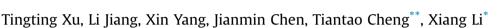
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Design and application of a novel integrated microsampling system for simultaneous collection of gas- and particle-phase semivolatile organic compounds



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HIGHLIGHTS

- An integrated microsampling system was used for atmospheric study.
- Theoretical and experimental verification confirmed the feasibility of this integrated microsampling system.
- The integrated microsampling system performed very well when applied to ambient aerosol sampling.
- The system is a simple, rapid, reusable, and cost-effective screening tool.

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ABSTRACT

An integrated microsampling approach based on active microextraction was developed to study semivolatile organic compounds (SVOCs). This microsampling system included an in-tube hollow-fiber solidphase microextraction device in combination with a sorbent tube that can be applied in parallel to collect gas- and particle-phase SVOCs. The preparation procedure, theory, and application of two devices were characterized and validated by the single fiber theory and scanning mobility particle sizer experiments. The influence of the optimization parameters (sampling time, flow rate, and breakthrough volume) on the extraction process was studied in detail. The performance of the system was tested via simultaneous collection of a range of gas and particle samples of ethylbenzene, xylenes, n-alkanes, polycyclic aromatic hydrocarbons, and monoterpenes. The collection efficiencies of the two devices were more than 90% for most target compounds. Typically, relative standard deviation values in the range 2-6% were obtained, depending on the compound. The calibration curves for each compound were reproducible and linear over the concentration ranges normally found in atmospheric samples. Both devices were used to collect ambient air samples during a haze period and performed well in the extraction of both gas- and particlephase SVOCs at the ng m^{-3} levels. The results from phase studies were used for further evaluation of the gas/particle partitioning of SVOCs. Application of this integrated microsampling system in the field validated its use as a simple, rapid, reusable, and cost-effective screening tool.

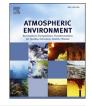
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1. Introduction

The physicochemical properties of atmospheric organic aerosol remain largely unclear, largely because we do not understand the fate of gas- and particle-phase volatile and semivolatile organic

http://dx.doi.org/10.1016/j.atmosenv.2016.11.022 1352-2310/© 2016 Elsevier Ltd. All rights reserved. compounds (VOCs and SVOCs). Gas-phase SVOCs, and VOCs with lower vapor pressures, can partition to the particle phase and then form secondary organic aerosol (SOA) after undergoing photooxidation in the atmosphere (Moise et al., 2015). Gas/particle partitioning phenomena play an important role in determining SOA formation processes and growth rates, which in turn affect global climate and human health (Scott et al., 2015; Zhao et al., 2013). Thus, the understanding and quantification of gas/particle partitioning are crucial for accurate estimates of SOA budgets.







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Numerous methods have been used for the sampling and analysis of atmospheric VOCs and SVOCs, from online to on-site and offline analyses (Parshintsev and Hyotylainen, 2015). Currently, mass spectrometry (MS) is the most commonly used technique for chemical analysis of these compounds, either alone or in combination with chromatographic methods (Barcelo, 1992; Hernandez et al., 2012). MS-based techniques provide the high sensitivity and selectivity required for the analysis of organic compounds. For the analysis of particle-phase SVOCs, aerosol particles are normally collected on filters or other substrates, with subsequent chemical analysis of the samples performed in the laboratory. Some filter types, namely, silica quartz (Maimone et al., 2011), glass fibers (Paolini et al., 2016), polytetrafluoroethylene (PTFE) (Moloi et al., 2002), microglass fibers with PTFE binder (Park et al., 2012), have been utilized most often for the sampling of particle-phase SVOCs with operation at high or medium volume flow conditions (ca. 70 or $1.0-2.3 \text{ m}^3 \text{ h}^{-1}$, respectively). The choice of filter is based on the ability to obtain consistently low organic blank levels. Quartz filters are far superior to others because of their thermal stability, high particle collection efficiency, and low cost (Parshintsev et al., 2011). Before use, quartz filters are usually baked at 550-700 °C for at least 6 h. However, gas-phase organic compounds are also adsorbed on quartz, resulting in positive artefacts (Maimone et al., 2011) in the measurement of particle-phase organics of 39%, or higher (Vecchi et al., 2009). The inhomogeneity of filter batches is also an issue, as filters from the same manufacturer may differ in capacity (Kirchstetter et al., 2001). After sampling, the target SVOCs are then extracted from filters by performing a Soxhlet (de Castro and Priego-Capote, 2010), microwave (Wang et al., 2016), ultrasonic bath (Aydin et al., 2007), solid-phase extraction (Andrade-Eiroa et al., 2016), pressurized liquid extraction (Carro et al., 2013), or accelerated solvent extraction (Gueguen et al., 2013), using polar solvents or mixtures. These preparation processes are timeconsuming and require the use of additional analytical equipment and hazardous materials. Finally, the extracts are analyzed by gas chromatography (GC) or GC/MS. It is clear that current filter sampling methods are inadequate for the accurate determination of concentrations of particle-phase SVOCs. Methods to overcome these deficiencies are not yet available.

