

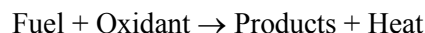
CHAPTER 4. COMBUSTION EMISSIONS

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Many of the toxic air pollutants, or air toxics, to which we are exposed are combustion-related ([Kinney *et al.*, 2002](#); [Lim and Turpin, 2002](#); [SCAQMD, 2000](#); [Manchester-Neesvig *et al.*, 2003](#)). The International Agency for Research on Cancer (IARC) list of Group 1 carcinogens includes a large number of these agents, including benzene, diesel exhaust, benzo[*a*]pyrene (B[*a*]P, a polycyclic aromatic hydrocarbon [PAH]), indoor emissions from coal combustion, and 1,3-butadiene. Gasoline engine exhaust is a Group 2B carcinogen, although it contains benzene, B[*a*]P, and 1,3-butadiene. (Tobacco smoke is also a combustion product with similarities to other combustion emissions, but is not covered here.) The World Health Organization, in the Global Burden of Disease study, found that emissions from indoor fuel combustion and urban particulate matter (PM) (much of which is combustion-related) are leading causes of premature mortality from environmental exposures ([Ezzati *et al.*, 2006](#)). The Multiple Air Toxics Exposure Study II (MATES-II; [SCAQMD, 2000](#)) of air toxics in the Los Angeles, California, region found that of the air toxics studied, the four compounds that had the greatest potential risk, combining both the estimated potential exposure and carcinogenicity, were primarily from

combustion, including diesel PM, 1,3-butadiene, benzene, and carbonyls (e.g. aldehydes, primarily formaldehyde). Combustion-related sources, including automobiles and indoor heating and cooking, are widespread and tend to be associated with more populated areas, leading to high potential exposures and health risks.

Combustion is the reaction between a fuel and oxidant accompanied by the release of heat:



Typically, the fuel is carbonaceous, such as gasoline, wood, or coal, and the oxidant is the oxygen in air, although there are non-carbonaceous fuels, notably hydrogen. While such processes are rare, combustion can take place without using air to provide the oxidant. The heat generated by combustion is typically used for cooking, heating, or producing power. The main products of hydrocarbon fuel combustion are carbon dioxide (CO₂) and water. However, combustion can lead to emissions of other compounds due to impurities in the fuel, the presence of nitrogen in air, or incomplete combustion. Specific sources of potentially carcinogenic air pollutant emissions that involve combustion include internal combustion engines (ICEs) (e.g. diesel, gasoline, turbine), external combustion

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boilers (as used for electricity generation), cement kilns, biomass burning (for cooking, heating, land management, and unplanned fires), waste combustion, and more ([Smith, 2002](#); [Lim, 2004](#); [Zielinska et al., 2004a](#), [2004b](#); [Lemieux et al., 2004](#)).

Here, we address combustion-related emissions, beginning with a brief discussion of combustion-derived pollutants, the combustion process, and toxic pollutant formation. Combustion sources are identified and the associated types of emissions discussed. Automotive emissions and biomass burning are major contributors to potentially harmful exposures, and are addressed in more detail in other chapters. When source emissions are discussed, potential controls are also identified.

Combustion-derived air toxics

A large fraction, but not all, of the air toxics emitted and/or formed during combustion are organic molecules and carbonaceous structures. Some are relatively simple molecules, such as formaldehyde (HCHO), increasing in complexity to compounds like 1,3-butadiene, aromatics, PAHs, and dioxins. Some sources emit soot, which is condensed organic material, part of which may approach elemental carbon. (Elemental carbon [EC] is currently operationally defined, i.e. it is dependent on the technique used for quantification. Black carbon [BC] is similar, although not identical, to EC, and concentrations of BC and EC tend to be highly correlated.) Soot is not composed of a single type of molecule but is made up of a variety of lower-volatility compounds, including PAHs, possibly on a core of a structure that resembles EC (although it can contain a variety of impurities). [Table 4.1](#) lists several combustion-derived organic air toxics, some of which, such as benzene, 1,3-butadiene, and diesel exhaust, have been classified by IARC as known human carcinogens. Several these species are semivolatile and may be found either

