significantly different (Wilcoxon signed-rank (WSR) test; $p < 0.05$) to AAS regardless of the input values (site/sampler based or default) used to calculate them. However, annual average PUF-PAS-derived air concentrations were within the same order of magnitude as AAS measurements except for the particle-phase polycyclic aromatic hydrocarbons (PAHs). Underestimation of PUF-derived air concentrations for particle-phase PAHs was attributed to a potential overestimation of the particle infiltration into the PUF-PAS chamber and underestimation of the particle bound fraction of PAHs.

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1. Introduction

Global and regional air monitoring networks have been established to monitor the progress of regulatory measures to reduce or eliminate emissions of pollutants, including a number of semi-volatile organic compounds (SVOCs), under the auspices of the Stockholm Convention (SC) on Persistent Organic Pollutants (POPs) and the Aarhus Protocol on POPs under the Convention on Long-Range Transboundary Air Pollutants (UN/ECE, 2010; UNEP, 2011).

Air is an important and effective transport route from sources of potential pollutants to the wider environment, and hence ambient air is a core matrix within the Global Monitoring Plan (GMP) framework as well as the European Monitoring and Evaluation Programme (EMEP) (UN/ECE, 1988; UNEP, 2011). Frameworks such as the GMP were established as a means to share information, create broad scale monitoring and to achieve some consistency (standardization or comparability) in monitoring procedures so that data are comparable (UNEP, 2011). Monitoring networks can also inform the understanding of the fate and transport of pollutants, which is essential in determining exposure in human and environmental health risk assessment.

Passive air samplers (PAS) have increasingly been used in regional and global scale air monitoring programs in the past

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decade to complement active air sampling, particularly the polyurethane foam (PUF) disk samplers (Hung et al., 2010; Jaward et al., 2004; Klánová et al., 2009). PAS are easy to use, do not require electricity and are relatively low cost compared to the alternative, active sampling (Harner et al., 2006a). These characteristics make PAS such as the PUF disk PAS (PUF-PAS) suitable to increase the spatial coverage of air monitoring networks, particularly in countries with low available resources and in high need of information (Poza et al., 2006). However, there are a number of factors that may affect the inter-comparability of passive sampling data between different monitoring networks or programs, as identified by Melymuk et al. (2014). For example, amongst the PAS most commonly used in monitoring networks (PUF-PAS, XAD-resin based PAS and XAD sorbent-impregnated PUF-PAS) (a) different deployment times, sample size and sampling frequencies, (b) PUF-PAS particle sampling assumptions, (c) the PAS sampling rate, as well (d) the spatial coverage of PAS networks are identified as key limitations for data compatibility (Melymuk et al., 2014).

In the present study we focused on the calculation of air concentrations of target compounds, which is currently perceived to be semi-quantitative, as a potential key limitation in ensuring inter-comparability of PAS measurements between monitoring programs (Melymuk et al., 2014). The amounts of compounds accumulated in PUF-PAS, for example, are routinely converted into air concentrations through application of sampling rates specific for a certain type of sampler (sorbent, housing). The sampling rates for PUF-PAS in particular can also be affected by physicochemical properties of compounds of interest (particle/gas partitioning), and environmental conditions at the site (temperature, wind speed) (Bohlin et al., 2014; Chaemfa et al., 2008; Klánová et al., 2008; Tuduri et al., 2006). For example, the sampling rates for PCBs increased by an order of magnitude between external wind speeds of $3.5\text{--}4\text{ m s}^{-1}$ and 7 m s^{-1} for the flying saucer configuration of the PUF-PAS (see Fig. S1 for a diagram of such a PUF-PAS configuration and Klánová et al. (2008) showed that the equivalent sample volume of the PUF-PAS for PAHs ranged from 300 m^3 to 400 m^3 at $-6\text{ }^\circ\text{C}$ and $150\text{--}250\text{ m}^3$ at $22\text{ }^\circ\text{C}$.

