

SMOKE FROM WILDLAND FIRES

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INTRODUCTION

Biomass burning is a major contributor of toxic air pollutants, particulate matter (PM), and greenhouse gases, but unlike some anthropogenic sources, poorly quantified. On a global basis, the net effect on human health and impact on climate change has not been adequately determined. The temporal and spatial distribution of these emissions is very difficult to estimate and the interannual variability of smoke production can range by an order of magnitude or more. With the advent of sensors operating over a temporal scale of hours, progress in estimating smoke emissions should be advanced. Still, the exposure of people to smoke emissions on a regional scale will continue to be very difficult to quantify.

Measurement techniques such as open-path Fourier transform infrared (FTIR) spectroscopy have allowed for measurements of oxygenated compounds and other more reactive compounds *in situ* rather than sampling for later analyses. Many different vegetation types have been examined for the release of CO₂, CO, and CH₄. Mass concentrations of products of incomplete combustion including particles less than 2.5 µm in diameter (PM_{2.5}) can generally be correlated with CO and CH₄. For example, Hao *et al* (1) reported the concentrations of many aliphatic hydrocarbons of C₆ and lower carbon number as being correlated with the concentration of CH₄. Some compounds such as those sulphur and nitrogen-based compounds depend both on the efficiency of the combustion process and the chemical composition of the vegetation burned by the fires. Utilizing ratios of air toxic compounds to CO, CH₄, and/or PM_{2.5} allows reasonable estimates of

exposure to a number of other compounds based on the characterization of one or two compounds. Of course, this assumes that the emissions are from biomass burning or that the contribution of biomass burning can be apportioned.

FACTORS AFFECTING INCOMPLETE COMBUSTION

Combustion efficiency is defined as the ratio of carbon released as CO₂ to total carbon. For convenience, we use modified combustion efficiency which is the ratio of carbon released as CO₂ divided by the sum of CO₂ and CO. The smouldering combustion phase, which is generally a very low-intensity combustion process, produces high emissions of particulate matter, CO, and other products of incomplete combustion. Fuel properties can markedly affect the amount of smouldering combustion and the overall mixture of combustion products. The mixture of products of combustion produced by a fire in a particular vegetation type may result in 90 per cent of the vegetation being consumed through flaming combustion (e.g., savanna ecosystems), and this would be quite different from a fire burning in a vegetation type where 90 per cent is consumed through smouldering combustion (peat, rotten logs, deep duff, etc.).

DISCUSSION OF PRODUCTS OF COMBUSTION

In the study of smoke related to its effect on the health of wildland firefighters, Ward *et al* (2) discussed several combustion products and classes of combustion products. These substances are categorized as follows:

1. Particulate matter
2. Polynuclear aromatic hydrocarbons
3. Carbon monoxide
4. Aldehydes
5. Organic acids
6. Semivolatile and volatile organic compounds
7. Free radicals
8. Ozone
9. Inorganic fraction of particles

Particulate matter

Particulate matter from wildland fires is highly visible, affects ambient air quality, and has various effects on human health. Particles are abundantly produced by forest fires with source strengths exceeding 0.6 tonnes per second on some large fires (3). The mass of particles can be separated into two modes: (i) a fine-particle mode generally considered to be produced during the combustion of organic material with a mean-mass diameter of 0.3 micrometres; and (ii) a coarse particle mode with a mean-mass diameter larger than 10 micrometres. Research, both from ground-based sampling (4) and airborne sampling systems, shows the bimodal distribution with a small fraction of the total mass (less than 10 per cent) between 2 and 10 micrometres (5). Smouldering combustion releases several times more fine particles than flaming combustion. The fine particles account for up nearly 100 per cent of the mass of particulate matter. The percentage of fine particles produced through flaming combustion ranges from 80 per cent to 95 per cent depending on the turbulence in the combustion zone and other factors. The smaller fine particles consist of 60 to 70 per cent organic carbon (4). Many known carcinogenic compounds are contained with the organic carbon fraction. Roughly, another 2 per cent to 15 per cent is graphitic carbon and the remainder is inorganic ash material (6). Particles are also known to carry adsorbed and condensed toxic gases and possibly free radicals.