Table 4.1 Examples of combustion-derived air toxics

Gas phase	Particulate matter	Semivolatile ^a
Formaldehyde	Diesel	Polycyclic aromatic hydrocarbons
Acetaldehyde		Polychlorinated biphenyls
Acrolein		Furans
Benzene		
Toluene		
o-, m-, p-Xylenes		
1,3-Butadiene		

^a May be found in either gas phase or primarily condensed phase as particulate matter.

as gases or as part of the PM, including many of the PAHs ([Simoneit et al., 2004](#); [Zielinska et al., 2004a](#), [2004b](#)).

Inorganic emissions of concern include acids, such as sulfuric and hydrochloric acid, sulfur and nitrogen oxides (NO_x), and minerals. These are usually derived from contaminants in the fuel, although NO_x are formed, in part, from the nitrogen that makes up the bulk of air. Sulfur, which is present in many fossil fuels, is oxidized during combustion to both sulfur dioxide (SO₂) and sulfur trioxide. Sulfur trioxide condenses with water to form sulfuric acid. Chlorine reacts with hydrogen during combustion to form hydrochloric acid. Coal and oil can contain a variety of minerals, including iron and silicon oxides. These minerals typically are emitted as small particles.

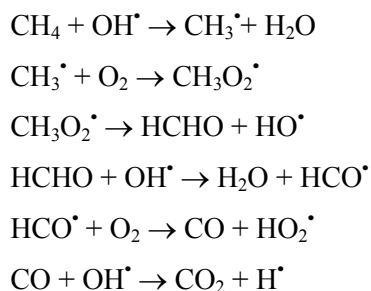
Combustion process

Combustion is a complex phenomenon involving chemical reactions and heat and mass transfer occurring on scales from atomic to potentially centimetres (e.g. a car engine), metres (e.g. a coal combustor), or kilometres (forest fires). Unless the fuel and oxidant are both simple molecules (e.g. hydrogen and oxygen, leading to

water), the products of combustion can be, and often are, as complex and varied as the combustion processes, forming as many compounds as originally present, if not more. Some of these species, such as 1,3-butadiene and PAHs, are known human carcinogens. Even a molecule as simple as methane burning in air can lead to larger molecules and soot.

The heterogeneity of real-world combustion leads to the complexity of the process and the wide range of compounds that are formed. In and around the combustion zone, the concentrations of species (e.g. the fuel, oxidant, and combustion products) vary by orders of magnitude over molecular scales. Temperature also can change rapidly over a few millimetres (if not less). In addition, flame fronts can move quickly; thus, the fuel can heat rapidly, start to combust, and then cool, quenching further reaction. Similarly, part of the flame can have excess oxygen while another part has excess fuel.

During combustion, the fate of organic fuel molecules is largely determined by the local conditions (e.g. temperature, abundance of oxygen, and time). If ample oxygen is present locally, i.e. directly where the combustion is occurring, the fuel will tend to oxidize and break down to smaller organic molecules until ultimately forming carbon monoxide (CO) and CO₂. For example, consider a mechanism sequence for methane oxidation:

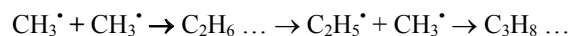


where the • indicates a very reactive radical intermediate.

A few characteristics of this process are important. First, even for a simple molecule

such as methane, the chemistry becomes complicated. This sequence shows only a fraction of the possible reactions; [Held and Dryer \(1998\)](#) show 89 reactions for methanol oxidation, excluding any reactions involving NO_x formation. If one considers combustion in fuel-rich conditions, the combustion mechanism becomes many-fold larger. For a two-carbon molecule, the complexity of the mechanism increases dramatically; more so for more complex molecules. Second, formaldehyde, an air toxic, is produced. It may also be destroyed later, but it is formed and can be emitted if the time for combustion is short (e.g. in an ICE). Larger molecules will lead to the formation of many other, more complex intermediates, including higher aldehydes (e.g. acetaldehyde from ethanol) and other air toxics (e.g. 1,3-butadiene). Third, CO₂ is the final end product, but only after several intermediate reactions. CO oxidation is relatively slow, so if conditions do not permit more complete combustion (i.e. low temperature, limited availability of oxygen, and short residence times), large quantities of CO can be emitted (e.g. in an automobile engine where residence time is limited and the car may be forced into an oxygen-limited condition during higher loads). Again, for larger molecules more reactions are required to form those ultimate products.

