

## Changes in indoor pollutants since the 1950s

Charles J. Weschler<sup>a,b</sup>

<sup>a</sup> Environmental and Occupational Health Sciences Institute, University of Medicine and Dentistry of New Jersey and Rutgers University, Piscataway, NJ 08854, USA

<sup>b</sup> International Centre for Indoor Environment and Energy, Technical University of Denmark, DK-2800 Lyngby, Denmark

### A B S T R A C T

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Over the past half-century there have been major changes in building materials and consumer products used indoors. Composite-wood, synthetic carpets, polymeric flooring, foam cushioning, plastic items and scented cleaning agents have become ubiquitous. The same is true for mechanical and electrical appliances such as washer/dryers, TVs and computers. These materials and products emit an array of chemicals including solvents, unreacted monomers, and additives. The consequent changes in emission profiles for indoor pollutants have been accompanied by modifications in building operations. Residences and non-residences are less ventilated than they were decades ago. Air-conditioned buildings are more numerous, especially in certain parts of the world. Most of these recirculate a high fraction of their air. The personal habits of building occupants, including the fraction who smoke indoors, have also changed. Taken together, these changes have altered the kind and concentrations of chemicals that occupants are exposed to in their homes, workplaces and schools. Since the 1950s, levels of certain indoor pollutants (e.g., formaldehyde, aromatic and chlorinated solvents, chlorinated pesticides, PCBs) have increased and then decreased. Levels of other indoor pollutants have increased and remain high (e.g., phthalate esters, brominated flame-retardants, nonionic surfactants and their degradation products). Many of the chemicals presently found in indoor environments, as well as in the blood and urine of occupants, were not present 50 years ago. Given the public's exposure to such species, there would be exceptional value in monitoring networks that provided cross-sectional and longitudinal information regarding pollutants found in representative buildings.

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### 1. Introduction

The chemicals found indoors are constant in neither kind nor concentration. Changes occur day-to-day, month-to-month, year-to-year and decade-to-decade. Chemicals that building occupants are exposed to today are substantially different from those that occupants experienced 50 years ago. Knowledge of such differences can aid in unraveling the effects that pollutants have on multiple aspects of human health.

This review presents general trends in the concentrations of indoor pollutants since the 1950s. It focuses on the United States, but the trends discussed have also been observed in other industrialized countries. Some of the restrictions or bans on certain chemicals may have occurred earlier or later in different parts of the world, but, to a large extent, the same chemicals have eventually been regulated. Many of the manufacturers of the materials, furnishings and products used indoors are international. Indeed, globalization has increased the extent to which indoor

environments in the United States, Europe, Asia and other parts of the world have come to resemble one another.

Initially, because of concerns with outdoor pollution and the understanding that outdoor pollution impacted indoor environments, the indoor pollutants that received the greatest attention were noxious substances that originated outdoors, especially sulfur dioxide, nitrogen oxides, ozone and airborne particles (Biersteker et al., 1965; Andersen, 1972 and references therein; Yocom et al., 1971; Sabersky et al., 1973; Thompson et al., 1973; Shair and Heitner, 1974). Subsequently attention turned to pollutants that were of particular concern indoors and readily measured; these included formaldehyde, radon, asbestos, tobacco smoke and nonpolar volatile organic compounds (National Research Council, 1981 and references therein). Over time, pesticides (Lewis, 2001 and references therein) and other semivolatile organic compounds (Weschler, 1980, 1984; Liroy et al., 1985) were measured indoors. As better analytical instruments were developed and instrument sensitivities improved, the number and types of compounds measured indoors increased. This was particularly true for organic compounds measured by capillary gas chromatographs interfaced to mass spectrometers. Presently researchers have begun to apply sophisticated techniques such as Proton Transfer Reaction-Mass

E-mail address: [weschlch@umdj.edu](mailto:weschlch@umdj.edu)

Spectrometry (PTR-MS) and Atmospheric Sampling Townsend Discharge Ionization Mass Spectrometry (ASTDI-MS) to measure species anticipated to be present in certain indoor settings (e.g., Wisthaler et al., 2005; Nøjgaard et al., 2007). Nonetheless, there remain compounds whose levels have not been directly measured, and whose presence indoors is only inferred (e.g., hydroxyl, nitrate, hydroperoxy and methylperoxy radicals).

How can we discuss indoor pollutant trends over a time-span that includes decades (i.e., prior to the 1970s) with few, if any, measurements of chemicals in indoor air? One way is to examine production figures for different chemicals over the time period of interest. This is a particularly valuable approach for chemicals that have primarily indoor uses such as certain plasticizers or flame-retardants. We can also look at the building materials that were common at different points in time. Given the composition of these materials, we can infer the major chemicals that they emitted. A similar approach applies to wall assemblies, floor coverings, architectural coatings, furnishings, cleaning agents and other products used indoors. However, the reader is cautioned that many of the inferences contained in this paper are “*best judgments*” about likely changes and do not have the certainty of findings based on direct empirical evidence.

The concentration of an indoor pollutant depends not only on its indoor emission rate, but also on the rate at which it is being transported from outdoors to indoors, and the rates at which it is scavenged by indoor surfaces, consumed by indoor chemistry and removed by ventilation or filtration. Changes in these source and sink terms are also examined in the present review.

Table 1 presents an admittedly subjective list of major events, actions and regulations that have affected the concentrations of pollutants in U.S. homes, offices and schools. Some of the entries refer to changes in the way that buildings were operated (e.g., air conditioning) or constructed (e.g., increased use of composite-wood products). Some refer to changes in the way that products were formulated (reduction of lead and mercury in paint, increased presence of synthetic fibers in carpets). Some refer to regulations that limited the use of certain products (pesticides, asbestos, chlorofluorocarbons). Some are deemed significant because they altered people’s thinking about environmental pollution in general (publication of “Silent Spring”) or indoor pollution specifically (publication of National Research Council report “Indoor Pollutants”). Some of the events listed were international in scope. Others are specific to the U.S., although analogous events often occurred in other countries. While a number of these will be called out in the course of this review, many are self-explanatory, and no further discussion will be provided. Regardless, stepping through the entries in Table 1 provides a sense of how and why an indoor environment in 2008 is so different from its counterpart in the early 1950s.

## 2. Building materials and products used indoors

### 2.1. Building materials

Numerous building materials emit chemicals into indoor air (Levin, 1989). This sub-section briefly discusses three that have come to dominate their respective categories, but were largely absent from buildings prior to the 1950s – composite-wood, PVC pipes and PVC wire/cable insulation.

**Composite-wood.** Following World War II, plywood began to replace solid wood in home construction, and, in the period from 1954 to 1975, U.S. plywood production rose from 4 billion to 16 billion  $\text{ft}^2\text{year}^{-1}$  (0.4–1.5 billion  $\text{m}^2\text{year}^{-1}$ ; APA, 2008). When plywood first came on the market, its adhesive resin was primarily urea-formaldehyde, and formaldehyde emission rates were relatively large ( $>1000 \mu\text{g m}^{-2}\text{h}$ ). Since then, both mandatory and

**Table 1**

Major events affecting indoor pollutants in the United States: 1948–2008.

1948	Chlordane first registered; uses include termite control
1950s	Synthetic-fiber tufted carpets begin to displace natural-fiber woven carpets
1950s	Rapid growth in “aerosol” air fresheners propelled by chlorofluorocarbons (CFCs)
1950	UL approves use of sprayed asbestos on structural surfaces in multi-storey buildings
1953	Over one million window air conditioners sold in the U.S. (43,000 sold in 1947)
1953	Industry standards reduce lead levels in paint to 1%
1954	U.S. plywood production reaches 4 billion $\text{ft}^2\text{year}^{-1}$ (0.4 billion $\text{m}^2\text{year}^{-1}$ )
1955	U.S. Air Pollution Control Act – first U.S. act addressing air pollution
1956	United Kingdom Clean Air Act; reduces smoke pollution; garners worldwide attention
1962	“Silent Spring” by Rachel Carson published
1962	90% of U.S. homes have TV sets
1963	Clean Air Act; initial enactment of legislation that will be revisited over next four decades
1964	First Surgeon General’s report on “Smoking and Health”
1965	Chlorpyrifos (Dursban®) registered
Mid-1960s	Permanent press clothing introduced – source of formaldehyde
Mid-1960s	Sales of indoor air fresheners grow; scents added to many household products
1970	U.S. EPA created
1970	Clean Air Act Extension requires EPA to protect public from airborne contaminants
1970s	Water-based (latex) paint displaces solvent-based (oil) paint for indoor applications
1972	DDT banned
1973	Arab Oil Embargo
1973	Arizona restricts smoking in public places
1973	U.S. EPA issues their first standards to reduce lead in gasoline
1973	U.S. EPA bans the use of sprayed asbestos for structural insulation and fireproofing
1975	Urea-formaldehyde foam insulation (UFFI) grows as an energy-saving measure in homes
1975	Andersen, Lundqvist & Molhave publish article on formaldehyde in Danish homes
1975	U.S. plywood production reaches 16 billion $\text{ft}^2\text{year}^{-1}$ (1.5 billion $\text{m}^2\text{year}^{-1}$ )
1975	Cars and light trucks manufactured with catalytic converters
Mid-1970s	Aerosol air fresheners replaced by other types due to concerns regarding CFCs & ozone layer
1976	Toxic Substances Control Act (TSCA) becomes law – regulates introduction of new chemicals
1978	All U.S. uses of mirex canceled
1978	Use of PCBs banned, except for totally enclosed applications
1978	EPA cancels use of chlordane on food crops and phases out other uses except for control of termites
1978	Paint with more than 0.06% lead banned for residential use
1978	First of the triennial international Indoor Air conferences, Copenhagen (Fanger and Valbjorn, 1978)
1978	Consumer Product Safety Commission (CPSC) proposes ban of benzene in consumer products
1979	Almost 25% of U.S. households have central air conditioning
1979	Oriented Strand Board (OSB) starts to gain market share
1981	National Research Council (NRC) issues report on “Indoor Pollutants”
1981	ASHRAE ventilation guidelines distinguish between smoking and nonsmoking spaces
1982	Consumer Product Safety Commission bans Urea-Formaldehyde Foam Insulation in homes/schools
1984	U.S. EPA restricts indoor use of pentachlorophenol (PCP); had been in paints and wood
1984	Finnegan et al. (1984) publish article on prevalence of sick building syndrome
1985	Department of Housing and Urban Development (HUD) limits allowable emission of formaldehyde from plywood and particleboard used in prefabricated and mobile homes
1985	U.S. EPA establishes Office of Indoor Air Quality
1986	Asbestos Hazard Emergency Response Act signed into law
1987	Montreal Protocol signed; substantial restrictions on chlorofluorocarbon (CFC) use
1987	U.S. EPA “TEAM Study” finds high exposures to indoor air pollutants
1988	U.S. EPA bans all uses of chlordane
1988	Indoor Radon Abatement Act directs EPA to identify areas with potential for high indoor radon

Table 1 (continued)

1989	U.S. EPA issues “Asbestos Ban and Phase-out Rule”; bans most asbestos containing products (overturned by courts in 1991, but ruling supports ban on specific products and “new uses”)
1990	Clean Air Act Amendments; address air toxics, gasoline formulation and evaporative emissions
1990	U.S. EPA bans mercury in interior latex paint
1992	U.S. EPA bans mercury in all interior paints
1994	California restricts smoking in workplaces
1995	Chlorofluorocarbon (CFC) production in the United States ends
Mid-1990s	Emissions from carpet backing and use of SBR backings decline (industry self-regulation)
1997	Almost 50% of U.S. households have central air conditioning
1998	EPA issues rule limiting VOC emissions from consumer products and architectural coatings
1998	National Research Council issues BEIR VI report “Health Effects of Exposure to Radon”
2000	3M voluntarily stops making certain perfluorinated compounds (e.g., PFOA and PFOS)
2000	More than 50% of U.S. homes have personal computers
2001	Registration withdrawn for chlorpyrifos (Dursban®) use in homes
2001	CDC publishes first National Report on Human Exposure to Environmental Chemicals
2004	Stockholm Convention on Persistent Organic Pollutants signed; global treaty
2005	Surgeon General releases National Health Advisory on Radon
2006	EU adopts Registration, Evaluation, Authorization and Restriction of Chemicals (REACH)

voluntary standards have been adopted that limit formaldehyde emissions, and the use of lower emitting phenol-formaldehyde resin has increased. Other composite-wood products that have been adopted by the construction industry include particleboard, oriented strand board (OSB) and medium density fiberboard. Each of these emits a mix of aldehydes and terpenoids (Baumann et al., 1999, 2000; Hodgson et al., 2002; Salthammer et al., 2003).

