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Characterise sources for exposure assessment of chemicals in indoor environment

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Smart new strategies to design, produce and use safer chemicals to minimise risks and prevent pollution are a priority for the U.S. Environmental Protection Agency (EPA). EPA is developing a comprehensive approach to enhance the Agency's management of existing chemicals and emerging materials under the Toxic Substances Control Act (TSCA). EPA's Office of Research and Development (ORD) has been conducting research to support safer, more sustainable use of chemicals in consumer products, building materials and chemicals used for other purposes, and to better protect human health. Many of the EPA's priority pollutants, including formaldehyde, per- and polyfluoroalkyl substances (PFASs), polychlorinated biphenyls (PCBs), flame retardants, etc., are released from a vast number of building materials and consumer products. The cutting edge research summarised here has improved the methods and techniques used to measure and model emissions of these indoor air chemical contaminants present in various buildings, e.g. schools, office buildings and residences. Our research develops new approaches and technologies to better identify and quantify sources of indoor air pollution and helps to inform and refine estimates of indoor exposure and evaluate risk management options.

Background

In the modern world, changes in building design to improve energy efficiency, advances in construction technology using synthetic materials, and existing and new chemicals used in building materials and consumer products have led to a more comfortable life while at the same time potentially producing high concentrations of airborne contaminants. Indoor air quality is critical to human health because people spend nearly 90% of their time indoors, where typical contaminant levels are two to five times higher than outdoors.[1]

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Pollutant sources surround us on a daily basis. Tens of thousands of chemicals are in use and hundreds more are introduced into the market every year. A variety of chemicals are used as additives or reactants in products and emitted from building materials, furnishings, consumer products, human activities such as cooking and cleaning, as well as various other sources. Volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) are among the broad classes of compounds found in indoor air. Exposure of people to these chemicals occurs via inhalation, dermal adsorption and ingestion. Exposure to indoor air pollutants has the potential to cause disability, disease and even death, especially for sensitive populations like children and the elderly. For example, exposure to formaldehyde can result in respiratory symptoms and irritation of the eyes, nose and throat[2] and may be linked to nasopharyngeal cancers and leukemia.[3] SVOCs such as PFASs, PCBs and organophosphorus flame retardants (OPFRs) accumulate in the human body because of their slow elimination rates. Acute and chronic adverse health effects, including carcinogenicity, immune and endocrine system disruption, and adverse reproductive and neurodevelopmental effects related to animals and humans have been reported for these chemicals.[4–7]

The TSCA, passed by the United States Congress in 1976 and administered by the U.S. EPA is our nation's primary chemical management law. TSCA regulates the introduction of new or already existing chemicals. The Frank R. Lautenberg Chemical Safety for the 21st Century Act, signed by the President in June 2016, amends TSCA and requires EPA to evaluate the safety of existing chemicals via a three-stage process: prioritisation, risk evaluation and risk management.[8] EPA's ORD has been developing methods to support activities associated with implementation of the TSCA. ORD's Chemical Safety for Sustainability (CSS) National Research Program supports EPA's priority of reducing risks associated with exposure to chemicals in commerce, the environment, products and food using high-throughput risk-based evaluation methods. Its ultimate goal is to enable EPA to address the impact of existing chemicals, anticipate the impact of new indoor chemicals and materials and evaluate complex interactions of chemical and biological systems to support EPA's decision-making processes.[9]

Experimental measurements and mathematical modelling are both essential components in exposure assessment. Model development depends heavily on experimental data even though generic defaults can be used in available transport, fate and exposure models. Collection of measured data is important to refine and improve high-throughput modelling of chemical exposure, especially for exposure pathways with limited data that characterise emissions from the source and/or contact with the receptor and for data-poor parameters.[10] Currently, data collection is lagging far behind the model development, because laboratory and field testing is costly, time-consuming and technically challenging to appropriately characterise chemicals in a broad range of indoor environments.

The mass transfer mechanisms for VOCs and SVOCs that are responsible for pollutant transport from sources to indoor air, building materials and dust include absorption, desorption, material/air, dust/air and material/material partitioning and particle formation. Particulate matter, either suspended or settled, plays an important role in human exposure. The interaction of SVOCs with airborne particles and settled dust is even more complex because of the particle size, porosity, chemical composition and other properties.

Understanding the transport mechanisms from the sources of these compounds into air and on house dust and interior surfaces in the indoor environment, and the development of experimental methods to estimate model parameters in the fate of indoor chemicals' and transport modelling are critical to help characterise human exposure.

This editorial summarises several indoor air research projects conducted by the EPA/ORD National Risk Management Research Laboratory. These projects characterise the sources for exposure assessment of chemicals in the indoor environment to support EPA's CSS National Research Program and EPA's chemical safety assessments, as well as to address examples of interest to EPA program office partners.