The above reaction mechanism assumed ample oxygen and sufficient time to react, and did not consider the possibility of organic radical intermediates reacting with each other to form larger organic molecules, which can be important to emissions of toxics. If, instead of reacting with oxygen, the methyl radical (CH₃•) reacts with another methyl radical, ethane can be formed, along with even larger molecules:



Larger molecules can then continue to react, ultimately forming various air toxics, including PAHs and soot, as discussed below.

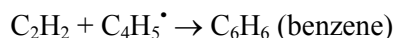
In general, the emissions of air toxics from combustion can be reduced by raising the temperature of combustion, increasing oxygen availability, and allowing a longer reaction time. For example, such conditions are in place for coal combustion in power plants, where relatively small (but non-zero) amounts of air toxics are emitted. However, ICEs have a more limited residence time in the combustion region, cooler reaction zones (particularly near the walls), and areas of reduced oxygen. Thus, one finds higher levels of products of incomplete combustion, such as CO, aldehydes, 1,3-butadiene, PAHs, and soot, emitted from such systems.

Polycyclic aromatic hydrocarbon emissions

PAHs can be formed during combustion when carbonaceous (organic) fuels are used. The formation of PAHs is not so surprising when the fuel (or the engine lubricant) already includes aromatics, but even those engines using so-called clean fuels, involving smaller organic molecules, such as methane, can produce PAHs.

Aromatics can grow to PAHs by addition of non-aromatic molecules to an already existing aromatic structure, or by reacting directly with other aromatic radicals. After a two-ring PAH is formed, by further reaction (again either by addition of non-aromatic radical intermediates or by reacting with aromatic radicals), three-ring and higher chains are formed.

Non-aromatic molecules (e.g. alkanes and olefins) also form PAHs, but at much lower efficiencies. While a variety of mechanisms exist, for example consider the reaction involving acetylene (C_2H_2) and the 1,3-butadienyl radical ($C_4H_5^\bullet$):



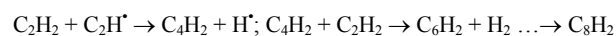
The benzene can then grow by addition of more organic chain radicals with and without aromatic structures. If significant levels of

oxygen are present, the 1,3-butadienyl radical will instead oxidize to aldehydes and other oxygenated organics with fewer carbon atoms.

Soot formation

Current evidence indicates that PM has somewhat greater health impact than other air pollutants. Results from [Laden *et al.* \(2000\)](#) and others ([Metzger *et al.*, 2004](#)) suggest that mobile source-derived and/or carbonaceous PM may have greater impacts than other components. The MATES-II study found that diesel PM is the major air toxic of concern in the Los Angeles, California, region ([SCAQMD, 2000](#)) ([Fig. 4.1](#)). These results raise the question as to the formation of carbonaceous PM emissions, for example soot during combustion.