**PVC pipes.** PVC pipes have partially replaced copper pipes in many indoor plumbing applications, including drain, waste and vent systems, as well as water distribution systems. PVC piping is rigid and does not emit plasticizers the way that flexible PVC products do (e.g., vinyl flooring and wall covering). However, PVC pipes often contain organotin compounds as stabilizers, and these semivolatile compounds are expected to migrate into indoor environments over time.

**PVC wire/cable insulation.** Following World War II, flexible PVC insulation began to replace rubber and textile braid insulation on wiring and cable used in both residential and commercial buildings. The total length of insulated wire and cable in homes, offices and schools has increased dramatically with the introduction of more extensive telephone systems, computer networks and both cable and satellite TV distribution systems. Plasticizers are required to make the PVC insulation flexible. These plasticizers, primarily phthalate esters such as di-2-ethylhexyl phthalate (DEHP), are emitted over the life of the product.

## 2.2. Products used indoors

**Carpeting.** Prior to World War II, most carpets were woven and consisted of cotton or wool. Nylon was introduced in 1947, and other synthetic fibers followed. During the 1950s, tufted carpets made of nylon, rayon, and acrylics displaced woven carpets. In 1950, ~10% of carpets were tufted; today, more than 90% are tufted (Carpet and Rug Institute, 2008). Polyester and polypropylene (olefin) carpets were introduced in the mid-1960s. Stain repellants (e.g., fluorinated surfactants) were applied with increasing frequency to the carpet fibers, while the carpet backing systems (backing, adhesive and pad) were often treated with flame-retardants.

As interest in indoor air quality increased in the 1980s and early 1990s, carpets and carpet backings were identified as major sources

of indoor pollutants (e.g., Hodgson et al., 1993). In 1992, the Carpet and Rug Institute (CRI) initiated its “Green Label” program to test emissions from carpets, cushions and adhesives. This voluntary program provided a labeling scheme to help consumers, architects, builders and facility managers specify products with reduced emissions of organic pollutants. Over the years, CRI has tightened its IAQ standards, prompted, in part, by guidelines and regulations that have been implemented in Europe. At present, carpeting is more prevalent in the U.S. than in Europe or Asia.

**Flooring.** Linoleum was a common flooring material that became less popular after World War II. In the 1950s, asphalt tile was the most widely used flooring material, but vinyl flooring was gaining in popularity. Today, vinyl is the most common flooring material in the U.S. (excluding carpet). Vinyl flooring requires plasticizers to remain flexible and avoid brittleness. It typically contains 30–40% plasticizer by weight (Bornehag et al., 2005a and references therein). The dominant plasticizer for PVC flooring has been di-2-ethylhexyl phthalate (DEHP). As concerns about the potential health effects of DEHP have increased (see Section 6.4), other plasticizers (e.g., DINP) have been substituted.

**Paints.** For indoor applications, water-based (latex) paints began to replace solvent-based (oil) paints in the 1950s. Water-based paints emit fewer volatile organic pollutants than do solvent-based paints. Their growth increased significantly during the environmentally conscious 1970s, resulting in fewer VOC emissions during indoor painting. The type of binder used in water-based paint determines the residual monomers and degradation products that the paint subsequently emits. Initially, the dominant binder in water-based paint was styrene-butadiene latex; this was later replaced by vinyl acrylic and acrylic latex (Martens, 1981). Common binders today include vinyl, acrylic and vinyl-acrylic blends. The most commonly used coalescing agent in latex paint is Texanol®, a mix of 3-hydroxy-2,2,4-trimethylpentyl-1-isobutyrate and 1-hydroxy-2,2,4-trimethylpentyl-3-isobutyrate (Corsi and Lin, in press). Texanol® isomers are semivolatile and continue to be emitted from latex-painted surfaces for months after application (Shields and Weschler, 1992; Corsi and Lin, in press). They have become common indoor pollutants as latex paints have grown in popularity.

Recently so-called “green” or “natural” paints, based on sustainable natural ingredients, have gained in market share. These products typically contain unsaturated organic compounds (e.g., linseed oil, limonene, other terpenoids) that can react with ozone. The resulting oxidation products include secondary organic aerosols (Lamorena et al., 2007).

First lead and then mercury have been removed from interior paints (see Section 6.5). Formaldehyde is still used as an in-can preservative in some paints.

**Furnishings.** Veneer on composite-wood has replaced solid wood in many furnishings. It is common today for kitchen cabinets, dressers, bed frames, desks and similar products to be constructed primarily with medium density fiberboard or a similar composite-wood material. The cushioning used in bedding, sofas and chairs has evolved from feathers and down to synthetic foams. Since the 1960s, most synthetic foams have been treated with flame-retardants. For example, polyurethane foam used in home cushioning often contains 10–30% (by weight) brominated flame-retardants (Hites, 2004).

**Office cubicles.** The design and layout of office space has changed to include the use of office cubicles. These were introduced in the mid-1960s and have proliferated. Their partitions are typically constructed of fiber-glass board, chosen in part for its sound deadening properties. Other constituents include fabrics, adhesives and finishes/coatings. Given their large surface area, emissions from cubicles can meaningfully impact air quality in office environments, as became apparent from cubicle emission measurements conducted in the mid-1980s. These, coupled with lawsuits,

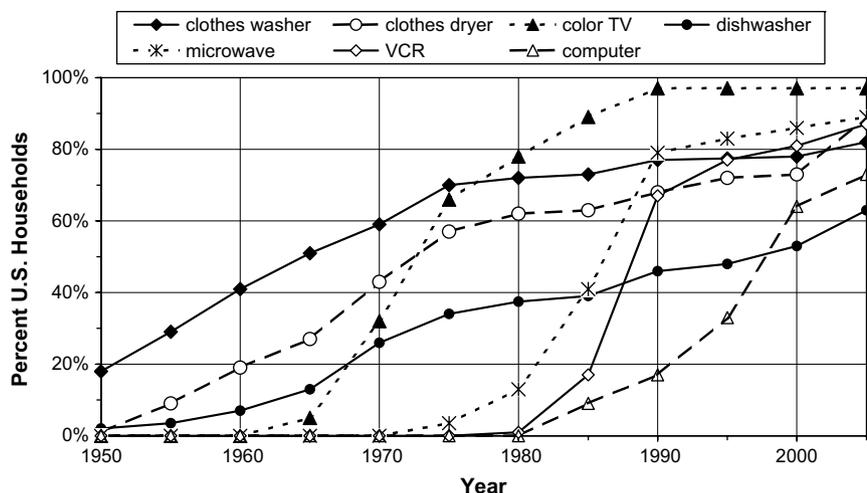


Fig. 1. Percent of U.S. households with selected appliances and electronic equipment for the period from 1950 to 2005 (adapted from Cox and Alm, 2008).

prompted an effort by manufacturers to reduce emissions. Between 1985 and 2005, the average level of formaldehyde emitted from cubicles decreased by 50%, chlorinated VOCs decreased more than 90% and total VOCs decreased 40–70% (Betts, 2005).

**Cleaning products.** Anionic surfactants such as fatty acid soaps, alkylsulfonic acid salts (e.g., linear alkyl benzene sulfonate), and fatty alcohol sulfates were commonly used in many detergents and cleaning products designed for home and commercial use. Over the past few decades, nonionic surfactants have been substituted for these anionic surfactants. An important class of nonionic surfactants is alkylphenol ethoxylates (APEs), especially nonylphenol ethoxylates, which constitute more than 80% of the APE market in North America (APE Research Council, 2008). These compounds have become common in the dust and air of indoor environments (see Section 6.4).

In the 1950s, benzene was found in many cleaning products, especially those containing a mix of aromatic solvents. Carbon tetrachloride was commonly used as a spot remover. Starting in the late 1980s there was a shift away from aromatic and chlorinated solvents. Both benzene and carbon tetrachloride have virtually disappeared from indoor cleaning products (see Section 6.3).

Various glycol ethers have been used as the active agents in window cleaners and multi-purpose cleaners designed to remove grease and grime (Nazaroff and Weschler, 2004; Singer et al., 2006a). The more toxic glycol ethers have been replaced with ethylene glycol monobutyl ether (2-butoxy ethanol), which is currently the dominant glycol ether used in cleaning products.

Today there is a trend towards “greener” cleaning agents, especially terpenoid based solvents. Many of the terpenoids (e.g., limonene,  $\alpha$ -terpinene,  $\alpha$ -terpineol, linalool) react at relatively fast rates with ozone and can serve as precursors for ozone oxidation products in indoor environments (Nazaroff and Weschler, 2004; Destailats et al., 2006; Singer et al., 2006b).

**Air fresheners.** In the early 1950s there was rapid growth in spray or “aerosol” air fresheners that were propelled by chlorofluorocarbons (CFCs). These products were typically ~1% active ingredients, ~24% solvent and ~75% CFC propellant. The delivery system produced fine droplets that spread out over a wide area and stayed airborne for minutes. In the mid-1970s Rowland and Molina (1974) published their *Nature* paper suggesting that CFCs were depleting the stratospheric ozone layer. Further research supported their hypothesis. The issue attracted wide public attention, and the use of aerosol air fresheners declined. The Montreal Protocol (1987) resulted in the elimination of CFC propelled air fresheners. However, other types of air fresheners took their place. These

included scented candles, spray products with alternative propellants, and, later, plug-in units. The sales of these products have continued to increase. In 2000, U.S. consumers spent \$0.9 billion on air fresheners; in 2005, \$1.5 billion (Storey, 2007). Industry testimony in the U.S. Federal Register (2007) stated that ~70% of U.S. homes use air fresheners. Many of the scenting agents used in air fresheners are unsaturated organic compounds that can react with ozone to produce both gas phase and condensed phase products (Liu et al., 2004; Singer et al., 2006a,b).