For collection of gas-phase VOCs and SVOCs, routine methods depend on integration of the sample, and require pumping of air through a medium (e.g., an impregnated filter, sorbent, impinge, or cryogenic trap) that retains the target compounds (Hofmann et al., 1997; Koblizkova et al., 2012; Matsuda and Cahill, 1985; Yu et al., 1995). Earlier work utilized impregnated filters for sampling VOCs or SVOCs in air, where only some filters were impregnated, and then placed either on top of each other or in specially fabricated multiple filter holders (Henneken et al., 2006). Owing to its simple configuration, results are usually obtained with greater uncertainty. Cartridges for housing solid sorbents, namely, polyurethane foam (PUF) (Chaemfa et al., 2014), charcoal (Cecinato et al., 2012), polyvinylbenzene resins (XAD) (Armstrong et al., 2013), reversed-phase surface-modified membranes (Chan et al., 2005), and silicone gum (Ansorge et al., 2012), are also often used to trap gas-phase VOCs or SVOCs. The choice of sorbent is determined by several factors, such as collection efficiency, capacity, and chemical stability, but each sorbent material has its own advantages and disadvantages. For example, charcoal tubes were widely used for the collection of light VOCs with high efficiency, but the release of VOCs from charcoal is generally quite difficult (Peltonen and Kuljukka, 1995). Recently, PUF has gained popularity, particularly for use in high-volume samplers because of its low pressure drop, low blanks, low cost, and ease of handling. For some effective applications, it has been reported that high-volume samplers were coupled with a PUF plug as a back-up sorbent for sampling of gas-phase SVOCs such as polycyclic aromatic hydrocarbons (PAHs) (Delgado-Saborit et al., 2013; Wang et al., 2013). However, this simultaneous sampling technique can cause redistribution of PAHs between the gas and particle phase. There is extensive evidence that PUF is susceptible to breakthrough for light SVOCs (e.g., naphthalene) (Peters et al., 2000; Xie et al., 2014). Diffusion denuder samplers can avoid the occurrence of breakthrough, as reported by Ahrens et al. (2011). Here, gas-phase SVOCs were collected first using an XAD-coated annular denuder, which was followed by glass or quartz fiber filters for collection of particle-phase SVOCs. However, this sampling method requires additional solvent processing steps (Ahrens et al., 2011; Delgado-Saborit et al., 2013).

Overall, these traditional methods do not easily support combined rapid sampling and analysis carried out completely, or mostly, in the field. Tedious sample extraction processes must also be taken into account. In addition, sampling times vary with ambient loadings, sampling rates, substrate blanks, and the sensitivity of the analytical method, and typically range from several hours to upwards of a day. Increasing evidence shows that SVOCs retained on filters decrease if the sampling duration is extended (Arp and Goss, 2008; Galarneau et al., 2006). Thus, shorter sampling times have been recommended to minimize losses of SVOCs during sampling. Data from ambient air measurements showed that SVOC concentrations determined after 6 h of sampling were up to 6.5 times higher than those obtained with 24 h sampling periods (Peltonen and Kuljukka, 1995).

In light of these issues, newer simultaneous sampling methods have been proposed for gas- and particle-phase SVOC measurements. Solid-phase microextraction (SPME) has been combined with a needle-trap device to determine gas- and particle-phase compounds, for example, PAHs in barbecue and cigarette smoke (Li et al., 2010). In this approach, both particles and gas-phase compounds were collected, where gas- and particle-phase compounds were sampled actively with a needle-trap, and gas-phase species were extracted passively by SPME. After collection, samples were analyzed directly by GC or GC/MS. This combination enabled the determination of compounds in both phases, which is important for relating chemical concentrations to potential environmental effects accurately. This approach offered several advantages over other methods: a simple structure, easy operation, solvent-free analysis, and short sampling time (Eom et al., 2012; Heidari et al., 2016; Koziel et al., 2001; Li et al., 2010; Niri et al., 2009). However, there are also some disadvantages, such as limited extraction capacity of the needle trap device (NTD) (Vitta et al., 2010). According to Li et al. (2010), the breakthrough volume of the needle trap was only 80 mL for acenaphthylene, acenephene, and fluorene, and 100 mL for anthracene, fluoranthene, and pyrene. Additionally, SPME is not a quantitative extraction technique and requires careful calibration, which may be difficult to perform on-site. These various sampling issues resulted in large uncertainties in gas/particle partitioning. For this reason development of a new more accurate sampling system is needed.

In this study, we developed an improved simultaneous sampling system with increased accuracy and extraction capacity. First, an intube hollow-fiber solid-phase microextraction (IT-HF-SPME) device was designed for collecting gas-phase SVOCs. IT-HF-SPME is an extension of the current SPME technique, which is accomplished using many capillary columns that are integrated into a hollow glass tube. Compared with SPME, the IT-HF-SPME technique has an improved extraction capacity that facilitates dynamic sampling (Chafer-Pericas et al., 2007). The analytes can be extracted by flushing the samples through the capillary which preconcentrates them on the inner surface of the capillary columns (Moliner-Martinez et al., 2015). This can be accomplished easily, by combining a sampling pump with the device to pass the sample in Download English Version:

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