Formaldehyde source emissions and sink effect

Formaldehyde has been of special concern as an indoor air pollutant because of its use in a wide range of products and the adverse health effects associated with exposure to this chemical. The Formaldehyde Standards for Composite Wood Products Act (S.1660) was signed into law in 2010. This far-reaching legislation requires that EPA establish processes to test, certify and label pressed-wood materials and finished goods. Additionally, EPA has been conducting registration reviews for antimicrobial biocides that release formaldehyde via the Reregistration Eligibility Decision process and Integrated Risk Information System (IRIS) formaldehyde assessment.

For EPA to address the potential risks to humans resulting from the use of composite wood products and biocides in occupational and residential settings, chamber studies were conducted to determine the amount of formaldehyde off-gassing from a variety of sources. The research activities include (1) development of emissions data and parameters for formaldehyde emitted from paint that contained formaldehyde-releasing biocides and 'conventional' and 'green' building materials and investigation of the effect of relative humidity on formaldehyde emissions and associated sink effects;(2) determination of partition and effective diffusion coefficients of formaldehyde for building materials under different relative humidity conditions that were used in formaldehyde mass transfer models; (3) measurement of formaldehyde emissions from composite and solid wood products in a full-scale chamber at different ventilation rates for up to 4000 hours to assess formaldehyde emissions from composite wood products; (4) development of a formaldehyde reference material to calibrate and assess the formaldehyde measurement method and eliminate the uncertainties involved for formaldehyde emissions testing; and(5) generation of formaldehyde Henry's law constants and the overall mass transfer coefficients for evaluating and developing improved emission models for formal-dehyde from the use of household and personal care products containing formaldehyde-releasing biocides.

Perfluorinated chemicals in consumer products and articles

Perfluorinated chemicals (PFCs) such as perfluorocarboxylic acids (PFCAs) have been found in numerous consumer products. Trace amounts of PFCAs have regularly been detected in humans, wildlife and environmental media. PFCAs came to the attention of EPA because of their widespread use, developmental toxicity in laboratory animals and other adverse health effects.EPA launched the perfluorooctanoic acid (PFOA) Stewardship Program, in which eight major manufacturers were committed to reducing global facility emissions and product content of PFOA and related chemicals by 95% by 2010 and to work toward eliminating emissions and product content by 2015. EPA also promulgated Significant New Use Rules under TSCA for long-chain perfluoroalkyl carboxylate (LCPFAC) chemical substances and perfluoroalkane sulfonates (PFSAs) chemical substances.[11] PFOA and PFOS are no longer manufactured in the U.S. as a result of this, with a few exceptions for limited industrial uses. However, PFOA and perfluorooctane sulfonic acid (PFOS) are still produced in other places around the world and they may continue to be imported into the U.S. in consumer goods.

Our study has been focused on how articles of commerce (AOCs) containing or treated by fluoropolymers and fluorotelomers impact human exposure in homes and offices. These research efforts were to (1) develop analytical methods to analyse PFCAs and the precursors, fluorotelomer alcohols (FTOHs), in AOCs;(2) apply the analytical methods that have been developed to identify the major PFCA sources and rank these sources in terms of source strengths in non-occupational indoor environments by determining the content of these chemicals in over one hundred AOCs in up to 13 article categories, including pre-treated carpeting, commercial carpet-care liquids, household carpet/fabric-care liquids and foams, treated apparel, treated home textile and upholstery, treated non-woven medical garments, treated floor waxes and stone/wood sealants, treated food contact paper, membranes for apparel, thread sealant tapes and pastes, non-stick cookware, dental floss and plaque removers and other miscellaneous AOCs; (3) conduct an independent assessment on the reduction of PFCA contents in consumer products collected from the U.S. market, primarily driven by the PFOA Stewardship Program; and (4) investigate FTOH sources and the fate of FTOHs in consumer products in the indoor environment.

PCB contamination and mitigation in schools

Protecting children from harmful chemical exposures is a top priority for EPA. PCBs, marketed as ‘Aroclors’ and other trade names, were commonly used in public and commercial building-construction materials from the 1950s through the late 1970s in the United States. Over 800,000 buildings that comprise 12 billion square feet (1,114,836,480 m^2) of interior space are estimated to have been constructed between 1958 and 1971.In addition, 46% of schools in the U.S. (approximately 55,000 schools) are estimated to have been built during that time based on results from a survey of indoor air quality programmes in schools.[12] Caulk, sealants, paint, fluorescent light ballasts and other products manufactured with PCBs are the primary sources of PCBs in buildings such as old school buildings. EPA announced new guidance for school administrators and building managers with important information about managing PCBs in caulk and tools to help minimise possible exposure in 2009. Our research was to determine the sources and levels of PCBs in schools, evaluate different strategies to reduce exposure and identify the best long-term solutions to minimise exposure.