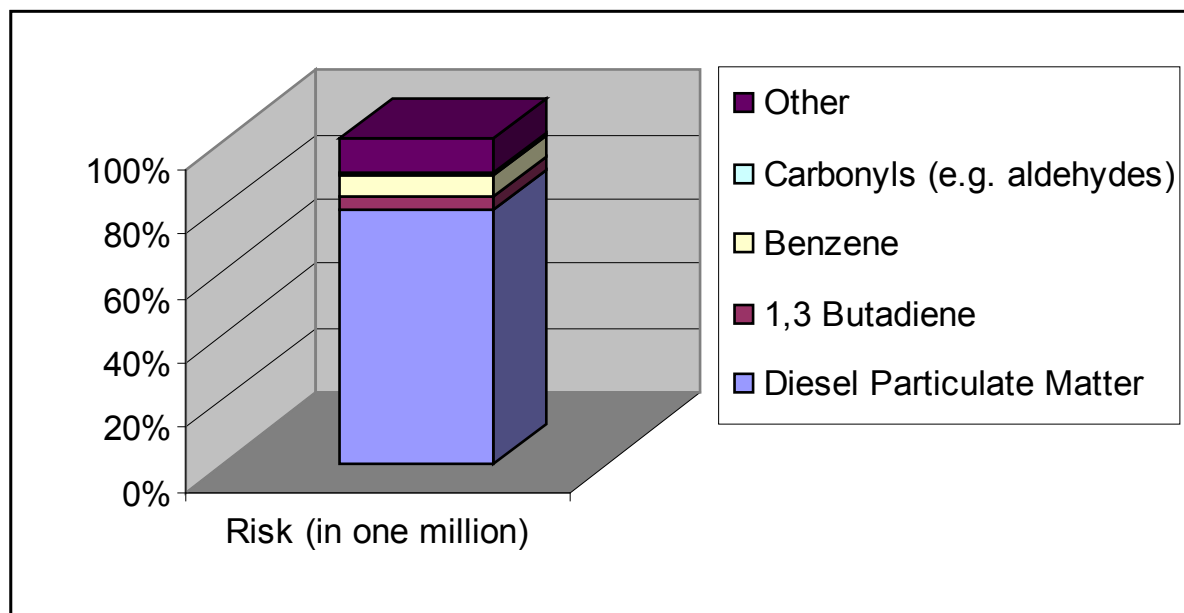
Soot formation is related to PAH formation, and aromatic fuels tend to produce more soot than others ([Flagan and Seinfeld, 1988](#); [Haynes, 1991](#)). In one mechanism, PAHs continue to grow as discussed above until they can condense and ultimately form solid particles. Again, even smaller organics can form soot; one mechanism is the formation of PAHs as discussed above, another is via the formation of polyacetylenes and continued reduction of the H:C ratio (soot has a very low H:C ratio, approaching that of EC). For example,



This process can continue until much larger, non-volatile structures (i.e. soot) are formed. Soot can also be produced by the removal of hydrogen from liquid carbonaceous fuels.

Critical to soot formation and growth is particle inception, where the first identifiable solid particles are formed. These particles are on the order of a nanometre and are composed of *sheets* of on the order of 100 carbon atoms. After inception, these particles can grow more rapidly by further reaction with organics on their surfaces

Fig. 4.1 Cancer risks at the MATES-II fixed sites



Risks are shown for all sources, including diesel particulates (top). The “other” portion is primarily non-combustion sources, although it includes PAHs not associated with diesel particulate matter. Compiled from [SCAQMD \(2000\)](#).

and by condensation of non-volatile species as the environment cools. They can also coagulate with other soot particles, leading to long-chain, fractal aggregates composed of hundreds of smaller spheres. The particles can grow by many orders of magnitude, often to diameters of $0.1\ \mu\text{m}$ or more, before being emitted.

Once formed, soot burns out more slowly. These kinetics partially explain why diesel vehicles emit soot even though they are operated fuel-lean (oxygen-rich overall). Near the fuel droplets and on cylinder surfaces, the combustion can be taking place in a fuel-rich environment, leading to formation of PAHs and soot. There is not ample time to oxidize the soot particles when they reach an oxygen-rich zone.

Soot from virtually any source (e.g. diesel engines, biomass burning, cooking of meat) is composed of a large number of different organic molecules, from very large, very low-volatility compounds to semivolatile species that are found

simultaneously in appreciable quantities in both the gas and particle phases.

Coke and char formation

Coal and fuel oil combustion can lead to the formation of char and coke. These are the carbonaceous residue particles that remain if the original solid or liquid fuel does not have time to fully combust. Char is formed as the volatile components in the coal escape due to the high temperatures, leaving the solid, nearly EC structure behind. Coke is formed from the liquid-phase pyrolysis of fuel oil. Ample reaction time can allow oxidation of char and coke.