Polynuclear aromatic hydrocarbons

Polynuclear aromatic hydrocarbons (PAH) is one class of compounds contained in the organic fraction of the fine particle matter. Some of the PAH compounds associated with the particles are carcinogenic. Benzo[a]pyrene, for example, is a physiologically active substance that can contribute to the development of cancer in cells of humans. Examples of PAH compounds are listed in Table 1 for prescribed fires in logging slash, laboratory fires of pine needles, fireplaces, and woodstoves. Not all of the compounds listed in Table 1 are of equal carcinogenicity. More data have been developed for benzo[a]pyrene than other PAH compounds for smoke from wildland fires. Ward *et al* (2) found for benzo[a]pyrene that emission factors increased proportionally to the density of live vegetation covering the prescribed fire units. This has not been verified for other ecosystems with live vegetation involved in flaming combustion.

PAH compounds are synthesized from carbon fragments into large molecular structures in low-oxygen environments, such as occurs inside the flame envelope in the fuel-rich region of the flame structure. If the temperature is not adequate to decompose compounds upon exiting from the flame zone, then they are released into the free atmosphere and condense or are adsorbed onto the surface of particles. Many different combustion systems are known to produce PAH compounds, and the burning of forest fuels is documented as one of these sources. Little is known about combustion conditions on wildfires, but recent experiments would suggest that emissions are not that different from prescribed fires when burning conditions are similar. Evidence suggests that for low-intensity backing fires, the ratio of benzo[a]pyrene to particulate matter is higher by almost two orders of magnitude over that for heading fire (7). For wood stoves, a relationship was established between burn rate and PAH production. Specifically, as the burn rate increased, total organic emissions decreased, but the proportion that was PAH compounds increased. DeAngelis *et al* (8) found the PAH emission rate to be highest over a temperature range of 500°C to 800°C. This would be consistent with the low-intensity backing fire results of McMahon and Tsoukalas (7).

Carbon monoxide

Carbon monoxide is a colourless and odourless toxic gas. It is produced through the incomplete combustion of biomass fuels. CO is second in abundance to CO₂ and water vapour. Carboxyhaemoglobin is created in the blood of humans in response to the exposure to CO, which replaces the capacity of the red blood cells to transport oxygen. Generally, a level of 5 per cent carboxyhaemoglobin results from three to four hours of exposure to CO of concentrations of 35 ppm and may result in people showing signs of disorientation or fatigue.

CO is produced more abundantly from smouldering combustion of forest fuels. Immediately following the cessation of flaming combustion, maximum levels of CO are produced. This phenomenon coincides with suppression activities, especially where direct attack methods are being used.

As the flames subside, CO is released at the highest rate and, typically, continues at a high rate during the first few minutes of the die down period. For fires burning under high drought conditions, the smouldering combustion can be self-sustaining and consume deep into the duff and in some cases, soil where the organic component of the soil makes up more than 30 per cent of the total. Tremendous amounts of smoke can be produced under severe conditions which is some times sustained for days and weeks.

Aldehydes

Aldehydes are compounds of which a few are extremely irritating to the mucous membranes of the human body. Some, such as formaldehyde, are potentially carcinogenic and in combination with other irritants may cause an increase in the carcinogenicity of compounds like the PAH compounds. Formaldehyde is one of the most abundantly produced compounds of this class and is released proportional to many of the other compounds of incomplete combustion. Formaldehyde is transformed rapidly to formic acid in the human body with formic acid being removed very slowly.

Acrolein is also known to be produced during the incomplete combustion of forest fuels. Acrolein is known to effect respiratory functions at concentrations as low as 100 ppb. Studies of pathogenesis in rabbits exposed to smoke from low-temperature combustion of pine wood suggest that low-molecular-weight aldehydes, including acrolein, are the most likely agents of injury. The ability of scavenger cells in the lung to engulf foreign material of bacteria is decreased through exposure to aldehyde compounds, which may accentuate infections of the respiratory system. Acrolein may have a high likelihood of making a discernible addition to the irritant character of smoke near firelines, and its concentrations could be as high as 0.1 to 10 ppm near fires.

