



A multi-residue method for the simultaneous analysis in indoor dust of several classes of semi-volatile organic compounds by pressurized liquid extraction and gas chromatography/tandem mass spectrometry



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ABSTRACT

People are exposed to multiple pollutants, especially indoors, in particular through ingestion of indoor settled dust. In the perspective of a cumulative risk assessment, a multi-residue analytical method based on pressurized liquid extraction (PLE) and gas chromatography/tandem mass spectrometry (GC/MS/MS) was developed for the simultaneous analysis in indoor dust of several classes of semi-volatile organic compounds (SVOCs) of health concern, from trace to highly concentrated compounds, including musk fragrances, organochlorines (OCs), organophosphates (OPs), polycyclic aromatic hydrocarbons (PAHs), polybromodiphenylethers (PBDEs), polychlorobiphenyls (PCBs), phthalates and pyrethroids. The method was validated in terms of limits of quantification (LOQ), and accuracy and precision via spiking experiments on an inert material (Celite® 545) and replicate analysis of the standard reference material SRM 2585 supplied by the National Institute of Standards and Technology (NIST). Method LOQs for 200 mg samples of sieved dust were 26 ng g⁻¹ for PCBs and some OCs, 65 ng g⁻¹ for musks, OPs, PAHs, PBDEs, pyrethroids, other OCs and some phthalates, 132 ng g⁻¹ for butylbenzylphthalate (BBP), 197 ng g⁻¹ for tributylphosphate and 1579 ng g⁻¹ for other phthalates. Quadratic calibration curves were established for each compound by analyzing at least five calibration solutions and exhibited determination coefficients higher than 0.999. The method was successfully applied to the SRM 2585 and seven real indoor dust samples. The results obtained on SRM 2585 demonstrate both excellent reproducibility and agreement with the indicative, reference or certified values and provide, for the first time, indicative concentrations for chlorpyrifos, diazinon, diisononylphthalate (DiNP) and tetramethrin. The results obtained on real dust samples illustrate the ability of the proposed method to quantify a wide range of SVOCs in a single analysis, making it appropriate for environmental monitoring programs or large-scale studies with a large number of samples.

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1. Introduction

People spend a considerable amount of time in indoor environments such as homes, workplaces and schools. As a result, exposure to indoor contaminants is of great concern due to numerous sources of contamination: introduction of outdoor materials, degradation of building materials, and widespread use of cleaning products, biocides, and electronic devices, as well as

indoor activities such as cooking or tobacco smoking. Among these indoor contaminants, semi-volatile organic compounds (SVOCs), that include a large number of chemicals such as pesticides, fragrances, plasticizers and flame retardants, have attracted growing interest over the past decade due to their possible health effects and the widespread exposure through different pathways: (i) non dietary ingestion of dust, (ii) ingestion of particles adhering to food, (iii) inhalation of air and airborne particles and (iv) dermal contact [1,2]. Among these different exposure pathways, ingestion of indoor settled dust is likely to be a non-negligible contributor to human exposure for some pollutants [3,4], especially for children due to hand-to-mouth behavior. Settled dust could also serve as a global indicator of indoor contamination [5], in particular because of the equilibrium partitioning of SVOCs between settled

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dust (particles deposited on surfaces, notably floors) and the two other compartments of the indoor environment, the gas phase and airborne particles (suspended particles in indoor air) [6,7].

A large number of SVOCs have been already investigated and identified in dust samples over recent years, including polybrominated diphenyl ethers (PBDEs) and other halogenated flame retardants, pesticides, phthalates, polycyclic aromatic hydrocarbons (PAHs), polychlorobiphenyls (PCBs), perfluorinated compounds (PFCs), alkaloids, alkylphenols and other phenol derivatives, dioxins and furans, synthetic musks, organophosphate esters (OPs), organotin compounds, parabens, polychlorobenzenes and ultraviolet (UV) filters (compounds designed to mitigate the deleterious effects of sunlight) [1]. For identifying chemicals of interest, a health ranking of SVOCs ingested through settled dust, based on toxicity and contamination data, was recently proposed [8]. Top-ranked chemicals were phthalates, pesticides, short-chain chlorinated paraffins, PBDEs, PFCs, organotins, PCBs, and PAHs. As many of them have reprotoxic and neurotoxic effects, it seems useful to get multi-residue methods for their simultaneous analysis in indoor dust samples. Such methods allow assessing cumulative exposure to these compounds, while taking into account economic constraints associated with environmental monitoring programs or large-scale studies with a large number of samples.

However, most previously published methods were dedicated to the analysis of a specific family of compounds, whereas multi-residue analytical methods were rarely reported [9–16]. So far, to the best of our knowledge, a method that allows the simultaneous analysis (same analytical procedure including preparation, extraction, clean-up and determination) in indoor dust of a wide range of SVOCs of interest belonging to different chemical classes has not yet been investigated, as often authors use several analytical procedures.