Ash formation

Coal and, to a lesser extent, heavy fuel oils contain non-combustible materials such as minerals, including silicon, nickel, aluminium, and calcium, and trace quantities of other metals

like selenium, cadmium, and so on as inclusions in the fuel. As the fuel burns, these inclusions become molten and agglomerate with each other. A small fraction of the mineral material can also vapourize and then condense as the temperature cools. The particles formed are initially quite small ($< 0.01 \mu\text{m}$) but grow due to the molten material agglomeration, condensation, and coagulation, leaving a large fraction of the produced particles $> 1 \mu\text{m}$. Condensing species include vapourized minerals, sulfuric acid, and organics. [Chowdhury \(2004\)](#) reported on the analysis of coal fly ash and found elevated levels of a variety of PAHs, including pycene.

Combustion sources and systems

There are a variety of combustion systems that lead to potentially toxic emissions. In more developed countries, emissions from traditional diesel engines (also referred to as compression ignition [CI] engines) and gasoline-fuelled spark ignition (SI) engines contribute significantly to human exposure. They are discussed briefly below and in more detail in other chapters. Biomass fuel combustion is similarly dominant in rural parts of developing countries and is also discussed in more detail in other chapters. Other major sources of concern are discussed below.

Internal combustion engines

ICEs, primarily diesel (or CI) and gasoline (or SI) engines, are typified by having limited time for combustion in the cylinder. This short time leads to a relative abundance of products of incomplete combustion. When traditional hydrocarbon-based fuels are burned, such products include: CO and NO_x ; unburnt organics (e.g. benzene that was present in the fuel); partially oxidized organics, such as aldehydes; products of pyrolysis, such as PAHs; and soot ([Haynes, 1991](#)). Part of the emissions from ICEs are due to incomplete combustion of fuel and lubricating

oil ([Sheesley et al., 2009](#); [Goldstein, 2012](#)). There is evidence that with time the lubricating oil in the crankcase is enriched in PAHs by leakage from the pistons, further increasing such emissions. ICEs also emit NO_x from fixation of the nitrogen in air. Automotive emissions have evolved over time as various controls, such as catalysts, have been implemented. These controls have dramatically reduced the amounts of emissions (by an order of magnitude for some compounds) but have altered the composition as well, and some pollutants, such as ammonia and hydrogen cyanide, can be formed due to the controls ([Baum et al., 2007](#)).

Alternative automotive fuels

ICEs, both CI and SI, can be operated on non-traditional fuels. This is often done to lower emissions, although other reasons exist (e.g. as a renewable energy source). For example, SI engines sometimes operate on alcohols (methanol and ethanol), natural gas (primarily methane), or liquefied petroleum gas (largely propane). Diesel engines can be operated on a range of fuels as well, including natural gas and biodiesel.

SI engines using alcohols and natural gas tend to have simpler, although not necessarily less toxic, emissions compared with those using gasoline. Alcohols used are largely one- and two-carbon molecules and naturally have oxygen present, which can lead to lower CO emissions and much lower emissions of air toxics such as 1,3-butadiene, benzene, and PAHs. While often viewed as a clean-burning fuel, alcohols tend to form greater quantities of aldehydes with the same carbon number as the parent fuel. For example, ethanol use leads to increased emissions of acetaldehyde, and methanol use leads to larger quantities of formaldehyde being produced ([NRC, 1996](#)). Like conventionally fuelled vehicles, part of the emissions are due to lubricating oil.

Combustion of municipal or medical solid waste

Combustion is a widely used practice to deal with municipal solid waste (MSW) (trash), competing with landfilling. While it has the advantage of greatly reducing the volume of waste to be disposed of, it can lead to emissions of compounds viewed as potentially toxic. Three classes of compounds stand out: chlorinated organics (e.g. polychlorinated biphenyls [PCBs], dioxins, and furans), PAHs, and mercury.