Aldehydes as a class of compounds have been difficult to quantify for forest fires and there are still many issues to be worked out. Some recent research by Reinhardt *et al* (9) suggest that acrolein is produced proportional to formaldehyde. On the other hand, Yokelson *et al* (10) using a very straight forward analytical technique were not able to identify acrolein in as high a

concentrations as those reported by Reinhardt *et al* (9) and in much less abundance than formaldehyde.

Organic acids

Organic acids are known to form from the combustion of biomass fuels. Yokelson *et al* (11) and McKenzie *et al* (12) have recently made significant progress in characterizing some of the emissions of organic acids including acetic and formic acid finding molar ratios to CO of 7.4 ± 6.2 and 1.5 ± 1.5 , respectively. Through the application of the molar ratios of different air toxic compounds to CO, McKenzie *et al* (12) reported possible exposure levels that were well below the allowable time weighted averages (TWA's) based on a peak exposure of firefighters of 54 ppm [based on Reinhardt's *et al* (9) data for peak exposure]. No single compound is present at a hazardous level except for vinyl acetate and 2-furaldehyde, which are suspected carcinogens (Table 2). It should be noted however, that the synergistic effects of some or all of these compounds and others have not been determined.

Semivolatile and volatile organic compounds

Semivolatile and volatile organic compounds in smoke contain a wide variety of organic compounds, many with significant vapour pressures at ambient temperatures. Some compounds are partitioned between the gaseous and liquid or solid phase at ambient temperature; e.g., benzene, naphthalene, toluene. Fires are known to produce a variety of these types of compounds, but little characterization work has been done. The phenolic compounds are important because they contain compounds that are very strong irritants and are abundantly produced from the partial oxidation of cellulosic fuels. Various phenolic compounds are used as starting materials in the manufacture of resins, herbicides, and pharmaceutical products. Other PAH compounds of low-molecular weight are contained with the semivolatile class of compounds. Because of the volatility and in some cases reactivity of these compounds, special sampling protocols are required including charcoal adsorption, porous polymer adsorption, and whole-air sampling. These materials are difficult to sample, and surrogate methods are needed for correlating exposures of the more volatile materials with the semivolatile components. Methane and carbon monoxide gases are often produced proportional to other products of incomplete combustion and may serve as indicators of their abundance.

Free-radicals

Free-radicals are abundantly produced through the combustion of forest fuels. The concern lies with how long these materials persist in the atmosphere and their reactivity when in contact with human tissues. Most of the chemical bonding is satisfied through recombination of free-radical groups by condensation within the few seconds of time it takes for the mixture of gases to exit from the flame which should reduce the overall toxicity of the smoke. However, some free-radicals persist up to 20 minutes following formation and may be of concern to people exposed to fresh aerosols. How much of the organic material remains in a reactive, free-radical state is unknown.

Ozone

Ozone concentrations close to fires that are high enough to be concerned about would not be expected. Ozone is formed photochemically near the top of smoke plumes under high sunlight conditions. Generally, ozone is formed in situations where smoke is trapped in valleys or under temperature inversion conditions of the atmosphere, or both. Fire crews working at high elevation locations may encounter elevated levels of ozone. Any effort to characterize exposure of people to smoke from vegetation fires must account for the potential exposure to ozone in areas where personnel are working at elevations close to the top of the atmospheric mixing layer.

RATIOS OF TOXIC AIR POLLUTANTS TO CO, CH₄, AND PM_{2.5}

In performing a risk assessment and establishing the relative importance of different compounds from a human health standpoint, a method is needed to estimate the exposure levels based on the measurement of CO and/or PM. Many of the compounds discussed are very difficult to measure which makes breathing space sampling nearly impossible for most of the toxic air compounds. Correlations of toxic air compounds to CO, CH₄, and PM_{2.5} has proven to be an effective way of estimating the release of a number of compounds (11-13).

