

# CHAPTER 3. CHARACTERIZING EXPOSURES TO ATMOSPHERIC CARCINOGENS

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Organic emissions are predominantly non-polar, lipid-soluble substances with oxygen-to-carbon ratios near zero; atmospheric chemistry transforms these and inorganic emissions into more oxidized, more water-soluble compounds ([Chung and Seinfeld, 2002](#); [Lioussse et al., 1996](#)). Atmospheric pollutants (particles and gases) are either emitted directly from sources (primary) or formed in the atmosphere (secondary) through chemical reactions involving precursor emissions and often sunlight ([Seinfeld and Pandis, 1998](#)). For example, photochemical oxidation forms aldehydes, ketones, acids, organic peroxides, and epoxides from anthropogenic and biogenic aromatics, alkenes, and alkanes ([Paulot et al., 2009](#); [Atkinson, 2000](#); [Finlayson-Pitts and Pitts, 2000](#)). In addition, oxygenated and nitro-polycyclic aromatic hydrocarbons (PAHs) are formed from atmospheric processing of combustion-generated PAHs ([Fan et al., 1995](#); [Finlayson-Pitts and Pitts, 2000](#)). Similarly, oxides of nitrogen ( $\text{NO}_x$ ; gas-phase species) react to form nitric acid (gas phase) and ammonium nitrate (particle phase); sulfur dioxide is oxidized to acidic or ammonium sulfate (particle phase). Some reactions result in the formation of new particles or add more oxidized material to pre-existing (primary) particles ([Ervens et al., 2011](#); [Seinfeld and Pankow, 2003](#); [Weber et al.,](#)

[1999](#)). For example, freshly emitted combustion particles are dominated by solid materials (black carbon, metals) and viscous organic liquids (e.g. PAHs, alkanes). A substantial portion of this primary particulate organic matter is believed to evaporate as it is diluted with cleaner air ([Robinson et al., 2007](#)). Subsequently, sulfates, nitrates, and polar organics formed through gas-phase photochemistry condense onto these primary seed particles ([Seinfeld and Pankow, 2003](#)) or form through aqueous chemistry in clouds and wet particles and remain in these particles after water evaporation ([Ervens et al., 2011](#)). As a result, atmospheric submicron particles are dominated by secondary oxidized species, except in close proximity to sources ([Zhang et al., 2007](#); [EPA, 2009](#)).

The degree of atmospheric *ageing* (degree of oxidation) of the air pollution mix has a substantial impact on the composition, properties, fate, and effects of the air pollution mixture. [Table 3.1](#) lists organic compounds commonly measured in atmospheric particles. While it is widely known that organic particulate matter (PM) includes alkanes, aromatic hydrocarbons, and dicarboxylic acids, a wide variety of types of organic compounds are present in atmospheric particles, including polyols, sugars, esters, amines, aldehydes, and even nitrocatechols ([Saxena and](#)

[Hildemann, 1996](#); [Claeys et al., 2012](#)). Typically only about 20% of organic PM is identified at the molecular level ([Rogge et al., 1993](#)). Most unidentified organic PM is made up of large, multi-functional compounds, including humic-like materials and oligomers ([Lin et al., 2010](#); [Turpin et al., 2000](#); [Rogge et al., 1993](#)). Water solubility affects the fate of some inhaled pollutants. Water-soluble gases (e.g. hydrogen peroxide) diffuse readily to the wetted surfaces in the mouth, nose, and upper airways and are efficiently removed ([Wexler and Sarangapani, 1998](#)). In contrast, gas-phase PAHs and ozone are transported into the air-exchange regions of the lung ([Hatch et al., 1994](#)).