MSW combustion is typically conducted at relatively lower temperatures and in less than optimal conditions with an inferior, more heterogeneous fuel than in utility boilers and automobiles. Even in controlled combustion with a relatively homogenous fuel, combustion is subject to widely varying conditions, leading to formation of undesirable products. The heterogeneity of MSW exacerbates problems, including having areas of lower oxygen and temperatures, leading to incomplete combustion. MSW also contains a much wider range of compounds than virtually any other combustion fuel, basically because it comes from whatever may be thrown away. This includes items like batteries, cans, plastics, newsprint, biomass, used oil, paint, and so on. Many of these contain toxic metals such as mercury, lead, and chromium. During the combustion process, these metals can be released, either as a gas or as PM, while a good fraction can be removed as ash. MSW combustion was a dominant source of mercury, although controls are effective at removing this pollutant. A sizable fraction of MSW can contain chlorine and other halogens, which can add to the organic molecules to produce dioxins and dibenzofurans.

MSW combustion emissions can be controlled in a variety of ways. First, the fuel can be prepared to improve the combustion characteristics, for example pelletized. MSW that has potentially harmful compounds can be removed (e.g. waste with harmful metals such as lead

and mercury). Combustion can be assisted by burning a higher quality fuel like natural gas to raise the temperature and obtain more complete MSW destruction. Post-combustion controls, such as scrubbers and fabric filters, have substantially lowered emissions.

Electricity generation

Coal is the dominant fossil fuel used for generating electricity and is the closest to being pure carbon. However, it still contains hydrogen and a range of other elements, including sulfur and nitrogen (possibly in relatively large amounts), chlorine, and metals such as iron, selenium, and mercury. The chlorine can react to form hydrochloric acid, the sulfur to form SO₂ and sulfuric acid, and the nitrogen to form NO_x, which is also formed from oxidation of the nitrogen in air. Coal-fired utilities remain one of the largest anthropogenic sources of both SO₂ and NO_x, both of which lead to the formation of acids in the atmosphere. Further, NO_x plays a critical role in driving the photochemical production of ozone and secondary particulates, which include nitrated PAHs and organic and inorganic acids.

Coal combustion is a major anthropogenic source of mercury to the atmosphere, along with MSW combustion ([Seigneur et al., 2004](#)). Methylmercury is toxic and bioaccumulates; levels in fish have become dangerously high. Much of the mercury emitted from power plants, however, is in the elemental, gaseous form, which is not assimilated directly by plants and animals. However, elemental mercury does slowly oxidize in the atmosphere and then deposits to the ground and surface waters, where it can enter the food chain. In addition, coal-fired utilities emit fly ash containing a variety of minerals and metals.

Controls on coal-fired utility boilers include filters (baghouses) (for controlling PM), scrubbers (SO₂, PM, mercury), electrostatic precipitators (PM), selective catalytic reduction (NO_x),

burner modification (e.g. low NO_x burners), and fuel modification, including the use of cleaner coals, washing, and using alternative fossil fuels such as natural gas. Such controls have effectively reduced emissions by > 80% for NO_x and SO₂ and by > 99% for PM.

Because of environmental and economic concerns, natural gas has become increasingly popular for producing electricity, while the use of fuel oil is declining. Natural gas combustion is typically cleaner than coal combustion for a variety of reasons. The gas phase allows for more homogeneous combustion conditions, and the smaller molecules being burned with ample residence time and excess oxygen lead to almost complete combustion to CO₂ and water. Trace species (e.g. small amounts of sulfur, but virtually no minerals and other contaminants) and thermal fixation of nitrogen do lead to small amounts of air pollutant emissions. When used as the primary fuel, natural gas is usually applied in turbines. Combustion is typically conducted at high temperatures with a reasonably long residence time, leading to more complete combustion and lower air toxics emissions.

Industrial process combustion

Several industries use combustion for purposes such as producing heat and destroying undesirable compounds. An example that may do both is using cement kilns to destroy toxic waste such as PCBs. Cement kilns are used in the process of calcining cement, which requires high temperatures. The residence time during combustion is relatively long, so it was proposed to burn toxic organics, which have a heating value, along with traditional fuels, to destroy the undesirable compounds. While this does destroy almost all of the original organic material, there can be some slippage (typically < 0.01%) in addition to the products of incomplete combustion. Much of the time coal is used, leading to emissions similar to those of coal-fired power plants.

Another fuel used by cement kilns is old tyres (or other waste with heating value), which can lead to somewhat higher levels of soot and PAHs, although the long residence time tends to minimize such emissions.