PUF-PAS sampling rates can be derived from (i) calibration studies based on parallel deployment of PAS and active air samplers (AAS) (Bohlin et al., 2014; Chaemfa et al., 2008, 2009a; Harner et al., 2013; Hazrati and Harrad, 2007; Klánová et al., 2008; Melymuk et al., 2011), (ii) use of depuration compounds (DCs), also termed performance reference compounds (PRCs) (Bartkow et al., 2004; Bohlin et al., 2010; Gouin et al., 2005; Harner et al., 2006b; Moeckel et al., 2009; Poza et al., 2006, 2012) and (iii) theoretical models (Harner, 2016; Petrich et al., 2013; Pevery et al., 2015). With the first method both gas-phase and particle-bound compounds are considered; however it can be influenced by the intermittent nature of many AAS records. For example, in some instances calibration studies are carried out using a high volume active air sampler (HVAAS) deployed for one 24-h period per week compared to month-long to three month-long periods, which are usual for PUF-PAS deployment (Harner et al., 2013; Klánová et al., 2008). Added to this, most calibration studies are case studies, i.e. one study at a specific period of the year and for relatively short periods (e.g., weeks to months), and thus may not account for seasonal influences (Bohlin et al., 2014; Chaemfa et al., 2008, 2009a; Melymuk et al., 2011). In reality, PUF-PAS for established monitoring programs (GAPS, EMEP, etc.) are deployed over the entire year (across seasons) and monitoring with PUF-PAS occurs for long periods (decades). DCs account for site-specific environmental conditions but are not applicable for particle-associated compounds. In addition, typically only a few DCs are used to calculate sampling rates for a more extensive list of analytes.

The choice of method to derive sampling rates adds uncertainty

to the determination of air concentrations, which can be compounded if the PUF-PAS has only partial efficiency for sampling particles, and hence particle-bound compounds (Melymuk et al., 2014). For example, for benzo[a]pyrene, a compound found primarily in the particle phase of the atmosphere, it has been estimated that only 8% of the air concentration may be captured by some PUF-PAS sampling configurations with potential losses possibly due to poor infiltration of particles into the PUF-PAS protective chamber, and hence under-sampling of particle-associated compounds by PUF-PAS (Melymuk et al., 2014). Therefore, our aim was to evaluate how well the theoretical PAS model (used to estimate the chemical-specific effective air sample volumes) described by Shoeib and Harner (2002) works to replicate the same levels and trends as seen in AAS at a long-term monitoring site. As passive sampling theory applies only to gas-phase chemicals a particle phase sampling rate as a fraction of the gas-phase sampling rate was estimated using long-term monitoring data or derived from the literature. In addition, and an octanol-air partition coefficient (K_{OA}) based model was used to estimate the fraction of compounds that might be associated with particles entering the PUF-PAS protective chamber and settling on or infiltrating into the PUF disk. PAS monitoring sites are globally distributed and a variety of PAS housing configurations are used by different monitoring programs, and hence there exist a variety of potential site- and sampler-based influences that contribute uncertainty to the calculation of air concentrations calculation from PAS measurements. Therefore, in this study a particular focus is given to how the choice of site- and sampler-based parameters, such as sampling rate and PAS particle-sampling efficiency affect the calculation of these air concentrations.

Air concentrations were calculated from PUF-PAS measurements using two sets of input variables including, (i) "site-based" variables, where up to nine years of PUF-PAS and HVAAS measurements from the sampler deployment site were used to first derive the gas-phase sampling rate (R_g), a particle-phase sampling rate (R_p) and, from these, C_{air} , and (b) a set of "default" input variables, where typical values of R_g based on PAS theory (Shoeib and Harner, 2002) and R_p derived from the literature, i.e., field measurements (Harner et al., 2013; Markovic et al., 2015) were used to calculate C_{air} . The method used to determine air concentrations of SVOCs from PUF-PAS measurements was a template provided by the GAPS network (Harner, 2016) using calculations and theory originally described in Shoeib and Harner (2002), Finizio et al. (1997) and Harner and Bidleman, 1998. C_{air} were compared to HVAAS measurements from the same period, i.e., a set of reference values.

2. Materials and methods

2.1. Sampling site

Passive and active air samplers were deployed at the Košetice observatory, located in central Czech Republic ($N49^\circ35'$; $E15^\circ05'$) (Fig. S1, SM). The site is operated by the Czech Hydrometeorological Institute and ambient air monitoring has been carried out at Košetice since 1988 (Holoubek et al., 2007b). The Košetice observatory is part of the European Monitoring and Evaluation Programme (EMEP), and hence, the monitoring design is based on the EMEP POP monitoring strategy (EMEP, 2009; Holoubek et al., 2007b). It is a background site located in a region with a moderately warm, moderately humid, highland climate (Vaña et al., 2001).

2.2. Air sampling, meteorological and particle data

HVAAS and PUF-PAS measurements for a range of SVOCs

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