Particles also penetrate into the air-exchange regions of the lung, delivering liquid- and solid-phase, water-soluble, and lipid-soluble compounds. Fresh combustion-generated particles comprising solid graphitic carbon, metals, and viscous organic liquids presumably retain their particle form after deposition, although their organics will absorb into lipid-like materials given the opportunity. When secondary particles comprising concentrated aqueous solutions deposit in the lung, they dissolve into the lung surfactant, delivering dissolved chemicals. These (largely) secondary accumulation-mode particles sometimes contain an ultrafine primary core, facilitating the deposition of ultrafine solid particles. Hygroscopic (secondary) particles can also transport water-soluble vapour (e.g. hydrogen peroxide, organic peroxides) into the lower lung ([Wexler and Sarangapani, 1998](#); [Morio et al., 2001](#)).

Urban air pollution is a combination of regionally generated pollutants that are substantially transformed through atmospheric processing, predominantly so-called secondary pollutants, and locally generated pollutants that are more likely to be dominated by primary emissions. For example, [Lee et al. \(2004\)](#) reported that fine PM (PM < 2.5 µm in diameter [PM<sub>2.5</sub>]) in urban-industrial New Jersey is dominated by secondary

**Table 3.1 Organic compounds commonly measured in atmospheric particles**

Compound class
<i>n</i> -Alkanes (C23–C34)
<i>n</i> -Alkanoic acids (C9–C32)
<i>n</i> -Alkenoic acids (C17–C18)
<i>n</i> -Alkanols (C25–C32)
<i>n</i> -Alkanals (C9, C26–C32)
Aliphatic dicarboxylic acids (C2–C9)
Ketocarboxylic acids (C2–C5)
Diterpenoid acids (C20)
Other multifunctional aliphatic acids (C3–C6)
Aromatic polycarboxylic acids (C8–C10)
Polycyclic aromatic hydrocarbons (C16–C24)
Polycyclic aromatic ketones and quinones (C17–C19)
Steroids (Cholesterol, C27)
Nitrogen-containing compounds (C6–C10)
Carbonyls (C2)
Phenol and substituted phenols (C6–C7)
Guaiacol and substituted guaiacols (C7–C11)
Syringol and substituted syringols (C8–C12)
Mono-, sesqui-, and triterpenoids (C10–C12, C30)
Sugars (levoglucosan, C6)

Compiled from [Rogge et al. \(1993\)](#) and [Schauer et al. \(1996\)](#).

formation and regional transport. They estimated that in the largest cities in New Jersey, no more than 25–30% of PM<sub>2.5</sub> is emitted or formed locally. In contrast, they reported that about 75% of PAHs in urban-industrial New Jersey are emitted locally. Concentrations of secondary species (e.g. sulfate, ozone, organic acids) are quite homogeneous across cities and large regions (states/nations) in places where regional transport is important (e.g. the eastern USA; [Chuersuwan et al., 2000](#)). In contrast, primary pollutants (e.g. carbon monoxide [CO], PAHs, black carbon) are elevated above the regional signal in urban areas, and very close to sources the concentrations of primary pollutants, such as road traffic, can be substantially elevated above the urban mix. Elevated CO concentrations in street canyons are well documented. Also, several studies have found elevated mass concentrations of black carbon, NO<sub>2</sub>, and PAHs and number

concentrations of ultrafine particles within 100 or 200 m of major roadways ([Roorda-Knappe et al., 1998](#); [Zhu et al., 2002](#); [Polidori et al., 2010](#)).

People are exposed to pollutants generated indoors at home or work, outdoors, and in other microenvironments (e.g. in transit). Pollution generated in these diverse environments is produced from a varied mix of sources and therefore will have a different composition and likely different effects ([Long et al., 2001](#)). Many pollutants are generated through personal activities. In this case, exposures are generally elevated above ambient (indoor or outdoor) concentrations because the individual is in close proximity to the source, such as when cooking or smoking tobacco products. Globally, the highest exposures to combustion-generated pollutants are found indoors in rural areas of developing countries and are the result of emissions from the combustion of unprocessed solid fuels from cooking ([Smith, 2002](#)).