**Appliances.** Fig. 1 displays the percent of U.S. households with various appliances for the period from 1950 to 2005. The fraction of homes with clothes washers and dryers grew rapidly from 1950 to 1970, while the fraction with dishwashers grew at a slower rate. These products required the use of new, specialized detergents. The fraction of homes with microwaves grew from 3% in 1975 to almost 80% by 1990; these were typically unvented and emitted cooking fumes while in use. Gas ranges without pilot lights were introduced in the early 1980s and became mandatory on new models starting in January 1990. This feature eliminated an indoor source of carbon monoxide and nitrogen oxides. Following the energy crisis in 1973, there was a temporary increase in the indoor use of unvented kerosene heaters. Both the CPSC and EPA cautioned that these were sources of nitrogen oxides and sulfur dioxide, which led to better venting practices.

A number of appliances manufactured prior to 1978 had transformers and capacitors that contained PCBs. When these leaked, the emitted PCBs eventually redistributed throughout the indoor environment.

**Electronic equipment.** Destailats et al. (2008) have published a review of indoor pollutants emitted by modern office equipment. Many of the pollutants tabulated in their review were absent from office environments 50 years ago. Photocopiers have replaced carbon paper and duplicating machines. Certain types of carbon paper were sources of PCBs, while duplicating machines emitted solvent vapors. Photocopiers can be a source of ozone, styrene, formaldehyde, other aldehydes, semivolatile organic compounds (SVOCs) from heat transfer fluids, and particles (Leovic et al., 1996; Wolkoff et al., 1993; Wolkoff, 1999; Brown, 1999). Personal computers and printers have replaced typewriters. Their casings and circuit boards emit plasticizers and flame-retardants, especially brominated flame-retardants. Certain laser printers have been identified as sources of airborne particles (He et al., 2007) and ozone (Lee et al., 2001; Smola et al., 2002). Computer monitors have evolved from CRTs to flat-panel displays; the latter tend to emit fewer organics than the former.

Residences have also experienced increased emissions from electronic equipment. To a first approximation, emissions from home electronics tend to be similar to those from office electronics. Fig. 1 shows the percent of homes with TVs, VCRs and computers for the period from 1950 to 2005. Color TVs increased sharply from 1965 to 1975. A similar rapid increase occurred for VCRs during the period from 1985 to 1990 and for computers during the period from 1990 to 2000. These TVs, VCRs and computers were new indoor sources of plasticizers and flame-retardants. TVs with cathode ray tubes are presently being replaced by flat-panel TVs with presumably lower emissions. At the same time, home entertainment systems are growing in size and number of components, which may result in increased emissions.

**Clothing.** Synthetic fibers are more common in clothing today than at the end of World War II, but they have not displaced natural fibers to the extent that they have in carpets. Over the years, a higher percentage of clothes have been treated with flame-retardants or stain repellants. The use of mutagenic tris(2,3-dibromopropyl)phosphate (tris-BP) as a flame-retardant in children sleepwear received considerable attention in the late 1970s (Blum et al., 1978). This compound was replaced by a chlorinated species, which was also found to be mutagenic (Gold et al., 1978).

The number of dry-cleaning establishments grew throughout the 1950s, meaning more dry-cleaned clothes in U.S. homes. Emission of tetrachloroethylene from dry-cleaned clothing was reported in the U.S. EPA TEAM study (Wallace, 1987). In the 1960s “easy care” and “permanent press” fabrics were introduced. These fabrics were treated with formaldehyde resins, and had meaningful formaldehyde emissions (Kelly et al., 1999). More recently, lower formaldehyde emitting resins have been introduced.

### 3. Chemical transformations

#### 3.1. Oxidation reactions

Chemical reactions among indoor pollutants alter the mix and concentrations of indoor pollutants (Weschler and Shields, 1997; Weschler, 2004). Indoor oxidation reactions have received the greatest attention to date. Oxidation products include free radicals, secondary ozonides, epoxides, aldehydes, ketones, acids, diacids, dicarbonyls and other oxygenated species (Weschler, 2000, 2006). Some of the products have low vapor pressures and contribute to the growth of secondary organic aerosols (Weschler and Shields, 1999; Weschler, 2003; Sarwar and Corsi, 2007 and references therein). Circumstantial evidence suggests that indoor

concentrations of ozone reactive compounds such as terpenoids (e.g., limonene,  $\alpha$ -pinene, linalool,  $\alpha$ -terpineol) have increased due to their growing use in cleaning agents and air fresheners. As noted earlier, ozone reactive chemicals (e.g., linoleic acid and linolenic acid) are also found in “green” or “natural” paints. This inferred increase in the level of ozone reactive compounds in indoor settings may, in turn, have resulted in increased production of ozone-derived pollutants such as formaldehyde, acrolein, nonanal, decanal, nonenal isomers and secondary organic aerosols.

#### 3.2. Hydrolysis reactions

Hydrolysis reactions are another important class of indoor reactions. The plasticizers and flame-retardants found in many indoor materials are often phthalate or phosphate esters (Wensing et al., 2005). Such esters can hydrolyze (react with water) to yield alcohols and acids. The hydrolysis of PVC on moist concrete has been a recognized indoor air problem for more than two decades (Norback et al., 2000). More recently, the hydrolysis of Texanol<sup>®</sup> isomers, used as coalescing agents in latex paint, has been identified as a source of highly unpleasant butyric acid (Corsi and Lin, in press). Over the past 50 years more and more products that contain esters have entered the indoor environment. At the same time, the fraction of homes with moisture problems appears to be growing (see Section 5.3). Taken together, this suggests that hydrolysis reactions have a larger impact on indoor environments today than they did several decades ago.

### 4. Personal habits

#### 4.1. Smoking

In 1964, the first U.S. Surgeon General's report on “Smoking and Health” was issued. In 1973, the state of Arizona restricted smoking in public places. In 1994, the state of California restricted smoking in workplaces. In 2003, New York City amended its smoking restrictions to include all restaurants and bars. Today many U.S. state and local governments, as well as many businesses, have bans on smoking in various indoor environments. Furthermore, as illustrated in Fig. 2, the percentage of the U.S. adult population that smokes has been declining over the last 4 decades; in 1965, 43% of U.S. adults smoked; in 1985, 30% smoked; and in 2006, 21% smoked (CDC, 2007). Today, even in those households with smokers, a sizable fraction no longer smokes indoors.

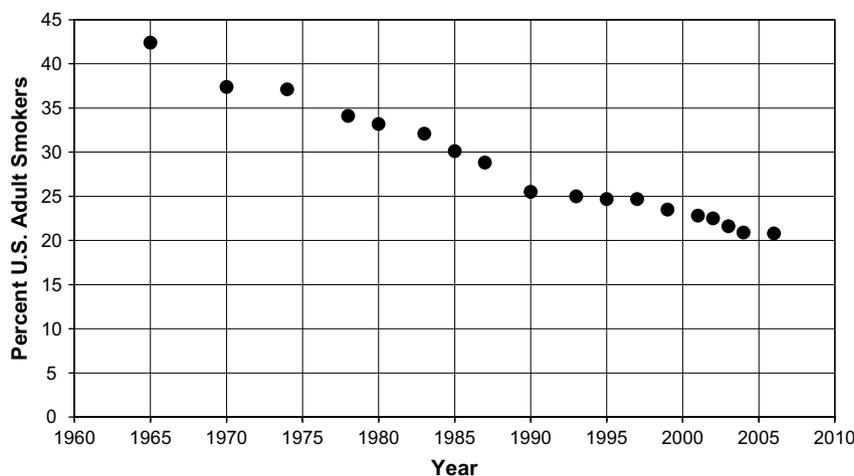


Fig. 2. Percent of U.S. adult smokers for the period from 1965 to 2006 (CDC, 2007).

Analogous restrictions and behavior changes have occurred in a number of European countries. As a consequence of these, there is less ETS in indoor environments and less exposure to ETS among nonsmokers in these countries. The latter has been documented for U.S. residents using biomarkers such as cotinine, a metabolite of nicotine that is present in the blood of nonsmokers exposed to ETS. Over the 11-year period from 1988–1991 to 1999–2002, blood cotinine levels for nonsmoking adults decreased by 75%; for adolescents, 69%; and for children, 68% (CDC, 2005). Although ETS levels have decreased in indoor environments, ETS exposure remains a major indoor health risk.

Anecdotal evidence suggests that in some indoor environments where there was previously smoking, ventilation rates were reduced after smoking was eliminated. This could result in increased concentrations of other pollutants emitted indoors, but unrelated to smoking.

#### 4.2. Time spent indoors

In 1950, 9% of U.S. homes had televisions; by 1954, more than 55% of U.S. homes had TVs; and by 1962, more than 90% of homes had TVs (TV history, 2008). By 2003, more than 60% of U.S. homes had personal computers (World Resources Institute, 2008). Concomitantly, the bandwidth of connections between home and the Internet has continued to grow, which results in more compelling Web-based applications. Over the past 15 years, the sales of electronic gaming units have mushroomed. For example, PlayStation® was introduced in 1994; by 2004, Sony had shipped 40 million units in North America and 100 million units worldwide (Sony Computer Entertainment, 2008). The above developments suggest that people, especially children, spend more time in their homes today than they did 50 years ago.

People are also in transit (another type of indoor environment) a larger fraction of the day than they were 50 years ago. As suburbs have grown, daily commutes by car or public transportation are longer. In large metropolitan areas (e.g., New York, Los Angeles, Atlanta), it is not unusual for workers to spend 3 h per day commuting.

#### 4.3. Pet ownership

Pet ownership, especially pets that are kept indoors, has increased (American Veterinary Medical Association, 2007). Hence,

it is anticipated that indoor environments contain more cat and dog allergens than they did several decades ago.

### 5. Building factors

#### 5.1. Tighter buildings

Steps to tighten building envelopes were included in energy conservation measures implemented following the Arab Oil embargo of 1973. Residential buildings constructed in the past two decades tend to be tighter and have lower air exchange rates than buildings constructed in the 1950s, 1960s and early 1970s (Weisel et al., 2005). Although conventional wisdom holds that the same trend applies to non-residential buildings, in a study of data from 139 commercial buildings, no correlation was seen between air leakage and building age (Persily, 1999).

Extensive literature reviews have concluded that low ventilation rates in offices are associated with building related health complaints (Seppanen et al., 2006; Wargocki et al., 2002). In a recent study of children living in 390 Swedish homes, Bornehag et al. (2005b) found that children with allergies lived in homes that had significantly lower ventilation rates than homes inhabited by children without allergies, and that there was an inverse dose–response relationship between ventilation rates and allergic symptoms. However, this association was for children living in Varmland, Sweden, where the outdoor air is quite clean. Whether such an association would be found in regions with highly polluted outdoor air remains to be determined. Indeed, the relationship between ventilation and health risks requires further scrutiny.