If this method is to be used, then it is important to give “safe-side” estimates or to use very specific information for the phase of combustion producing the smoke of concern. For example, ratios of benzo[a]pyrene to CO and/or to PM for different fuel types show a significant difference

between flaming and smouldering combustion and fuel type (Table 3). There is almost an order of magnitude difference between emission ratios of benzo[a]pyrene to CO for flaming in comparison to smouldering ratios. An average weighted emission ratio can be calculated based on the percentage of fuel consumed by phase of combustion producing the emissions contained within the breathing space. This can be done by assuming, for example, that the emissions along the fireline consist of 10 per cent from vegetation consumed during the flaming phase, 70 per cent for the first smouldering phase and 20 per cent for the final smouldering phase. The results are illustrated in Table 3. On the other hand, emissions released through flaming combustion are generally accompanied by the release of significant heat which lofts the emissions through convective forces acting on the smoke plume. Most of the emissions near the surface may be produced through smouldering combustion. It is recommended that emission ratios for smouldering combustion be used for assessing exposure, except for those conditions where 75 per cent to 80 per cent or more of the fuel is consumed through flaming combustion.

CONCLUSIONS

1. The mixture of particles, liquids, and gaseous compounds found in smoke from wildland fires is very complex. The potential for adverse health effects is much greater because of this complex mixture.
2. The particles are known to contain many important organic compounds some of which condense to form tarry droplets over a substrate material of ash or graphitic carbon or both.
3. The size distribution of smoke particles is such that a large percentage are respirable.
4. Gaseous compounds in the air adjacent to fires in association with the particles include carbon monoxide, methane, oxides of nitrogen and many organic compounds - some of which are carcinogens and many of which are irritants.
5. Other semi-volatile compounds have a significant vapour pressure at ambient temperature and pressure which results in a gas phase emission and many of these compounds to be important from a health standpoint, but have not been adequately quantified.

6. With the data available today, we still do not know what the overall toxicity of smoke is from wildland fires or how this toxicity varies from fire to fire.
7. The large variance in the concentration of smoke needs to be evaluated to assess the level of exposure and risk to humans.
8. The new USEPA PM_{2.5} air quality standard is designed to protect human health and suggests that health is most at risk from particles less than 2.5 µm in diameter.
9. Along with the combustion products is the dust, heat, and remoteness of many of the wildland fires making exposure to humans difficult to assess. The fire, fuel, and weather vary continuously, which changes the fire dynamics and the dilution occurring over time and space.

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Table 1
Comparison of polynuclear aromatic hydrocarbons from four sources: (i) prescribed fires in logging slash in western Washington and western Oregon (2); (ii) pine needle litter fuel of southeast Pinewood (7); (iii) fireplace emissions tests with green southern pine wood (8); and (iv) woodstove emissions tests with green southern pine wood (8). Carcinogenicity is from National Academy of Sciences (23) and is coded as follows: “-” is not carcinogenic; “±” is uncertain or weakly carcinogenic; and “++” or “+++” is strongly carcinogenic