Because people spend a large majority of their time indoors (for residents of the USA, 87% of their time; [Klepeis et al., 2001](#)), exposure to outdoor-generated (atmospheric) pollutants (as well as indoor-generated pollutants) predominantly occurs in indoor environments. For some pollutants, indoor sources and personal activities drive exposures, whereas other pollutant exposures are dictated by ambient outdoor sources. In homes without smokers in cities in the USA, particle-phase PAH concentrations found indoors are predominantly from outdoor sources ([Naumova et al., 2002](#)), and outdoor-generated PM<sub>2.5</sub> is the largest contributor to indoor concentrations of PM<sub>2.5</sub> ([Ozkaynak et al., 1996](#); [Meng et al., 2005](#)). Ozone, lead, manganese, cadmium, methyl tertiary butyl ether, carbon tetrachloride, trichloroethylene, and sulfate found indoors have been reported to be dominated by outdoor sources ([Weisel et al., 2005](#); [Yocom, 1982](#)). In contrast, indoor sources contribute more than outdoor sources to residential organic PM concentrations ([Polidori et al., 2006](#)). Results from the Total

Exposure Assessment Methodology (TEAM) study suggest that for most commonly measured volatile organic compounds, exposures are dominated by small sources close to the individual, usually indoors ([Wallace, 1986](#)). Formaldehyde, acetaldehyde, asbestos, chloroform,  $\alpha$ -pinene, and D-limonene exposures are also dominated by indoor sources ([Weisel et al., 2005](#); [Yocom, 1982](#)).

Assuming complete mixing, the concentration of a pollutant found in a home can be described with a single-compartment mass-balance model:

$$V(dC_i / dt) = PVaC_a - VaC_i + Q_i - kC_iV \quad (1)$$

where  $C_i$  and  $C_a$  are the indoor and outdoor compound concentrations ( $\mu\text{g}/\text{m}^3$ ),  $P$  is the fractional penetration of the compound through the building envelope,  $a$  is the air exchange rate ( $\text{h}^{-1}$ ),  $k$  is the compound loss coefficient ( $\text{h}^{-1}$ ) describing physical and chemical loss mechanisms indoors,  $Q_i$  is the strength of indoor sources ( $\mu\text{g}/\text{h}$ ), and  $V$  is the house volume ( $\text{m}^3$ ). Equation 1 describes the accumulation or depletion of a compound in the home as a function of: (i) the flux (mass/time) of compound into the home from outside due to air exchange, (ii) the flux out of the home with air exchange, (iii) the flux (mass/time) of compound introduced due to formation or emission of the compound indoors, and (iv) the flux (mass/time) lost by deposition or by reaction indoors. At steady state, this equation becomes:

$$C_i = \frac{PaC_a}{a+k} + \frac{Q_i/V}{a+k} = FC_a + C_{\text{pig}} = C_{\text{ai}} + C_{\text{pig}} \quad (2)$$

This equation describes the indoor compound concentration as the sum of the outdoor contribution (first term, Equation 2;  $C_{\text{ai}}$ ,  $\mu\text{g}/\text{m}^3$ ) and

the indoor contribution (second term, Equation 2;  $C_{\text{pig}}$ ). The quantity  $Pa/(a+k)$  or  $F$  describes the fraction of  $C_a$  that penetrates into and persists in indoor air (EPA, 2009).

Because the penetration through the building envelope ( $P$ ) and the loss rate indoors ( $k$ ) vary by pollutant, the composition of ambient air pollution is altered with outdoor-to-indoor transport. Non-polar gases (e.g. benzene, toluene, xylene) have very small loss rates indoors and penetration factors very close to unity, yielding  $F \approx 1$ . In other words, nearly 100% of a non-polar atmospheric gaseous compound is transported indoors and persists. In contrast, polar, water-soluble gases (e.g. formaldehyde, glyoxal, hydrogen peroxide, nitric acid) penetrate the building envelope with lower efficiencies and have larger loss rates indoors (Nazaroff and Cass, 1986; Lunden et al., 2003). For example, the loss rate coefficient ( $k$ ) for formaldehyde is  $0.36 \text{ h}^{-1}$  for a typical home (Nazaroff and Cass, 1986). There is some evidence that outdoor-to-indoor transport of some water-soluble vapours (i.e. hydrogen peroxide) is negligible, based on extremely high losses between offices connected by drop ceilings (Li et al., 2002).