#### 5.2. Air conditioning

In the U.S., air conditioning (AC) in residential and commercial buildings has increased dramatically over the last six decades. In 1947, 43,000 window air conditioners were sold; in 1953, that number was more than one million. The rate of growth in air conditioning was greatest in the five-year period from 1967 to 1972 when the percent of households with some type of air conditioning grew from 20% to 48% (Cox and Alm, 2008). Fig. 3 shows the change in air conditioning status of U.S. households for the period from 1978 to 1997 (Energy Information Administration, 2000). During this period the percent of households with window air conditioning actually decreased slightly, while the percent with central AC more than doubled. By 1997, 47% of U.S. households had central

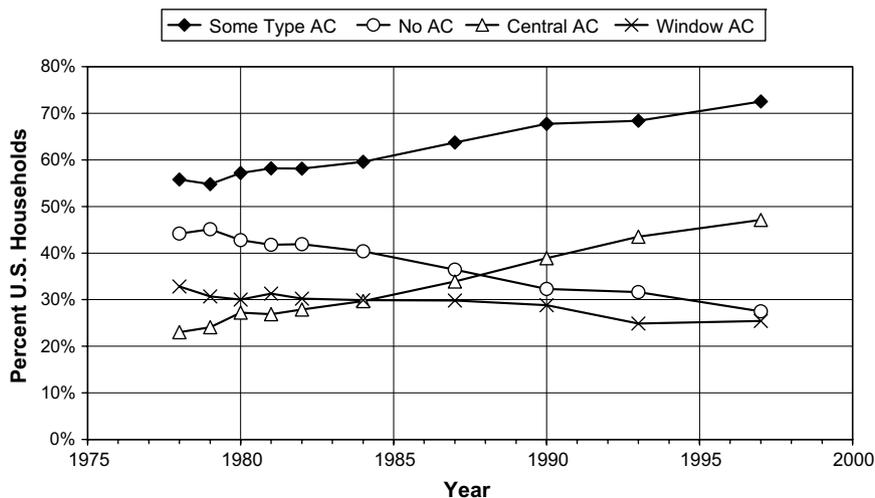


Fig. 3. Percent of U.S. households with some type of air conditioning, with no air-conditioning, with central air-conditioning and with window air-conditioning for the period from 1978 to 1997 (Energy Information Administration, 2000).

AC and 72% had some kind of AC. In the southern region of the U.S., that figure was 93% and most new homes were being constructed with central AC. To a major extent, air conditioning enabled the large influx of people into states such as Florida, Nevada and Arizona.

Air conditioning has changed the way that buildings are operated. Rather than opening windows and operating fans during periods of hot weather, occupants of air-conditioned homes close windows and turn on their “AC”. To save energy, home air conditioners tend to be operated in a mode where they recirculate most of the indoor air. In air-conditioned office buildings and schools, it is also common to recirculate indoor air, with more than 90% of the supply-air typically being recirculated-air. Hence air-conditioned buildings exchange indoor air with outdoor air at a much lower rate than buildings without air conditioning. Ventilation, instead of being high in warm weather, has become quite low.

For airborne pollutants with indoor sources, the reduced ventilation that accompanies the use of air conditioning results in higher indoor concentrations. On the other hand, air conditioning results in less outdoor-to-indoor transport of ozone, nitrogen dioxide and submicron particles. Additionally, in commercial buildings the installation of heating, ventilating and air conditioning (HVAC) systems has included filters that remove a fraction of particles from the airstream. Such filtration can meaningfully reduce indoor airborne particles, especially when efficient filters are employed (see *ASHRAE Standard 52-2, 2007* for information on removal efficiencies of HVAC filters).

### 5.3. Damper buildings?

As the number of homes in the U.S. has grown, houses have been built on land that was previously considered “wetlands”. There has also been a large increase in homes in hot, humid regions of the U.S. (e.g., Florida, Georgia, South Carolina), enabled by the growing availability of air conditioning. The combination of air conditioning and a hot, humid climate can often lead to moisture condensation on interior surfaces. Further aggravating matters, gypsum board has largely replaced plaster in home construction; the former does not provide the moisture buffering of the latter. Taken together, these changes suggest that the percentage of U.S. buildings with moisture problems has increased over the past several decades. This trend may be less pronounced in other countries.

## 6. Trends in indoor pollutants

Table 2 lists selected indoor pollutants and, for each pollutant, an up or down arrow to broadly summarize the trend in its indoor concentration since the 1950s. Most of these trends have been inferred; there are only a small number of pollutants for which indoor measurements exist over an extended period of time. The paragraphs in this section provide supporting information for the “best judgments” presented in the table. Compounds in Table 2 preceded by an asterisk are those for which substantial body burdens in U.S. residents have been reported (*NHANES, 2005*; see Section 6.6 and Table 3).

Some indoor pollutants are strongly influenced by outdoor concentrations; some have primarily indoor sources; and some are influenced by both outdoor and indoor sources. With the exception of short lived, highly reactive species, pollutants found outdoors will also be found indoors; changes in the concentrations of outdoor pollutants result in changes in the concentration of indoor pollutants. For relatively unreactive pollutants such as benzene or carbon tetrachloride, the indoor concentration will eventually equal the outdoor concentration in the absence of indoor sources. For reactive pollutants such as ozone or nitrogen dioxide, the indoor concentration depends on the air exchange rate; the larger

the air exchange rate, the larger the ratio of the indoor concentration to the outdoor concentration (*Shair and Heitner, 1974; Weschler et al., 1989*).

### 6.1. Inorganic gases

*Carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>) and sulfur dioxide (SO<sub>2</sub>)*. On average, the indoor concentration of each of these gases has decreased since the mid-1950s, partially due to decreases in their outdoor concentrations. The use of low sulfur fossil fuels has resulted in much smaller outdoor emissions of SO<sub>2</sub>. Catalytic converters, first introduced in 1975, have sharply reduced emissions of CO and NO<sub>x</sub> from motor vehicles. Indeed, the number of unintentional motor vehicle-related CO deaths decreased from 4.0 to 0.9 deaths/million-person-years between 1975 and 1996 (*Mott et al., 2002*). Indoor emissions of CO and NO<sub>x</sub> from gas appliances, including gas stoves, have decreased with the introduction of units that no longer use “pilot lights”. Less indoor smoking has further contributed to reduced indoor emissions of these pollutants.

*Ozone (O<sub>3</sub>)*. In contrast to carbon monoxide, nitrogen oxides and sulfur dioxide, the outdoor concentration of ozone has increased over the past five decades (with the exception of a few regions, such as southern California, that have implemented specific ozone-reduction measures). This is true not only in urban areas but also at rural and background sites (*London and Kelley, 1974; Seinfeld and Pandis, 1998; Derwent et al., 2002*). The increase in outdoor ozone levels has been tempered by average reduction in ventilation rates and concomitant decrease in the transport of ozone from outdoors to indoors. Reductions in outdoor-to-indoor transport are anticipated to be greater in those regions of the country with a higher fraction of air-conditioned buildings.

Indoor sources of ozone are more common than they were several decades ago. Perhaps the most important of these is photocopiers (*Leovic et al., 1996; Destailats et al., 2008*), which were nonexistent in the early 1950s and have become ubiquitous in offices and schools and are even found in many homes. Other indoor sources include so-called “ionic air cleaners” (*Britigan et al., 2006*), ozone generators (*Boeniger, 1995; Weschler, 2000*) and electrostatic precipitators.

*Radon*. Major indoor sources of radon are soil, ground water and construction materials. The 1981 NRC report on indoor pollutants highlighted radon and its potential health risks. In 1984 the news media publicized the case of Stanley Watras, a worker at a nuclear power plant in Pennsylvania who set off radiation alarms on his way *into* work. Subsequently testing showed that the radon level in his home was roughly 650 times higher than typical background levels. In 1988, the Indoor Radon Abatement Act required that EPA list and identify areas in the U.S. where indoor radon levels might be elevated. In 1998 the NRC issued its BEIR VI report “Health Effects of Exposure to Radon” (*National Research Council, 1998*). This report concluded that radon was the second leading cause of lung cancer in the U.S. In 2005, the U.S. Surgeon General issued a Health Advisory noting the serious health threat posed by indoor radon and that lung cancer deaths from radon were almost entirely preventable. Various states have adopted guidelines or regulations regarding radon testing during real estate transactions. As a result of such actions, the number of U.S. homes mitigated for radon increased from 35,000 in 1986 to 80,000 in 2003 (*Gregory and Jalbert, 2004*), suggesting that the general trend in indoor radon concentrations has been downward over the past two decades.

### 6.2. Very volatile organic compounds (VOCs)

*Formaldehyde and acetaldehyde*. Formaldehyde is, or has been, used in the resin of composite-wood products, in acid-cured cabinet and floor finishes, as a preservative in paints and cosmetics,

**Table 2**  
Selected pollutants and trends in their indoor concentrations since the 1950s. Compounds preceded by an asterisk are those for which substantial body burdens have been reported (NHANES, 2005).

