

STUDIES OF EMISSIONS FROM OIL FIRES

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ABSTRACT: Over 45 mesoscale burns were conducted to study various aspects of diesel and crude oil burning *in situ*. Extensive sampling and monitoring of these burns were conducted at downwind stations, upwind stations, and in the smoke plume. Particulate samples were taken in air and analyzed for polycyclic aromatic hydrocarbons (PAHs). PAHs were found to be lower in the soot than in the starting oil, although higher concentrations of the larger molecular PAHs were found in the soot and residue, especially for diesel burns. Overall, the amount of PAHs in the soot and residue were about 2 to 8% of that in the starting oil. This implies a destruction of PAHs by 92 to 98%. Particulates in the air were measured by several means and were found to be greater than recommended exposure levels up to 500 meters downwind at ground level, depending on the size and type of fire. Diesel fires emit much more particulate matter and have longer exposure zones. Combustion gases including carbon dioxide and carbon monoxide are below exposure level maximums. Volatile organic compound (VOC) emissions are extensive from fires, but the levels were less than from an evaporating crude oil spill. Over 140 compounds were identified and quantified. Carbonyls, including aldehydes and ketones, were found to be below human health concern levels.

Emission data from over 45 experimental burns have been used to develop prediction equations for over 150 specific compounds or emission categories. These are used to calculate safe distances and levels of concern for a standard burn size of 500 square meters, an amount that would typically be contained in a boom. The safe distance for a crude oil burn of this size is about 500 m and for a diesel burn, much further.

Introduction

In situ burning of oil spills has been tried over 30 years ago, but is only being slowly accepted as an oil spill cleanup option—largely because of the lack of understanding of the combustion products and the principles governing the combustibility of oil-on-water. Extensive research has recently been conducted to understand the many facets of burning oil. A consortium of several agencies in the United States and Canada has joined forces to study burning and to conduct large-scale experiments. This effort has resulted in data that should lead to broader

acceptance of *in situ* burning as an acceptable spill countermeasures alternative.

Burning has distinct advantages over other countermeasures. First and foremost, it offers the potential to rapidly remove large quantities of oil. *In situ* burning has the potential to remove as much oil in 1 day as several mechanical devices could in 1 month. Application of *in situ* burning could prevent a large amount of shoreline contamination and damage to biota by removing oil before it spreads and moves to other areas. Secondly, *in situ* burning requires minimal equipment and much less labor than any other technique. It can be applied in areas where other methods cannot be used because of distances and lack of infrastructure. Thirdly, burning of oil is a final solution compared to mechanical recovery. When oil is recovered mechanically it still has to be transported, stored, and disposed of. Fourth and finally, burning may be the only option available in certain situations. Oil with ice and on ice are examples of situations where practical alternatives to burning do not exist.

There are disadvantages to burning. The first and most visible disadvantage is the large black smoke plume that burning oil produces. The second disadvantage is that the oil must be a minimum thickness to burn. Thin slicks will not burn. Oil rapidly spreads, forming thinner and thinner slicks. Most oils will spread rapidly and, in time, will be far thinner than their minimum burning thickness. An approach to dealing with such oils is to concentrate them into thicker slicks using fire-resistant booms and then burn them. This has the obvious disadvantage of requiring boats, booms, people, and time. It should be noted, however, that concentrating oil for burning requires less equipment than does collecting oil using skimmers.

The concern over atmospheric emissions remains the biggest barrier to the widespread use of burning. Extensive studies on emissions have been conducted recently. Unfortunately, burning of all kinds is often a questionable process because of concern over combustion by-products. Analysis is still difficult, although technology does permit analysis of key compounds and comparison to ambient levels of pollution.

Emissions include the smoke plume, particulate matter precipitating from the smoke plume, combustion gases, unburned hydrocarbons, organic compounds produced during the burning process, and the residue left at the burning pool site. Soot particles, although consisting largely of carbon particles, have a

variety of chemicals absorbed and adsorbed. Complete analysis of the emissions from a burn involves measuring all of these components.