In order to determine the extent of the threat to public health posed by SVOCs indoors, it seems necessary to study the three compartments of the indoor environment (gas phase, airborne particles and settled dust) [1]. Regarding airborne particles, a multi-residue analytical method for the simultaneous determination of 55 SVOCs of health concern by thermal desorption coupled with gas chromatography/mass spectrometry (GC/MS) was recently proposed [17]. Investigated SVOCs were musks, organochlorines (OCs), OPs, PAHs, PBDEs, PCBs, phthalates and pyrethroids. These were selected according to their health interest [8] and their compatibility with a multi-residue GC/MS analysis.

The present work aims to optimize and validate a multi-residue method based on pressurized liquid extraction (PLE) and gas chromatography/tandem mass spectrometry (GC/MS/MS) for the simultaneous analysis of these same SVOCs in indoor settled dust. The objective of this study was to propose an efficient method that provides sufficient selectivity and sensitivity for all the target SVOCs covering trace (such as PCBs and PBDEs) and highly concentrated compounds (such as phthalates), while being appropriate for environmental monitoring programs or large-scale studies with several hundreds of samples. This involved compromises between analytical performances and costs of analysis. To achieve this, several solutions were considered: (i) automation of the extraction procedure, (ii) simplification of the clean-up procedure (without fractionated elution), making it suitable for all compounds, to reduce sources of error, time and cost, (iii) use of some native surrogate standards to monitor recoveries instead of labelled standards that are generally very expensive, and (iv) use of tandem mass spectrometry that provides excellent selectivity and sensitivity by minimizing matrix interference and background noise. The accuracy and precision of the proposed method were assessed via replicate analysis of the standard reference material SRM 2585 supplied by the National Institute of Standards and

Technology (NIST), and its applicability tested for several real dust samples.

2. Experimental

2.1. Selection of SVOCs

SVOCs were selected using a ranking method based on toxicity and indoor exposure levels [8]. A literature review provided data on the occurrence of chemicals and indoor settled dust concentrations. The toxicity reference values were retrieved from toxicity databases or were calculated from No Observed Effect Levels (or Lowest Observed Effect Levels) and uncertainty factors. The chemicals at the top of the ranking list were phthalates, pesticides, short-chain chlorinated paraffins, PBDEs, PFCs, organotins, PCBs, and PAHs. 55 SVOCs among these top-ranked chemicals were then selected according to their compatibility with a multi-residue GC/MS analysis. Investigated compounds are mentioned in Table 2.

2.2. Reagents and chemicals

Acetone and dichloromethane (DCM) (PLUS-for residual pesticide analysis) were purchased from Carlo Erba Reagents (Val de Reuil, France). Certified standards of aldrin, *cis*- and *trans*-chlordane, 4,4'-DDE, 4,4'-DDT, dieldrin, *alpha*-endosulfan, endrin, heptachlor, *alpha*-HCH, *gamma*-HCH, metolachlor, chlorpyrifos, diazinon, dichlorvos, atrazine, oxadiazon, cyfluthrin, cypermethrin, deltamethrin, permethrin, tetramethrin, tributylphosphate, acenaphthene, anthracene, benzo[a]pyrene, fluoranthene, fluorene, phenanthrene, pyrene, PCB 77, PCB 105, PCB 126, butylbenzylphthalate (BBP), di-*n*-butylphthalate (DBP), di(2-ethylhexyl)phthalate (DEHP), diethylphthalate (DEP), diisobutylphthalate (DiBP), diisononylphthalate (DiNP), dimethoxyethylphthalate (DMEP), dimethylphthalate (DMP), fenchlorphos (surrogate standard), fenprothrin (surrogate standard), methoprotrotryne (surrogate standard), and 2,3,4-trichloronitrobenzene (TCNB, internal standard (ISTD)) were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Purity of certified standards was above 97%, except for permethrin (94%). It should be noted that DiNP, cyfluthrin, cypermethrin, permethrin and tetramethrin were acquired as a mixture of isomers. Individual standard stock solutions (1 g L^{-1}) were prepared in acetone by accurately weighing 25 mg ($\pm 0.1 \text{ mg}$) of certified standards into 25 mL volumetric flasks, and then stored at -18°C . Nonane solutions (50 mg L^{-1}) of BDE 85, 99, 100 and 119 were purchased from Wellington Laboratories (Guelph, ON, Canada). Cyclohexane solutions (10 mg L^{-1}) of galaxolide (HCHB) and tonalide (AHTN) were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). A mixture (PCB Mix 21) containing 10 mg L^{-1} of 8 PCBs (PCB 28, 31, 52, 101, 118, 138, 153 and 180) in cyclohexane was supplied by Dr. Ehrenstorfer GmbH (Augsburg, Germany). Calibration solutions were prepared by appropriate dilution of individual standard stock solutions and commercial solutions in dichloromethane.

The Standard Reference Material SRM 2585 (Organic Contaminants in House Dust) was purchased from the National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA).

Celite[®] 545 was purchased from Merck KGaA (Darmstadt, Germany). Chromabond[®] NH₂ (aminopropyl modified silica) glass columns (3 mL/500 mg) were purchased from Macherey-Nagel GmbH & Co. KG (Düren, Germany).

2.3. Sample collection and preparation

The real dust samples were taken from household vacuum cleaner bags collected in French dwellings. After collection, samples were returned to the laboratory and stored at 5°C until sieving.

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