Pollutant	Trend	Comment
<i>Inorganic gases</i>		
Carbon monoxide	↓	Less indoor smoking; reduced outdoor concentration
Nitrogen Dioxide	↓	Less indoor smoking; fewer unvented combustion appliances
Nitric Oxide	↓	Less indoor smoking; fewer unvented combustion appliances
Ozone	?	Increased outdoor concentration (with So. CA exception); reduced outdoor-to-indoor transport; increased indoor sources (photocopiers; ionic air cleaners)
Sulfur dioxide	↓	Reduced outdoor concentration
Radon	↓	Increased awareness; mitigation measures
<i>Very volatile organic compounds (VVOC)</i>		
Formaldehyde	↓	Elimination of UFFI; reduced emission from composite-wood products; less indoor smoking
Acetaldehyde	↓?	Reduced emission from some products; increased indoor chemistry?
Acrolein	↓	Less indoor smoking; cooking remains important source
1,3-Butadiene	↓?	Decreased outdoor level; emitted when cooking with some oils
Isoprene	–	Emissions from occupants relatively unchanged
<i>Volatile organic compounds (VOC), aldehydes</i>		
Hexanal	↑?	Increased use of composite-wood, increased indoor chemistry?
Nonanal	↑?	Increased indoor chemistry?
Decanal	↑?	Increased indoor chemistry?
<i>VOC, aliphatics</i>		
<i>n</i> -Alkanes (e.g., <i>n</i> -octane)	–	Continued use of aliphatic solvents
Branched alkanes	–	Continued use of aliphatic solvents
<i>VOC, aromatics</i>		
Benzene	↓↓	Restricted use as solvent (see Fig. 4a and c)
Toluene	↓	Decreased outdoors; reduced use of aromatic solvents
Xylene isomers	↓	Decreased outdoors; reduced use of aromatic solvents
Ethylbenzene	↓	Decreased outdoors; reduced use of aromatic solvents
Trimethylbenzene isomers	↓	Decreased outdoors; reduced use of aromatic solvents
Styrene	↓	Decreased outdoors; reduced use of aromatic solvents
<i>VOC, terpenoids</i>		
Limonene	↑	Increased use of terpenoid solvents and scents
$\alpha$ -Pinene	↑	Increased use of terpenoid solvents and scents
Linalool	↑	Increased use of terpenoid solvents and scents
$\alpha$ -Terpineol	↑	Increased use of terpenoid solvents and scents
<i>VOC, chlorinated</i>		
Dichloromethane	↓↓	Reduced use as solvent
Chloroform	↓	Reduced levels in drinking water, reduced use as solvent
Carbon tetrachloride	↓↓	Reduced use as solvent
1,1,1-Trichloromethane	↓↓	Montreal protocol; reduced use as solvent
Trichloroethylene	↓↓	Reduced use as solvent
Tetrachloroethylene (Perc)	↑,↓	Increased use in “dry-cleaning”, followed by reduced use in “dry-cleaning”
Dichlorobenzene	↓	Reduced use of “mothballs”
*Dichlorophenols	↓	Restricted use as herbicides and pesticides
<i>VOC, fluorinated</i>		
Freon 11	↑,↓	Increased use (air conditioning), followed by Montreal protocol
Freon 12	↑,↓	Increased use (air conditioning), followed by Montreal protocol
Freon 113	↑,↓	Increased use (solvent), followed by Montreal protocol
<i>VOC, other</i>		
*Dimethyl phthalate	↑	Increased use in personal care products and cosmetics
*Diethyl phthalate	↑	Increased use in personal care products and cosmetics
Cyclopentasiloxane (D5)	↑	Increased use in personal care products/antiperspirants
<i>Semivolatile organic compounds (SVOC), biocides/fungicides/preservatives</i>		
*Triclosan	↑	Increased use in disinfecting soaps and cleaning products
Bis(tributyltin)oxide(TBTO)	↑,↓	States began to restrict use in indoor paints in 1988
Butylated hydroxytoluene (BHT)	↑	Increased use as an antioxidant in a variety of products
*Pentachlorophenol (PCP)	↑,↓	Biocide in wood, paints, stains; indoor use restricted in 1984
*Trichlorophenols	↑,↓	Used as pesticides; no longer manufactured
<i>SVOC, combustion byproducts</i>		
ETS	↓	Less indoor smoking; smaller % of population smoking
Dioxins	↓	Reduced outdoor concentrations
Furans	↓	Reduced outdoor concentrations
*PAHs	↓	Less indoor smoking
<i>SVOC, degradation products</i>		
*Bisphenol-A	↑	Increased use of polycarbonate products
<i>SVOC, flame-retardants</i>		
*BDE-47	↑,↓	Increased use in foams and electronics; recent curtailed use
*BDE-99	↑,↓	Increased use in foams and electronics; recent curtailed use
*BDE-209	↑	Increased use in foams and electronics
Tris(chloropropyl)phosphate	↑,↓	Used as flame-retardant for clothing; use restricted

Table 2 (continued)

Pollutant	Trend	Comment
<i>SVOC, heat transfer fluids</i>		
*PCBs	↑,↓	Production peaked in early 1970s; severely restricted in 1978
Polydimethyl siloxanes	↑	Increased use as heat transfer fluid in photocopiers
<i>SVOC, personal care products</i>		
Musk compounds	↑	Increased use of synthetic musks in personal care products
<i>SVOC, pesticides and herbicides</i>		
Aldrin	↑,↓	Agriculture uses canceled 1970, termiticide use canceled 1987
*Chlordane	↑,↓	Growth in mid-1960s; registration withdrawn in 1988
*Chlorpyrifos	↑,↓	Registered for indoor use 1965; registration withdrawn in 2001
*p,p'-DDT	↓	Banned in 1972
*p,p'-DDE	↓	Expected to track trend for DDT
*Dieldrin	↑,↓	Agriculture uses canceled 1970, termiticide use canceled 1987
*Malathione	↓	Restrictions on usage
*Mirex	↑,↓	Introduced in 1962; all pesticide uses canceled in 1978
*Permethrin	↑	Substitute for other pesticides; recent growth
<i>SVOC, plasticizers</i>		
*Dibutyl phthalate	↑	Continued increased use in plasticized products
*Butylbenzyl phthalate	↑	Continued increased use in plasticized products
*Di-2-ethylhexyl phthalate	↑,↓	Recent elimination from toys and other products
Triphenylphosphate (TPP)	↑?	Increased use as plasticizer?
<i>SVOC, stain and water repellents</i>		
*Perfluorinated surfactants	↑,↓	In 2002, 3M stopped producing some perfluorinated compounds
<i>SVOC, nonionic surfactants and coalescing agents</i>		
*4-Nonylphenol	↑	Increased use of nonylphenol ethoxylates (nonionic surfactants)
Texanol® isomers	↑	Increased indoor use of latex paints
<i>SVOC, waxes and polishes</i>		
Fatty acids	?	Longtime ingredient in waxes and polishes
Sesquiterpenes	?	Longtime ingredient in waxes and polishes
<i>Metals and mineral fibers</i>		
Asbestos	↓	Regulations restricting use
*Cadmium	↓?	Less indoor smoking; other sources uncertain
*Lead	↓	Eliminated from gasoline and indoor paint
*Mercury	↓	Eliminated from indoor paint
<i>Others</i>		
Allergens (from dust mites, cats, dogs, etc.)	↑?	More damp buildings; more pets; less dusting and cleaning
Mold/fungi	↑?	More damp buildings; restrictions on moldicides and fungicides
Airborne particles	↓	Less indoor smoking; lower outdoor concentrations

in the coating for permanent press fabrics, in certain insulation materials (urea-formaldehyde foam and fiber-glass) and in finishes that coat some paper products (CPSC, 1997). It is also generated during combustion and the oxidation of compounds with terminal double bonds. Formaldehyde was one of the first pollutants measured indoors (Andersen et al., 1975). In the 1950s, 1960s and early 1970s, composite-wood products, especially plywood, emitted large amounts of formaldehyde. Starting in the mid-1960s, permanent press fabrics became another important indoor source (Kelly et al., 1999). In the mid-1970s, following the energy crisis, the use of urea-formaldehyde foam insulation (UFFI) in homes, especially in difficult-to-access wall cavities, became popular. By the mid-1980s, with limits on allowable formaldehyde emissions from composite-wood products and bans on the use of UFFI in homes and schools, indoor formaldehyde concentrations were decreasing.

In contrast to formaldehyde, there is little long-term data on indoor levels of acetaldehyde. Indoor smoking, a significant source, has decreased. However, acetaldehyde derived from ozone-initiated chemistry appears to have increased.

*Acrolein, 1,3-butadiene and isoprene.* Acrolein is an airborne pollutant that has both acute and chronic effects on human health (U.S. EPA, 2003). Measurements by Seaman et al. (2007) indicate that indoor acrolein levels are much larger than outdoor levels. The dominant indoor sources appear to be cooking, smoking and emissions from wood based materials. Indoor acrolein levels have likely decreased as indoor smoking has decreased. 1,3-Butadiene

and isoprene are other unsaturated VOCs found indoors. Butadiene is present in outdoor air and transported indoors. It is also emitted indoors by rubber products. As its level in outdoor air has gone down, its indoor level has likely followed. Isoprene's indoor concentration is driven by emissions from human occupants, houseplants and wood based materials. Indoor levels of isoprene are not anticipated to have changed greatly over the past five decades.

### 6.3. Volatile organic compounds (VOCs)

*Common indoor VOCs.* Starting in the late 1970s, there have been numerous measurements of volatile organic compounds (VOCs) in indoor air. Reviews of VOC indoor concentrations include Brown et al. (1994); Wolkoff (1995) and Hodgson and Levin (2003). In their 2003 review Hodgson and Levin concluded, "average indoor concentrations of some toxic indoor air contaminants, such as benzene, 1,1,1-trichloroethane and tetrachloroethylene, have decreased" in comparison to the previous decade. The authors suggest that this was partially driven by the 1990 amendments to the Clean Air Act, which resulted in reduced outdoor concentrations of targeted air pollutants. Fig. 4 presents a similar comparison. It is based on measurements made from 1981 to 1984 as part of the U.S. EPA TEAM study (Wallace, 1987) and measurements made from 1999 to 2001 as part of the RIOPA study (Weisel et al., 2005). These studies included measurements of outdoor, indoor and personal concentrations of selected VOCs in several U.S. cities. Fig. 4a

**Table 3**

95th percentile concentrations for more abundant compounds detected in blood and urine samples of U.S. residents, based on sampling from 2001–2002 (NHANES, 2005) unless otherwise noted.

Compound	Blood levels
<b>PCBs</b>	
2,2',3,4,4',5,5'-Heptachlorobiphenyl (PCB 180)	0.61 ng g <sup>-1</sup> serum
2,2',4,4',5,5'-Hexachlorobiphenyl (PCB 153)	0.85 ng g <sup>-1</sup> serum
2,3',4,4',5-Pentachlorobiphenyl (PCB 118)	0.29 ng g <sup>-1</sup> serum
2,4,4',5-Tetrachlorobiphenyl (PCB 74)	0.21 ng g <sup>-1</sup> serum
<b>PBDEs</b> (NHANES for 2003–2004 (Sjodin et al., 2008 Tables 2 and 3); factor of 150 used to convert lipid serum concentrations)	
BDE-47	1.05 ng g <sup>-1</sup> serum
BDE-99	0.28 ng g <sup>-1</sup> serum
BDE-100	0.24 ng g <sup>-1</sup> serum
BDE-153	0.44 ng g <sup>-1</sup> serum
<b>Perfluoroalkyls</b> (NHANES for 2003–2004 (Calafat et al., 2007))	
Perfluorooctanesulfonic acid (PFOS)	55 ng g <sup>-1</sup> serum
Perfluorooctanoic acid (PFOA)	9.8 ng g <sup>-1</sup> serum
Perfluorohexanesulfonate (PFHxS)	8.3 ng g <sup>-1</sup> serum
Perfluorononanoate (PFNA)	3.2 ng g <sup>-1</sup> serum
<b>Organochlorine pesticides/herbicides</b>	
p,p'-DDT	0.18 ng g <sup>-1</sup> serum
p,p'-DDE	15 ng g <sup>-1</sup> serum
Chlordane	0.35 ng g <sup>-1</sup> serum
Mirex	0.41 ng g <sup>-1</sup> serum
Dieldrin	0.15 ng g <sup>-1</sup> serum
<b>Environmental tobacco smoke</b>	
Cotinine (metabolite of nicotine)	2.19 ng g <sup>-1</sup> serum (non-smokers)
<b>Metals</b>	
Cadmium	1.3 ng g <sup>-1</sup> serum
Lead	44 ng g <sup>-1</sup> serum
Mercury	4.6 ng g <sup>-1</sup> serum
Compound	Urine levels
<b>PAHs</b>	
Fluorene	~3.8 μg g <sup>-1</sup> creatinine (Σ metabolites)
Phenanthrene	~1.7 μg g <sup>-1</sup> creatinine (Σ metabolites)
Pyrene	0.243 μg g <sup>-1</sup> creatinine
Benz[a]pyrene	0.184 μg g <sup>-1</sup> creatinine
Naphthalene	34.5 μg g <sup>-1</sup> creatinine (Σ metabolites)
<b>Organochlorine pesticides/herbicides/anti-microbials</b>	
Pentachlorophenol	2.3 μg g <sup>-1</sup> creatinine
2,4,6-Trichlorophenol	11.6 μg g <sup>-1</sup> creatinine
2,4-Dichlorophenol	18 μg g <sup>-1</sup> creatinine
2,5-Dichlorophenol	527 μg g <sup>-1</sup> creatinine
Chlorpyrifos	9.2 μg g <sup>-1</sup> creatinine (based on metabolite 3,5,6-trichloro-2-pyridinol)
cis-Permethrin	3.8 μg g <sup>-1</sup> creatinine (Σ metabolites)
Triclosan	364 μg g <sup>-1</sup> creatinine (NHANES for 2003–2004 (Calafat et al., 2008a))
<b>Organophosphate pesticides</b>	
Dialkyl phosphates	~10–30 μg g <sup>-1</sup> creatinine
Methyl parathion	2.9 μg g <sup>-1</sup> creatinine
<b>Carbamate pesticides</b>	
Propoxur	<1 μg g <sup>-1</sup> creatinine
<b>Phthalates</b>	
Diethyl phthalate	1860 μg g <sup>-1</sup> creatinine
Dibutyl phthalate	81 μg g <sup>-1</sup> creatinine
Butylbenzyl phthalate	90 μg g <sup>-1</sup> creatinine
DEHP	267 μg g <sup>-1</sup> creatinine (Σ metabolites)