The concentration and composition of outdoor-generated  $\text{PM}_{2.5}$  also changes with outdoor-to-indoor transport (Hodas et al., 2012; Meng et al., 2007). Particle penetration through the building envelope and deposition indoors lead to  $F$  values of about 50% for  $\text{PM}_{2.5}$  ( $F = 0.3\text{--}0.8$ ; Chen and Zhao, 2011). In addition, since penetration and loss rates are different for different PM species and for particles of different sizes, outdoor-to-indoor transport modifies the composition, source contributions, and size distribution of ambient  $\text{PM}_{2.5}$  (Meng et al., 2007; Hodas et al., 2012).  $P$  and  $k$  are a strong function of particle size (Riley et al., 2002; Thatcher and Layton, 1995; Abt et al., 2000). Different particle formation mechanisms (e.g. combustion, abrasion) yield material that differs in composition and size distribution. For example,  $F$  for primary

fine ( $< 2.5 \mu\text{m}$ ) soil dust, which is mechanically generated and thus predominantly in supermicron particles, is quite small ( $F_{\text{soil}} \approx 0.04$ ; Meng et al., 2007). With the exception of nitrate, the penetration and persistence of secondary aerosol is much larger ( $F_{\text{secondary}} \approx 0.8$ ; Meng et al., 2007). There is some evidence that primary combustion-generated particles have somewhat smaller  $F$  values ( $F_{\text{combustion}} \approx 0.5$ ; Meng et al., 2007;  $F_{\text{elemental carbon}} \approx 0.6$ ; Lunden et al., 2008), and aerosol physics suggests that infiltration factors for freshly formed ultrafine particles are considerably lower (Riley et al., 2002). Thus, in the indoor environment, *PM of outdoor origin* has a composition that is enriched in accumulation-mode constituents relative to supermicron and ultrafine particle constituents (Meng et al., 2007).

Aerosol thermodynamics also changes the composition of atmospheric particles found indoors. The most dramatic effect can be seen for nitrate. Particulate ammonium nitrate and gaseous nitric acid are transported indoors, where nitric acid is readily lost by deposition and sorption to indoor surfaces (Lunden et al., 2003; Hering et al., 2007). This loss disturbs the equilibrium between nitrate and nitric acid, resulting in transformation of ammonium nitrate to gaseous nitric acid and ammonia. Thus, only about 12% of ambient outdoor particulate nitrate is found in indoor air ( $F_{\text{nitrate}} \approx 0.12$  at an air exchange rate of  $0.45 \text{ h}^{-1}$ ; Hodas et al., 2012). In locations where nitrate is a major outdoor particle constituent, this process substantially reduces  $F$  for  $\text{PM}_{2.5}$  as well as substantially altering the particle composition. The implication is that people are exposed to much less ammonium nitrate than found at ambient sampling sites.

Changes in gas-particle partitioning of semivolatile organics (including PAHs, organic acids, and brominated flame retardants) can also occur with outdoor-to-indoor transport. Indoor-outdoor temperature differences can drive this process. Also, indoor concentrations of

particulate organic matter are often considerably larger than outdoor concentrations, presumably because of indoor sources of organic PM (Polidori *et al.*, 2006). Since semivolatile organics absorb into organic PM, the presence of additional organic material indoors can drive partitioning of these trace compounds (emitted indoors or outdoors) into the particle phase (Naumova *et al.*, 2003). However, semivolatile organic gases are also lost to indoor surfaces. Changes in gas-particle partitioning of semivolatile species with outdoor-to-indoor transport are unlikely to have a large impact on the bulk composition of PM but could have a noticeable impact on the partitioning and fate of semivolatile organic air toxics.