We have investigated the potential exposure to PCBs in schools and other buildings by conducting laboratory studies, where we: (1) characterised potential primary sources of PCB exposure, such as caulk, coating and light ballast, in schools, and provided new data and models for ranking the primary sources of PCBs; (2) investigated transportation of PCBs, including from primary to secondary sources, the subsequent emissions to the air and migration to the dust; and (3) evaluated mitigation methods to reduce exposure to PCBs in caulk and other sources, including encapsulation and chemical destruction. The experimental data provided necessary knowledge and parameters to develop a general purpose computer program for dynamic modelling of emissions, transport and sorption of SVOCs in the indoor environment.

SVOCs in indoor environments

Many of the EPA's priority pollutants under TSCA are SVOCs and non-volatiles, and these pollutants are released from a vast number of building materials and consumer products.[13] Due to their low volatility, SVOCs are emitted slowly but produce long-term effects in the indoor environment. Better understanding of the transport mechanisms of these compounds from sources to air, airborne particles, house dust and interior surfaces in the indoor environment is essential to estimate indoor exposure. The existing mass transfer models for diffusion sources and sink materials include models based on Fick's Second Law for porous materials and the Langmuir and Freundlich models for nonporous materials. Material/air partition coefficients (K_{ma}), solid-phase diffusion coefficients (D_m), adsorption/desorption rate constants (e.g. k_a and k_d), mass-transfer coefficients (k_g), and initial material phase concentration (C_0) are critical parameters in these models. The experimental challenges that hindered advancement in the understanding of indoor SVOC emissions and fate are their low concentrations in the air, difficulty of measuring the slow emissions and strong sorption. The development of relatively rapid and practical experimental methods to estimate these model parameters are therefore a prerequisite for the application of the models, which has since attracted considerable research interest in the literature.

Using OPFRs, tris(2-chloroethyl) phosphate, tris (1-chloro-2-propyl) phosphate and tris(1,3-dichloro-2-propyl) phosphate, as the representative of SVOCs, we have: (1) developed and improved a dual SVOC chamber testing method to characterise the sorption properties of SVOCs on building materials and consumer products such as concrete, ceiling tile, vinyl flooring, carpet, latex-painted gypsum wallboard, open cell polyurethane foam, mattress pad and liner, polyester clothing, cotton clothing and uniform shirts; (2) estimated critical parameters of mass transfer models, including material/air partition and solid-phase diffusion coefficients, for SVOCs in those building materials and consumer products by combining a sink model and the experimental data; (3) measured and modelled surface sorption dynamics of SVOCs on impervious surfaces to inform SVOC source emission chamber testing; (4) compared measurement methods for parameters controlling the emissions of SVOCs in indoor environments and examined a rapid parameter estimation method based on the data obtained; and (5) investigated the partitioning of SVOCs between the gas phase and settled dust and migration of SVOCs from materials into house dust settled on the surface materials.

Spray polyurethane foam insulation research

Spray polyurethane foam (SPF) is produced on-site by mixing and spraying reactive chemicals for insulation and as an air sealant. During application, chemicals contained on 'Side A' react exothermically with polyols contained on 'Side B' to form the cellular structure of the SPF. Side A contains very reactive chemicals known as isocyanates. Side B contains a polyol, which reacts with isocyanates to make polyurethane, and a mixture of other chemicals, including catalysts, flame retardants, blowing agents and surfactants.[14] There is a potential for eye, skin and inhalation exposure to hazardous chemicals with all types of SPF products. EPA and other federal agencies have been investigating emissions and other implications associated with the use of SPF insulation (SPFI). We are working on developing full-scale emissions test protocols to characterise application and post application SPFI emissions using EPA's in-house indoor air research facilities. The methods, data and protocols developed from the study will be used to evaluate emission predictions in buildings where sources are located in conditioned and unconditioned spaces and provide the scientific basis for ASTM International consensus methods for measuring and modelling emissions from SPFI that EPA and others can use to evaluate and reduce exposures and risks of SVOCs.

Ending remarks

In summary, these combined research activities have been identified general principles and guidelines for extending the overall approach to assess and manage indoor air pollutants. Results will advance computational exposure models required to transform chemical evaluation. Using these research results, scientists can gain a deeper understanding of the hazards and risks of a large number of chemicals and better develop, evaluate and apply measurement-based modelling tools to be used in parameterisation and evaluation of individual models of near-field exposure and the development of integrated and evaluated consensus models for rapidly predicting human exposures to chemicals. These research activities support EPA Program Offices and other stakeholders as they refine existing risk assessments and make policy decisions to minimise exposure and protect human health and the environment from thousands of existing and emerging chemicals indoors.

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