**Table 3 (continued)**

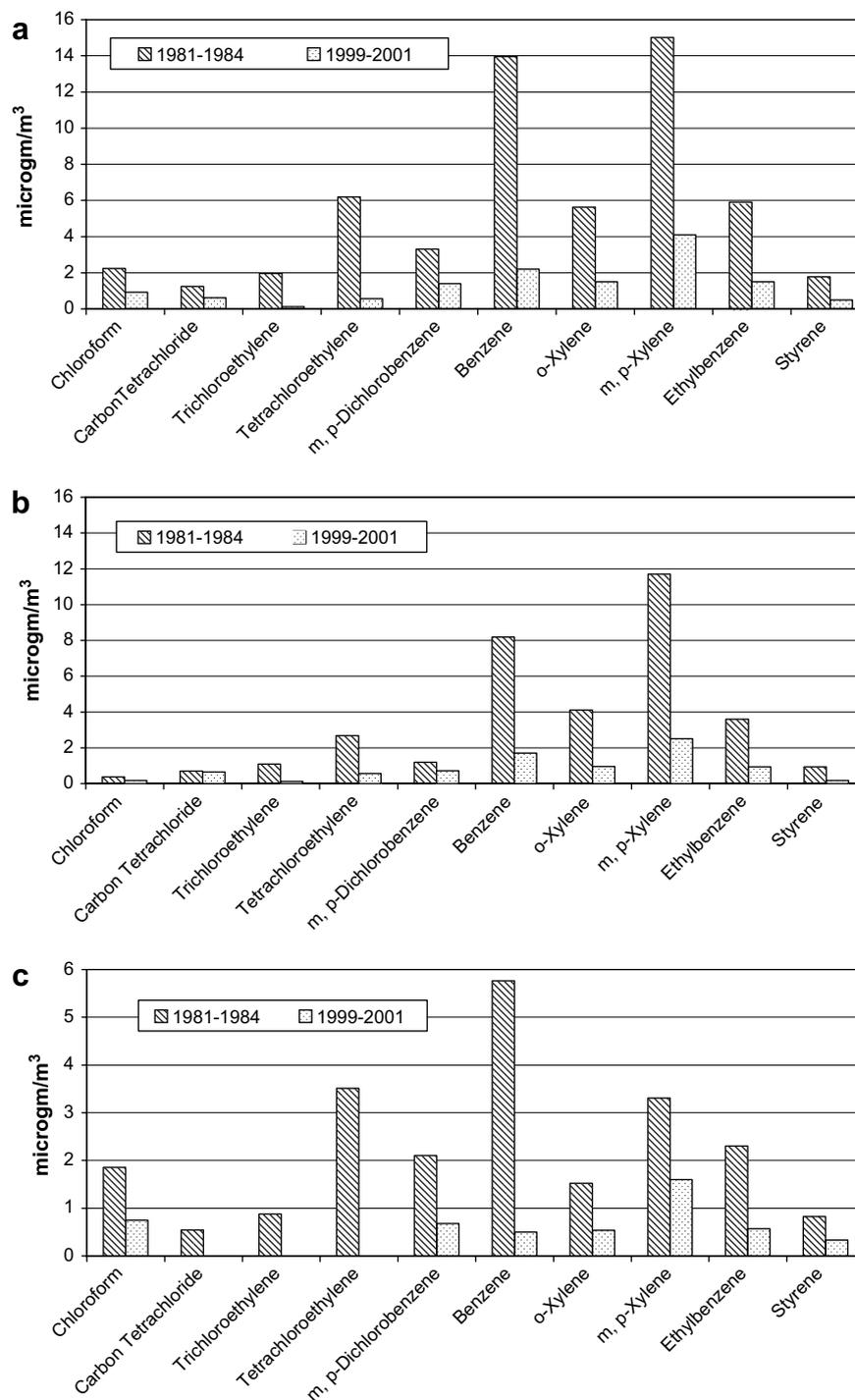
Compound	Urine levels
<b>Others</b>	
Bisphenol-A	11.2 μg g <sup>-1</sup> creatinine (NHANES for 2003–2004 (Calafat et al., 2008b))
4-Nonylphenol	1.4 μg g <sup>-1</sup> creatinine (lower limit; Calafat et al., 2005)
<b>Metals</b>	
Cadmium	0.92 μg g <sup>-1</sup> creatinine
Lead	2.0 μg g <sup>-1</sup> creatinine
Mercury	3.0 μg g <sup>-1</sup> creatinine

compares median indoor concentrations of 10 VOCs measured in both studies; the results from the TEAM study are based on 715 samples collected in Los Angeles, CA and Bayonne/Elizabeth, NJ; the results from the RIOPA study are based on 553 samples collected in Los Angeles, CA, Houston, TX and Elizabeth, NJ. It is apparent from Fig. 4a that the median indoor concentration of each of the displayed compounds decreased significantly from the 1981–1984 period to the 1999–2000 period. The largest relative decreases are seen for trichloroethylene (94%), tetrachloroethylene (91%) and benzene (84%). Fig. 4b is analogous to Fig. 4a, but displays the outdoor concentrations. The outdoor concentrations for most of the displayed compounds have also decreased significantly, indicating that decreasing outdoor concentrations are partially responsible for the observed decrease in indoor concentrations.

For a pollutant that is not significantly removed by surfaces or indoor chemistry, the difference between its indoor and outdoor concentrations is its indoor emission rate (mass per unit time) normalized by the ventilation flow rate (volume per unit time). Fig. 4c compares normalized indoor emission rates, derived in this fashion, for the 10 VOCs displayed in Fig. 4a and b. It is apparent from Fig. 4c that indoor emission rates of carbon tetrachloride, trichloroethylene and tetrachloroethylene dropped from meaningful to negligible levels over the almost two decades that separate the TEAM study and the RIOPA study. The decrease in the normalized emission rate of benzene was almost as great (~91%). The normalized indoor emission rates of each of the other VOCs in the plot decreased by at least 50%, despite the fact that ventilation rates had likely decreased somewhat over the intervening period (which would have resulted in larger normalized emission rates, if all else had remained the same).

The TEAM study included homes with smokers whereas the RIOPA study excluded homes with smokers. However, this is insufficient to account for the magnitude of the decrease in aromatic VOCs between the two studies. The RIOPA study included homes from Houston, whereas the TEAM study did not. However, the Houston results were not substantially different from the Los Angeles or Elizabeth results. Basically, the changes in indoor concentrations and indoor emission rates are consistent with changes that we know occurred in the period between 1981–1984 and 1999–2000. These include a shift away from chlorinated solvents and, to a lesser extent, aromatic solvents in products intended for indoor use; restrictions on the production of trichloroethane as a consequence of the Montreal Protocol; restrictions on the use of benzene (see below); less indoor smoking, and reductions in the concentrations of aromatic compounds in outdoor air due to 1990 amendments to the Clean Air Act.

**Benzene.** Benzene warrants special comment. Common in products in the 1950s and 1960s, its indoor use was already decreasing in the 1970s. In 1978, the U.S. Consumer Product Safety Commission (CPSC) proposed a ban of benzene in consumer products. In 1980, CPSC proposed that manufacturers identify consumer products to which benzene had been intentionally added. Two years later, IARC (1982a,b) classified benzene as a known human carcinogen. Eventually CPSC withdrew its proposed formal ban on benzene, since extensive surveys indicated



**Fig. 4.** (a). Median indoor concentrations of VOCs measured in both the 1981–1984 TEAM study and the 1999–2001 RIOPA study. (b). Median outdoor concentrations of VOCs measured in both the 1981–1984 TEAM study and the 1999–2001 RIOPA study. (c). Median indoor emission rates, normalized by ventilation flow rates, of VOCs measured in both the 1981–1984 TEAM study and the 1999–2001 RIOPA study. TEAM study:  $n = 715$ ; Wallace, 1987; RIOPA study:  $n = 553$ ; Weisel et al., 2005.

that benzene was no longer being intentionally added to consumer products. Today, almost half the U.S. population's exposure to benzene comes from inhaling air that contains tobacco smoke (National Toxicology Program, 2005).

*Chlorofluorocarbons, low molecular weight phthalates and siloxanes.* The use of chlorofluorocarbon refrigerants and solvents (sold by Dupont as "Freons") increased from the early 1950s into the mid-1980s. In the years following the 1987 signing of the Montreal Protocol, their indoor concentration decreased sharply. In 1995 chlorofluorocarbon production in the U.S. ceased.

The use of dimethyl- and diethyl phthalates in personal care products and cosmetics, as well as the use of cyclopentasiloxane (D5) in antiperspirants, has increased during the past several decades.

#### 6.4. Semivolatile organic compounds (SVOCs)

There are fundamental differences between VOC and SVOC emissions. For volatile organic compounds found within the matrix of a material at the time of manufacture (e.g., solvents, unreacted

monomers, byproducts), emissions tend to occur independent of their external environment and decrease over the life of the material. Such emissions often deplete the reservoir of VOCs present within a material during the initial weeks or months that the material is present in an indoor setting. In contrast, semivolatile organic compounds found within the matrix of a material (e.g., plasticizers, flame-retardants, preservatives) tend to be emitted at rates that depend on external factors such as partitioning into the gas phase, the convective mass transfer coefficient and sorption onto indoor surfaces (Xu and Little, 2006). Furthermore, SVOC emissions do not meaningfully deplete the reservoir of SVOC within the material, since the amount of SVOC additive emitted from the material in an hour or a day is only a small fraction of the total amount that is typically present in the material. Plasticized vinyl floors or flame-retarded mattresses continue to emit meaningful amounts of plasticizers or flame-retardants over their entire lifetime. Similarly, when low volatility SVOCs are applied or emitted in indoor environments (e.g., pesticides, stain repellants, dioxins), they will be depleted at a slow rate and will continue to impact their indoor setting for years after their initial release (Weschler and Nazaroff, 2008).