Compound	Logging slash (mean±SD)	Pine needles (mean±SD)	Fire places	Wood stoves	Carcino- genicity
(µg of compound per g of particulate matter)					
Anthracene/phenanthrene	42±29	185±72	575	6345	-/-
Methylanthracenes	61±38	NA	692	3147	NA
Benz[a]anthracene/chrysene	17±8	43±25	117	2276	+/+
1,2-benzanthracene	17±8	NA	NA	NA	+
Chrysene/triphenylene	29±11	NA	NA	NA	NA
Dibenzanthracenes/ Dibenzophenanthrenes	NA	NA	4	3	NA
Fluoranthene	47±23	51±29	125	1153	-
Benzo[fluoranthene	NA	11±11	133	865	NA
Benzo (ghi) fluoranthene	11±5	NA	NA	NA	-
Benzo (a) fluoranthene	7±4	NA	NA	NA	NA
Benzo (b) oranthenes	26±9	NA	NA	NA	++
Benzo (j) oranthenes	26±9	NA	NA	NA	++
Benzo (k) oranthenes	26±9	NA	NA	NA	-
Pyrene	42±24	73±46	133	1153	-
Benzo(a)pyrene	13±14	3±2	NA	NA	+++
Benzo(e)pyrene	13±5	6±3	NA	NA	-
Benzopyrenes/perylene	NA	NA	117	578	NA
Perylene	3±2	2±2	NA	NA	NA
Indenopyrene	13±14	NA	NA	NA	NA
Indeno(1,2,3-c,d)pyrene	NA	NA	NA	NA	+
Anthanthrene/dibenzopyrene	6±8	NA	8	1	-
Benzo[ghi]perylene	15±19	NA	117	288	-

NA: Not available

Table 2
Listing of ratios of toxic air pollutants to CO determined for a variety of fuel types. The bold type values in column 3 are the ratios recommended for use in making risk assessments and are calculated from the highest 1 to 3 values listed for each compound in column 2.

	Literature values	Values to be used for risk assessment
	mean molar ratio to CO ($\times 10^{-3}$)	mean molar ratio to CO ($\times 10^{-3}$)
	1.50±0.93 ⁽¹²⁾	1.50±0.93⁽¹²⁾
5-methylfuraldehyde	0.30±0.19 ⁽¹²⁾	0.30±0.19⁽¹²⁾
2-acetylfuran	0.33±0.16 ⁽¹²⁾	0.33±0.16⁽¹²⁾
Phenol	0.32±0.2 ⁽¹²⁾	0.32±0.2⁽¹²⁾
o-cresol	0.27±0.13 ⁽¹²⁾	0.27±0.13⁽¹²⁾
m/p-cresol	0.52±0.25 ⁽¹²⁾	0.52±0.25⁽¹²⁾
Guaiacol	0.17±0.081 ⁽¹²⁾	0.17±0.081⁽¹²⁾
4-methylguaiacol	1.00±0.83 ⁽¹²⁾	1.00±0.83⁽¹²⁾
Vanillin	0.50±0.57 ⁽¹²⁾	0.50±0.57⁽¹²⁾
Acetol	1.20±1.7 ⁽¹²⁾	1.20±1.7⁽¹²⁾
Vinyl acetate	1.70±2.1 ⁽¹²⁾	1.70±2.1⁽¹²⁾
2-cyclopenten-1-one	0.20±0.13 ⁽¹²⁾	0.20±0.13⁽¹²⁾
Acetic acid	7.40±6.2 ⁽¹²⁾ ; 22.6 ⁽¹¹⁾ ; 8.70±6.1 ⁽¹⁵⁾ ; 1.60±2.4 ⁽¹⁶⁾ ; 8.00±4 ⁽¹⁷⁾ ; 3.20±0.4 ⁽¹⁵⁾ ; 2.60±6.8 ⁽¹⁷⁾	7.40±6.2 ⁽¹¹⁾ ; 22.6 ⁽¹¹⁾ ; 8.70±6.1 ⁽¹⁵⁾ 12.1
Formic acid	1.50±1.5 ⁽¹²⁾ ; 1.6 ⁽¹⁴⁾ ; 9.1 ⁽¹¹⁾ ; 2.60±2 ⁽¹⁵⁾ ; 0.17±0.27 ⁽¹⁶⁾ ; 20.0 ⁽¹⁷⁾ ; 35.00±22 ⁽¹⁷⁾	9.1 ⁽¹¹⁾ ; 35.00±22 ⁽¹⁷⁾ ; 1.6 ⁽¹⁴⁾ ; 15.2
Propanoic acid	0.39±0.19 ⁽¹²⁾ ; 0.66 ⁽¹⁴⁾	0.66⁽¹⁴⁾
3-oxobutanoic acid	0.41±0.44 ⁽¹²⁾	0.41±0.44⁽¹²⁾
Methanol	11.00±9 ⁽¹²⁾ ; 18.0 ⁽¹¹⁾	11.00±9 ⁽¹²⁾ ; 18.0 ⁽¹¹⁾
Methane	29.00±11 ⁽¹²⁾ ; 55.00 ⁽¹³⁾ ; 83.4 ⁽¹¹⁾ ; 45.00±13 ⁽¹⁸⁾ ; 140.00±93 ⁽¹⁹⁾ ; 58.00±18 ⁽²⁰⁾ ; 71.00 ⁽¹³⁾ ; 91.00±3.1 ⁽²¹⁾ ; 76.00±13 ⁽²²⁾	83.4 ⁽¹¹⁾ ; 140.00±93 ⁽¹⁹⁾ ; 91.00±3.1 ⁽²¹⁾ ; 104.8
Ethane	2.50±1.2 ⁽¹²⁾ ; 9.4 ⁽¹¹⁾ ; 4.00±1.4 ⁽¹⁸⁾ ; 6.80±5.2 ⁽²¹⁾	9.4 ⁽¹¹⁾ ; 4.00±1.4 ⁽¹⁸⁾ ; 6.80±5.2 ⁽²¹⁾ 6.7
Ethene	12.00±9 ⁽¹²⁾ ; 13.5 ⁽¹¹⁾ ; 17.00±9.1 ⁽¹⁸⁾ ; 12.00±8.7 ⁽²¹⁾	12.00±9 ⁽¹²⁾ ; 13.5 ⁽¹¹⁾ ; 17.00±9.118 ⁽¹⁸⁾ 14.2