Measurement of emissions

Extensive measurement of burn emissions began in 1991 with the instrumentation of several burns conducted at Mobile, Alabama to measure various physical facets of oil burning (Fingas *et al.*, 1993). Analysis of the data from these burns showed several interesting facts and several data gaps. Monitoring of burns continued for several years. In 1992, two further series of burns were monitored for emissions (Booher and Janke, 1997; Fingas *et al.*, 1993). In 1993, two major burns were conducted at sea specifically to measure emissions, but many other measurements were taken as well (Fingas *et al.*, 1994a, b, 1995a, b). Further tests were conducted in 1994, 1997, and 1998 (Fingas *et al.*, 1996a, b, 1998, 1999a, 2000; Lambert *et al.*, 1998). These tests and numbers of fires monitored are summarized in Table 1.

The emissions monitored at these burns were intended to be comprehensive and used the best field samplers or instrumentation available at the time. Measurement techniques, however, have progressed through the years and are continually improving. Measurements taken are summarized in Table 2. The emphasis on sampling has been the air emissions at ground level. This is the primary concern and also the regulated value. This paper will focus on the same measurements.

Volatile organic compounds (VOCs) were sampled using sorption tubes (in 1991 and 1992) and by taking whole air samples. Whole air samples were also taken using 6-L pre-evacuated (to 0.05 mm Hg) stainless steel canisters (Summa canisters, Scientific Instrumentation, Moscow, Idaho). A fixed orifice with an integral stainless steel frit was used to restrict the flow to about 200 mL/min, which was precalculated to fill during the time of one burn. Upwind and background samples were always taken. Analysis was by GC-MS. Over 150 compounds were measured by 1997 and over 90 hydrocarbons identified in the vapors from an evaporating or burning slick.

Table 1. Summary of studies used to measure *in situ* burn emissions.

Location	Year	Burns (#)	Monitored (#)	Oil type	Prime purpose	Burn area range (m ²)	Time of burns (min)*	Instruments (#)*	Target compounds (#)*
Mobile	1991	14	14	Louisiana crude	Physics	37–231	20–60	30	70
Mobile	1992	6	6	Louisiana crude	Physics	36–231	20–60	30	70
Calgary	1992	20	3	Crude, diesel	Emissions	37	20–70	25	40
Newfoundland	1993	2	2	Crude (ASMB)	Emissions	467–600	60–90	200	400
Mobile	1994	3	3	Diesel	Physics	199–231	60–80	95	400
Mobile	1997	9	8	Diesel	Boom tests	25	60	95	400
Mobile	1998	12	12	Diesel	Boom tests	25	60	67	400
	Total	66	48						

* Values are approximate or rounded off.

Table 2. Samples taken and target emission measurements.

Sample taken	Sampler	Measurement parameter	Secondary parameters	Additional parameters
Soot at ground level	High volume sampler	Dioxins and dibenzofurans	Particulates	PAHs
	High volume sampler	Sized particulates (PM-10, PM-2.5)		PAHs
	Sampling pump	PAHs	Particulates	
	RAM, DataRam	Particulates		
	Cascade sampler	Particle size	PAHs	
Soot in smoke plume	Sampling pump low volume	PAHs	Particulates	Metals
	Blimp, remote-controlled		Particulates	PAHs
	helicopter, research aircraft			
Air at ground level	Summa canister	VOCs	CO ₂ , CO ₁ , NO _x	
	Sampling pump low volume	VOCs		
	CO ₂ meter	Carbon dioxide		
	SO ₂ meter	Sulphur dioxide		
	NO ₂ meter	Nitrogen dioxide		
	CO meter	Carbon monoxide		
	SO ₂ impinger	Sulphur dioxide in acid form		
	DNPH cartridge	Carbonyls		
	Long-path IR	VOCs	CO ₂ , CO	
Oil		PAHs	Metals	Full analysis
Burn residue		PAHs	Metals, toxicity	Full analysis
Water under burn		PAHs	Organics	Toxicity

Carbonyls were sampled by using a Gilian 513A to pump air through a DNPH (2,4-dinitrophenylhydrazine)-silica cartridge attached via a Tygon tube. The cartridge contains 350 mg of silica coated with 1.0 mg of DNPH. The sample was subsequently analyzed using HPLC.