It is convenient to review semivolatile organic compounds in terms of their source or use categories. Not all of the categories listed in Table 2 will be explicitly discussed. Instead the focus is on those sources that have had the largest impact on indoor environments.

*Biocides, fungicides and preservatives.* Until the mid-1980s, pentachlorophenol (PCP) was used as a wood preservative for foundation and structural lumber in residential and commercial buildings. It was also the active biocide in a large number of products used indoors, including wood preservatives, paints, wood stains, and sealers. Restrictions on its indoor use were adopted by EPA in 1984 and became effective after failed industry lawsuits in 1986. Nonetheless, due to its extensive indoor uses, PCP was widely distributed in homes built prior to 1984. During the early 1980s, indoor concentrations in the range of 0.1–10  $\mu\text{g m}^{-3}$  were common in treated wood structures (Levin and Hahn, 1986). Given its moderate vapor pressure, it has not persisted in indoor environments as long as less volatile chlorinated species (e.g., DDT or higher molecular weight PCB congeners). However, treated wood can constitute a very large and persistent reservoir. Additionally, some of the salvaged wood that is used to build or remodel houses today may have been pressure treated with PCP when it was first produced. Rudel et al. (2003) found PCP in the air of 58% of 120 Cape Cod homes sampled from 1999 to 2001; its median concentration was 1.8  $\text{ng m}^{-3}$ . Between 2000 and 2001, Wilson et al. (2007) measured very similar levels in the air of 251 homes and 42 daycare facilities in Ohio and North Carolina. PCP is still detected in the urine of almost all U.S. residents (NHANES, 2005), a legacy of its once ubiquitous use.

*SVOCs derived from combustion.* This category includes PAHs, dioxins and furans. Among the PAHs, benz[a]pyrene (B[a]P) is a known carcinogen and has received substantial attention. Major indoor sources include outdoor air, cooking, smoking and unvented combustion appliances. Given the reduced percentage of U.S. households with smokers (Fig. 2), the concentration of B[a]P has likely decreased.

*Flame-retardants.* In the U.S., fires that originate in homes or offices have declined over the past several decades, partially due to policies that require flame-retardants in various materials and consumer products (Birnbaum and Staskal, 2004). However, during this same period the concentration of flame-retardants has increased in indoor environments. Certain flame-retardants that were once common are no longer in use. These include polybrominated biphenyls (PBBs) that were discontinued in the early 1970s after an incident in which PBBs were mistakenly mixed with

animal feed (Dunckel, 1975), and tris(2,3-dibromopropyl)phosphate (tris-BP) that was found in the urine of children whose sleepware had been treated with this agent (Blum et al., 1978). Polybrominated dipheynyl ethers (PBDEs) are flame-retardants that have been in widespread use for only the past two decades. They are used in foam cushioning and mattresses, electronic devices such as televisions and computers, as well as a variety of other household products. Hites (2004) has published a critical review that examines the accumulation of PBDEs in different environments, as well as in people. He concludes: "By now it is clear that PBDEs are ubiquitous environmental pollutants and that their concentrations in most environmental compartments are exponentially increasing with doubling times of about 4–6 yr." As their levels increase in indoor environments, their levels increase in the occupants of those environments (Section 6.6).

*Heat transfer fluids.* Although this review classifies polychlorinated biphenyls (PCBs) as heat transfer fluids (used in oil-filled transformers, capacitors and fluorescent lamp ballasts), they had numerous other indoor uses including stabilizers for PVC wire-insulation, flame-retardants (see above), pesticide extenders, additives (in sealants, adhesives, paints, and floor finishes) and in carbonless copy paper. Their production peaked in the late 1960s/early 1970s. In 1971, Monsanto, the major U.S. producer, voluntarily cut back production. In 1978, their use (except for totally enclosed situations) was banned. Commercial PCB mixtures often contained a small fraction of polychlorinated dibenzofurans (furans) or polychlorinated dibenzodioxins (dioxins), which were unintended contaminants of the production process. These co-occurring furans and dioxins are worth noting since they tend to be much more toxic than PCBs (Agency for Toxic Substances and Disease Registry, 2000). During the 1970s and 1980s, indoor PCB levels were typically in the 100–500  $\text{ng m}^{-3}$  range (e.g., MacLeod, 1981). By in the late 1990s, indoor levels were in the 1–10  $\text{ng m}^{-3}$  range (Menichini et al., 2007 and references therein). Rudel et al. (2003) found PCBs in the air of more than 30% of 120 Cape Cod homes sampled from 1999 to 2001. In a follow-up study of two highly contaminated homes, PCB containing wood floor finish that had been used in the 1950s and 1960s was identified as a strong, continuing source of PCBs (Rudel et al., 2008). Transformers, capacitors and fluorescent lamp ballasts produced before 1978 are among other important indoor source of PCBs. Hence, although their indoor levels have been declining, PCBs are still present in indoor air and dust, and are expected to remain in contaminated indoor environments for years to come.

*Personal care products.* The constituents of personal care products have received remarkably little attention as indoor pollutants (Daughton and Ternes, 1999). These have evolved over the past several decades to include many complex semivolatile organic compounds. Prime examples are synthetic aromatic nitro-musks and polycyclic musks that have largely replaced natural musk fragrances. Such compounds have been widely used as scented agents in cosmetics, perfumes and cleaning products and have been reported at tenths of  $\mu\text{g m}^{-3}$  in indoor air and  $\text{mg kg}^{-1}$  levels in indoor dust (Fromme et al., 2004). Recently, production of nitro-musks has decreased due to concerns about their potential toxicity (OSPAR, 2004).

*Pesticides and herbicides.* Until it was banned in 1972, DDT was used chiefly as an agricultural pesticide. Nonetheless, DDT (and its degradation product DDE) can still be measured in indoor air and dust, as demonstrated by samples collected in 120 Massachusetts' homes (Rudel et al., 2003). Its presence indoors may be due to outdoor-to-indoor air transport or tracked-in dust. It was also occasionally applied indoors as a pesticide of convenience. Although indoor DDT levels are likely lower than they were in the 1950s and 1960s, its presence in Massachusetts' homes sampled almost 30 years after it was banned indicates its persistence in indoor environments.

Chlordane is a pesticide that was first registered in 1948; it had numerous applications, both outdoors and indoors. Its history has been repeated by a number of pesticides that have succeeded it. Chlordane production continued to increase through the 1960s, and it became the pesticide of choice for termite control. Starting in 1974, U.S. government agencies placed restrictions on its indoor and outdoor use. By 1983, the only permissible use was as a termiticide. In this year both National Public Radio and 60 Minutes aired shows on chlordane and its presence indoors after sub-slab application as a termiticide. Such publicity increased public awareness of their exposure to chlordane, and in 1988 all sales and uses of chlordane in the U.S. were halted. Despite this, chlordane has persisted in the environment. Approximately 12 years after all uses of chlordane were halted, Rudel et al. (2003) measured a median air concentration of  $0.3 \text{ ng m}^{-3}$  for the sum of chlordane isomers in 120 Cape Cod, MA homes. During this same period, the RIOPA study measured geometric mean concentrations of  $1.3 \text{ ng m}^{-3}$  in NJ homes,  $2.0 \text{ ng m}^{-3}$  in CA homes and  $4.2 \text{ ng m}^{-3}$  in TX homes (note that the levels increase for regions more likely to have termite problems). Chlordane is a “legacy indoor pollutant”; once present indoors, its primary removal mechanism is via ventilation. This is a slow process for a contaminant when the airborne concentrations are  $0.1\text{--}1 \text{ ng m}^{-3}$  and milligram amounts were originally applied (Weschler and Nazaroff, 2008). Trends in chlordane’s body burden are discussed in Section 6.6.

The indoor use of chlorpyrifos, an organophosphate pesticide, follows a history similar to that of chlordane. It was extensively used in U.S. buildings until 2002, and was routinely applied in milligram amounts using a crack and crevice approach. It has sufficient volatility to migrate from the original point of application to all other indoor surfaces. Chlorpyrifos can also be considered a “legacy pollutant”. Its indoor levels are decreasing, but at a slow rate.

In the United States, mirex was used in pesticide formulations starting in 1962. All U.S. uses of mirex as a pesticide were banned in 1978. Mirex was also used in as a flame-retardant in plastics, rubber, paint and electronics.

The use of pyrethroids has grown considerably over the past decade as the use of other pesticides has been eliminated or curtailed. Permethrin is an example of a pyrethroid pesticide now commonly found indoors but absent several decades ago.

**Plasticizers.** Since the 1950s, increasing amounts of phthalates have been used as plasticizers, especially for flexible polyvinyl chloride (PVC). PVC flooring (Clausen et al., 2004), wall covering (Uhde et al., 2001) and electrical cable insulation are each meaningful emitters of phthalates, and can contain these additives at levels of 30–40% (by wt). Until recently, di-2-ethylhexyl phthalate (DEHP) was the primary phthalate used in flexible PVC. It accounted for more than half of all phthalate ester plasticizers produced and was the dominant SVOC in indoor environments (Bornehag et al., 2005a; Wensing et al., 2005). Recently, the use of DEHP, as well as butyl benzyl phthalate, has been reduced as a consequence of concerns regarding potential health effects (Oie et al., 1997; Adibi et al., 2003; Bornehag et al., 2004; Swan et al., 2005; Hauser et al., 2006). However, given its low vapor pressure and apparently slow indoor degradation rate, DEHP is likely another indoor “legacy pollutant”.

**Stain repellants and water repellants.** Perfluorinated surfactants are an important class of stain and water repellants that have been used extensively on carpets, drapery and upholstery fabrics found indoors. The best known of these is Scotchgard®, a 3 M product that was introduced in 1956 and voluntarily phased out, together with other perfluorinated compounds, in 2002. Perfluorinated surfactants are robust, low vapor pressure compounds anticipated to persist for years after their introduction into an indoor environment. Shoeib et al. (2004, 2005) have measured perfluorinated

sulfonamides in indoor air and dust samples. Given the production history of these compounds, one can infer that their indoor levels increased from the 1960s through the 1990s, and then began to decline after 2002. Such a trend has been reported for the body burdens of selected perfluorinated compounds (Section 6.6).

**Surfactants (nonionic) and coalescing agents.** Nonionic surfactants have displaced anionic surfactants in detergents and cleaning products. These compounds and their degradation products (e.g., 4-nonyl phenol) are now commonly found in indoor environments. For example, Rudel et al. (2003) have detected nonylphenol, nonylphenol ethoxylates and octylphenol ethoxylates in the air and dust of Cape Cod homes. 4-Nonylphenol was detected in the air of all 120 homes sampled, at a median concentration of  $110 \text{ ng m}^{-3}$ .

Indoor Texanol® levels have increased with increasing use of latex paints. In measurements made over the last two decades, indoor airborne Texanol® concentrations have typically been in the range of 0.1–5 ppb or  $0.8\text{--}45 \text{ } \mu\text{g m}^{-3}$  (Corsi and Lin, in press).