Table 2(continued)

	Literature values	Values to be used for risk assessment
	mean molar ratio to CO (x10 ⁻³)	mean molar ratio to CO (x10 ⁻³)
Glycol	10.8 ⁽¹¹⁾	10.8 ⁽¹¹⁾
Formaldehyde	17.3 ⁽¹¹⁾	17.3 ⁽¹¹⁾
Ammonia	26.0 ⁽¹¹⁾	26.0 ⁽¹¹⁾
HCN	4.0 ⁽¹¹⁾	4.0 ⁽¹¹⁾
1,3-butadiene	1.10 ⁽¹⁾	1.10 ⁽¹⁾
Benzene	2.13 ⁽¹⁾	2.13 ⁽¹⁾
Toluene	1.79 ⁽¹⁾	1.79 ⁽¹⁾
o-xylene	0.24 ⁽¹⁾	0.24 ⁽¹⁾
m,p-xylene	0.43 ⁽¹⁾	0.43 ⁽¹⁾
n-hexane	0.06 ⁽¹⁾	0.06 ⁽¹⁾
Pyruvic aldehyde	6.2 ⁽¹⁴⁾	6.2 ⁽¹⁴⁾
Crotonic acid	0.21 ⁽¹⁴⁾	0.21 ⁽¹⁴⁾

Table 3

Example of application of data for prescribed fires in the Pacific Northwest used as an estimate of emissions exposure of 10 per cent flaming, 70 per cent primary smouldering, and 20 per cent secondary smouldering. The ratios can be multiplied by the concentration of CO to calculate either B[a]P or PM exposure. If only PM exposure is available, CO can be calculated and B[a]P estimated along with other air toxics found in Table 2.

Phase of combustion	CO (ppm)	PM ($\mu\text{g}/\text{m}^3$)	B[a]P ($\mu\text{g}/\text{m}^3$)	B[a]P/CO ($\mu\text{g}/\text{m}^3/\text{ppm}$)	B[a]P/PM ($\mu\text{g}/\text{g}$)	PM/CO ($\mu\text{g}/\text{m}^3/\text{ppm}$)
F	140	15740	0.1284	0.0009	8.2	112.4
S1	113	8391	0.1608	0.0038	42.8	74.3
S2	26	1214	0.1024	0.0067	126.4	46.7
Weighted	98	7690		0.0040	56.1	78.2