Outdoor-to-indoor transport of ozone has also been observed to facilitate the oxidation of volatile organic compounds emitted indoors, altering the properties and fate of these organic emissions. Products of these reactions are more water-soluble than their precursors, and include aldehydes, peroxides, and organic acids (Weschler and Shields, 1999; Li *et al.*, 2002). These reactions can also form secondary organic PM indoors (Weschler and Shields, 1999).

Temporal and geographical variations in  $F$  could, under certain circumstances, lead to a downward bias and underestimation of effects derived from epidemiological analyses (Dominici *et al.*, 2000; Zeger *et al.*, 2000; EPA, 2009). Variability in  $F$  has been identified as a source of exposure error in epidemiological analyses that use central-site  $PM_{2.5}$  as a surrogate for exposure to ambient fine particles (Dominici *et al.*, 2000; Zeger *et al.*, 2000; Hodas *et al.*, 2012).  $F$  varies spatially and temporally because (i) the air exchange rate depends on the housing stock, indoor-outdoor temperature difference, and ventilation practices (e.g. windows); (ii) the atmospheric conditions, season, and source mix affect particle properties (and  $P$ ,  $k$ ); and (iii) air conditioning and filtration affect particle losses indoors (Riley *et al.*, 2002; Hodas *et al.*, 2012). In fact, several studies have shown a reduced

risk of PM-associated mortality or morbidity with increased prevalence of air conditioning (Janssen *et al.*, 2002; Zeka *et al.*, 2005; Franklin *et al.*, 2007; Bell *et al.* 2009). Air conditioning use increases particle losses indoors, decreases  $F$ , and thus decreases exposure to ambient  $PM_{2.5}$ . Further, homes that use air conditioning are more likely to have their windows closed, reducing air exchange rates (and  $F$ ) compared with homes with open windows (Breen *et al.*, 2010). Certainly, the use of air conditioning varies seasonally and geographically and is one reason, among others, for heterogeneity in  $F$ .

Several studies are currently under way that explore to what degree  $F$  modifies associations between air pollutants and health effects in epidemiological studies. One such study found that the variation in ozone mortality coefficients across cities in the USA could be partially explained by variations in air exchange rate and thus variations in the fraction of outdoor ozone found indoors (Chen *et al.*, 2012). Certainly, the epidemiological model matters. For example, in a case-crossover study, factors that differ across subjects but are largely constant within subjects (e.g. differences in air exchange rates or particle losses stemming from differences in housing stock and residential proximity to local sources) would not be expected to contribute to exposure error (or bias) in this type of study design. Similarly, when case-control periods are confined to one calendar month, *seasonal* variability in  $F$  (i.e. due to seasonal differences in the air pollutant mix) would be controlled by design. Matching case and control periods by weekday and hour also addresses, to a degree, within-subject variability in human activity patterns.

In summary, the air pollution mixture is spatially and temporally heterogeneous. Primary pollutants (more lipid-soluble) exhibit steep concentration gradients close to sources, and secondary pollutants (more water-soluble) are often homogeneously distributed on a regional scale. The chemical and physical properties of

the pollutants affect their atmospheric lifetime, penetration and persistence in indoor environments, and lung deposition. Human exposure to the ambient air pollution mixture is affected by: transport of outdoor pollutants into indoor environments; the modifications of the pollutants indoors; and time–activity patterns of people. This is because most exposure to outdoor pollutants comes from exposure to these pollutants indoors. Under some circumstances, variations in the fraction of an ambient pollutant found indoors can affect pollutant–mortality associations that are based on outdoor concentrations.

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