Use of industrial process boilers to provide heat and steam is also widespread. Emissions from such sources are fuel-specific, with coal, natural gas, and oil being the dominant fuels used. Emissions from industrial boilers will be similar to those from utility boilers using the same fuel, recognizing that the controls may not be as extensive since the source is smaller.

While combustion often is cited for increasing emissions, flaring has been used to reduce emissions of volatile organic compounds (VOCs), including toxic compounds such as benzene and 1,3-butadiene. While this approach is effective at removing the parent VOC, it can lead to the production of lesser amounts of organic toxics, for example PAHs. Catalytic destruction is also used to help remove unwanted VOCs before emission from industrial facilities.

Residential coal combustion

While not widely used in residential applications in developed countries, coal is still used for heating and cooking in developing countries where it is a plentiful resource. In this case, the conditions are not as favourable to complete combustion as in typical industrial or utility facilities, and post-combustion controls will be minimal or non-existent. In such cases, emissions can be quite large and will include soot, char, ash, PAHs, and CO. The enclosed environments further lead to potentially very high exposures ([Smith, 2002](#)).

Biomass combustion

Burning modern fuel (usually biomass) occurs both intentionally (e.g. for heating, cooking, or land management) and unintentionally (forest

fires). On a worldwide scale this process leads to more PM emissions (and likely more toxics) than does the combustion of fossil fuels, which is typically done under more controlled conditions. In developed countries, however, biomass combustion usually occurs in less densely populated areas and the resulting exposures are not as severe (unless one includes cigarette smoking, the major exposure to air toxics of all sources). However, biomass combustion is still a major contributor to PM in many urban areas ([Schauer et al., 1996](#); [Zheng et al., 2000, 2002, 2005](#)) and is discussed in more detail in other chapters.

Anthropogenic fugitive combustion

Building fires can lead to the emissions of PCBs, for example from the formation of PAHs and soot during the low-temperature smouldering and burning of plastics and electrical wiring insulation. This occurred after the attack on the World Trade Center, where the smouldering wreckage led to exposure to air toxics ([Pleil et al., 2004](#)). Fires in scrap tyre stockpiles and areas with concentrated refuse catching fire can lead to emissions of toxics (including organic-laden PM) ([Christian et al., 2010](#); [Lemieux et al., 2004](#)).

Meat cooking

While meat cooking is not often considered a major source of PM, Cass and co-workers ([Kleeman et al., 1999](#); [Zheng et al., 2002](#)) have found surprisingly large quantities of carbonaceous PM in urban areas to be due to meat cooking. In this case, the fat from the cooking process can be volatilized and/or partially combusted to form less-volatile compounds. The volatilized material then condenses back onto pre-existing particles. Compounds formed from meat cooking include PAHs.

Summary

Combustion is a ubiquitous process leading to environmental air toxics exposures and emissions of a wide range of species. Combustion-generated compounds can be either organic (e.g. aldehydes and PAHs) or inorganic (e.g. acids and metals such as mercury). In more developed countries, motor vehicle emissions (both on- and off-road) play an important role in exposure to air pollution, and key pollutants include PM (diesel and SI), benzene, 1,3-butadiene, aldehydes, and other organics, although levels are decreasing due to enhanced controls. Use of alternative fuels does not eliminate emissions of air toxics, although they will generate mixtures that differ from those of conventional fuels. In developing countries, indoor combustion of biomass or coal is a continuing source of concern, along with increasing exposures to automobile emissions as vehicle fleets grow.

Electricity generation tends to produce lower emissions of organic compounds as combustion is more complete. Power plants do emit PM (the amount largely depending on how well controlled the facility is), sulfur oxides, NO_x, and, of particular concern recently, mercury, although various controls have proven effective at reducing emissions. Historically, MSW combustion has also been one of the major sources of mercury and leads to the formation of halogenated organics such as dioxins and furans.

Control of air toxics emissions from combustion sources generally relies on improving combustion conditions and post-combustion controls, including scrubbers, filters, electrostatic precipitators, flaring (which can form other pollutants), and catalytic destruction.

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