**Semivolatile terpene oxidation products.** As noted in Section 3.1, indoor levels of oxidation products derived from terpene and terpene alcohol precursors appear to have increased over the past 50 years. These products include semivolatile compounds (e.g., Glasius et al., 2000) that partition between the gas phase, the surface of airborne particles, and other indoor surfaces. Partitioning to airborne particles results in increased levels of secondary organic aerosols (SOA). The nature of the SVOCs derived from such oxidation reactions varies with the chemical structure of the precursor.

#### 6.5. Metals, mineral fibers and particles

**Lead.** The major sources of lead in the indoor environment have been paint and outdoor-to-indoor transport of emissions from motor vehicles burning leaded gasoline. In 1953, industry standards reduced lead levels in paint to 1%. In 1973, the U.S. EPA issued their first standards to reduce lead in gasoline. As a result of the subsequent phase-out of lead in gasoline, its concentration in outdoor air has decreased almost 98% since 1980 (Chemical & Engineering News, 2008). In 1978, the Lead Based Paint Poisoning Prevention Act banned paint with more than 0.06% lead from residential use. These measures have had a dramatic effect – the percentage of children aged 1–5 with blood-lead levels greater than  $10 \text{ } \mu\text{g dL}^{-1}$  has declined from 88% (1976–1980) to 8.6% (1988–1991) to 4.4% (1991–1994) to 1.6% (1999–2002) (CDC, 2005).

**Mercury.** Historically, the major sources of mercury in indoor environments have included paints, preservatives, thermometers, certain types of switches and lamps, and outdoor-to-indoor transport. A major source of mercury in outdoor air remains combustion of coal that contains mercury. In 1990, the U.S. EPA banned mercury in interior latex paint, and in 1992 it banned mercury in all interior paints. Mercury’s presence as a preservative in other products intended for indoor use has also been severely restricted. Indoor levels of mercury have likely declined over the past 15 years.

**Cadmium.** Cadmium from smelting activities can be present in outdoor air and transported indoors. However, for most of the U.S. population, cigarette smoking and exposure to environmental tobacco smoke is the major source of cadmium. The CDC (2005) found that about 5% of the U.S. population aged 20 and older had urinary cadmium levels high enough to be of concern with respect to kidney injury. Given the decline in indoor smoking, indoor levels of cadmium have likely declined over the past several decades.

**Asbestos.** Asbestos fibers are flexible, strong, durable and flameproof. These properties led to its use in many products, including thermal and acoustic insulation, flooring, textiles, felts, and coating materials. In 1950, Underwriters Laboratories approved its spray application to steel structural supports in multi-storey buildings to prevent their deformation during fires. This use

resulted in substantial releases of asbestos fibers into indoor environments. In 1973 the U.S. EPA banned asbestos spraying for insulation and fireproofing of structural materials. In 1978 the EPA broadened the ban to include all spraying, with the exception of situations where it was encapsulated in a binder that was not friable after drying. Between 1950 and 1978 it is estimated that half a million tons of asbestos was sprayed on surfaces in the U.S. (National Research Council, 1981, p. 113).

Vinyl asbestos floor tiles, popular from the late 1940s to the early 1970s, were another significant indoor use of asbestos. In the early 1990s they were estimated to be present in 42% of U.S. buildings (OSHA, 1994). Fortunately, these tiles release few asbestos fibers unless they are damaged. Regardless, asbestos floor tiles are no longer used in buildings, and the fraction of buildings that contain them has been steadily decreasing over the past 20 years.

In 1989 the U.S. EPA issued a final rule banning most asbestos containing products. This rule was challenged in court and overturned in 1991, but the court ruling retained the ban on asbestos in flooring felt, rollboard, and corrugated, commercial or specialty paper, as well as “new uses” of asbestos (U.S. EPA, 2007).

*Airborne particles.* There are multiple sources of indoor airborne particles (Abt et al., 2000), including outdoor air, indoor combustion (especially smoking and cooking), abrasion, vacuuming, re-suspension, laser printers (He et al., 2007) and ozone-initiated chemistry (see Section 3.1). The concentration of particles in outdoor air has declined due to regulations on incinerators, industrial emissions and motor vehicle exhaust. The percent of adult smokers has also declined (see Fig. 2). Although emissions of indoor particles from laser printers and indoor chemistry are inferred to have increased, these increases are anticipated to have been overshadowed by reductions in particles derived from indoor smoking and outdoor-to-indoor transport. That is, on average, indoor particle levels are probably lower today than in the 1950s.

## 6.6. Body burdens

Table 3 lists the 95th percentile levels of the more abundant compounds detected in blood and urine samples collected from over 2000 U.S. residents in 2001–2002 (NHANES, 2005, unless otherwise noted). These blood and urine concentrations provide an indication of the net amount of an environmental pollutant that enters the body from all pathways. While ingestion of food containing these pollutants is often the dominant route of exposure (e.g., PCBs, DDT, mercury), inhalation, ingestion of dust, and dermal absorption can also be important contributing pathways. Almost all of the compounds in Table 3 have been identified in indoor air and dust. Indeed, most of materials that contain PBDE flame-retardants and phthalate ester plasticizers are intended for indoor use. Wilford et al.'s (2005) analysis indicates that ingestion of indoor dust is a major exposure pathway for PBDEs, especially for children. Fromme et al. (2007) used duplicate meal samples, measured indoor air/dust concentrations and back-calculated intake from excreted phthalate metabolites, to show that non-food pathways are important exposure routes for dibutyl and diisobutyl phthalates. Exposures to chlordane and chlorpyrifos in indoor air and dust have been inferred to contribute to their overall body burdens, especially in homes that have been treated with these pesticides for termites (Dearth and Hites, 1991; Morgan et al., 2005; NHANES, 2005). A large study of preschool children's exposure to pentachlorophenol concluded that inhalation was their dominant route of exposure (Wilson et al., 2007). The airborne concentrations of 4-nonylphenol measured by Rudel et al. (2003) suggest that inhalation also contributes meaningfully to its presence in humans. Basically, if a pollutant exists at elevated concentrations in indoor air and dust, humans will inhale, ingest (dust and foods exposed to

indoor air prior to consumption), and dermally absorb it, contributing to the compound's resulting body burden.

For some of the chemicals listed in Table 3, the body burdens appear to be relatively steady or increasing. In a 1991 article that examined chlordane accumulation in humans, Dearth & Hites wrote “there has been no measurable decline in these concentrations levels (adipose tissue) even after 10 years of regulation.” NHANES data for chlordane metabolites in blood are available for two different time periods: 1999–2000 ( $n = 1661$ ) and 2001–2002 ( $n = 2249$ ). In the two-year period between surveys, there was no decline in the blood levels of the chemicals that constituted technical grade chlordane and its metabolites (NHANES, 2005, pp. 331–337). Measurements of PBDE in archived U.S. human serum samples collected from 1985 to 2002 indicate that the serum concentrations of PBDE congeners have been increasing (Sjodin et al., 2004).

For other chemicals listed in Table 3, the body burdens appear to have peaked and are presently declining. In the same serum samples just discussed, collected from 1985 to 2002, blood levels of a PBB and a PCB congener decreased (Sjodin et al., 2004), consistent with restrictions on their use starting in the 1970s. In Section 4.1 we noted that between 1988–1991 and 1999–2002, blood cotinine levels for nonsmoking adults decreased about 75% (NHANES, 2005). This is indicative of reduced exposure to nicotine from environmental tobacco smoke. In the case of fluorinated surfactants, earlier biomonitoring showed that concentrations in human serum had been increasing (Houde et al., 2006). In 2000 the major world producer of perfluoroalkyl compounds, the 3M Company, began a phase-out of certain fluorinated precursors. Calafat et al. (2007) have recently reported that serum concentrations of PFOS, PFOA and PFHxS are lower in samples collected from U.S. residents in 2003–2004 than in samples from 1999–2000. These results are supported by comparisons of perfluoroalkyl levels in serum samples of American Red Cross Blood donors collected in 2000–2001 and 2006, which indicate that the blood levels of certain perfluoroalkyls are declining (Olsen et al., 2008).

A striking feature of the compounds in Table 3 is that many have been produced in meaningful amounts only within the last several decades. These include compounds currently found in tenths to tens of nanograms per gram of blood serum (e.g., PBDEs, perfluoroalkyls, chlordane, mirex and dieldrin) and compounds or their metabolites found in microgram to milligram per gram of urine creatinine (e.g., chlorpyrifos, cis-permethrin, triclosan, methyl parathion, various phthalate esters, and 4-nonyl phenol). Humans were not exposed to these compounds two or three generations ago. We know why we are using them: to make plastics perform better, to reduce the risk of fire, to kill pests, to minimize mold growth, to help paint spread, to improve the scent of cleaning products. However, at present we cannot be confident that long-term exposure to these compounds, either individually or in a mixture, is benign.

## 7. Conclusions

The health risks from indoor pollutants in 2008 differ from those in the 1950s. Indoor exposures to a number of “known” carcinogens (e.g., benzene, formaldehyde, asbestos, environmental tobacco smoke and radon) and “reasonably anticipated” carcinogens (e.g., chloroform, trichloroethylene, carbon tetrachloride and naphthalene) have decreased. Indoor exposures to other recognized toxicants such as carbon monoxide, sulfur dioxide, nitrogen dioxides, lead and mercury have also declined. Conversely, indoor exposures to suspected endocrine disruptors have markedly increased; these include certain phthalate ester plasticizers, certain brominated flame-retardants, bisphenol-A and nonylphenol. Pesticide exposures have changed from compounds that were

found to have health risks (e.g., chlordane, chlorpyrifos and mirex) to compounds that are thought to be relatively safe (pyrethroids).

It is apparent that changes in indoor emissions of volatile pollutants (e.g., inorganic gases, VVOCs, VOCs) have impacted indoor environments faster than changes in emissions of less volatile pollutants (e.g., SVOCs, heavy metals, fibers). For example, restrictions on the use of chlorinated solvents influenced indoor environments over a shorter time interval than did restrictions on the use of chlorinated pesticides. While VOC emissions from a product tend to decrease sharply during the first weeks or months of a product's life, SVOC emissions tend to continue throughout the life of a product (Section 6.4). Additionally, SVOCs sorb to other indoor surfaces after they are released to the air, and, in the case of low volatility SVOCs (e.g., DEHP, BDE209, DDT), these sorbed SVOCs continue to desorb long after the host material is removed. In a related fashion, if there is a large residue of a heavy metal or asbestos indoors, it will persist until that residue is deliberately removed.

Given the importance of this subject, it is striking how little directly measured, year-to-year data exist on the kind and concentration of indoor pollutants. It would be exceptionally valuable to establish monitoring networks, in both the U.S. and elsewhere, that provided cross-sectional and longitudinal information about the state of pollutants in representative buildings. Operated in parallel to networks that presently monitor pollutants outdoors and in body fluids, such networks would vastly enhance our knowledge and understanding of the chemicals that we daily inhale, ingest and absorb.

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