Polycyclic aromatic hydrocarbons (PAHs) were sampled in air using sorbent tubes initially, but later from particulates collected on high volume samplers. Analysis of PAHs was also conducted from various particulate sampling including fractionation samplers, PM-10, PM-2.5 or cascade samplers, and filters from low- and medium-volume pumps. Analysis was by standard methods using GC-MS.

Heavy metals on soot were collected using Gilian personal samplers on a 37-mm, 0.8- μ m cellulose ester (MCE) filter. Analysis was by ICP using standard methods.

Polychlorinated dibenzo-p-dioxin/furan (DX/DF) was measured on particulate samples. High volume samplers (PS-1, General Metal Works) were employed to collect cumulative samples at upwind and downwind locations. Sampling media were 10-cm glass-fiber filters followed by a 5×7.5 cm polyurethane foam plug (PUF). Flow rate was nominally at 200 L/m. These same samples were used to measure TSP, or total suspended particulate levels, and sometimes were analyzed for PAHs, other organics or metals.

PM-10 particulate air sampling was performed using a General Metal Works model PS-1 instrument. The sampling media consisted of a 3"-diameter quartz fiber filter. Some of these filters were also analyzed for PAHs.

PM-2.5 particulate air sampling was accomplished using a Partisol PM-2.5 sampler (Rupprecht & Patashnick, Albany, New York). A tared 47 mm Teflon filter was placed in the apparatus and used to collect sample.

Real-time particulate measurements were taken with RAMs or DataRAMs. The MIE Ram-1 instrument was used to perform real-time aerosol monitoring and measure relative concentrations of airborne particulates. This instrument responds to a physical particle size of 0.1 to 30 microns. The DataRAM (MIE Inc., Bedford, Massachusetts) is an updated version of the RAM. The advantage of this unit over the RAM is its internal data logging and processing capabilities. The apparatus is capable of employing several different sampling head configurations. These are total particulate, the 0–10 μ m particulate fractions or the 0–2.5 μ m particulate fractions.

Sulphur dioxide in gaseous form was measured using the Biosystems Cannonball at a flow rate of 1 L/min. This data was logged electronically. Sulfur dioxide in acid form was measured using a Gilian pump and a sodium hydroxide-filled impinger. After each burn, the impinger fluid is titrated to determine the amount of sulfuric acid/dioxide.

Carbon dioxide was measured using two electronic instruments, the Metrosonics AQ501 and the Armstrong CD-1 carbon dioxide analyzer. All these data were recorded at intervals of 1-minute averages of 10-second measurement intervals. Carbon dioxide also was measured in some Summa grab samples using gas chromatography. The Metrosonics instrument also measures carbon monoxide, moisture, and temperature.

Nitric oxides were measured using the Biosystems Cannonball. Electronic output data was recorded in manner similar to the above.

Results

Summarized data appear in the references. These data are too extensive even to provide encapsulating summaries; however, qualitative statements will be made regarding these.

Particulates. All burns, especially those of diesel fuel, produced an abundance of particulate matter. The concentrations of particulates from diesel at the same distances were approximately 4 times that for similar-sized crude oil burns. PM-10 concentrations were sometimes about 0.7 of the TSP concentration, as would be expected, but sometimes were the same as the TSP. The same is true of the PM-2.5 concentrations. This may be indicative that most material is of PM-2.5 category or that the sampling units break the particles into smaller ones. Table 3 shows the typical particulate levels of burns of crude and diesel fuel for a medium size boom area of 250 m². These values are calculated using three-way regression analysis from all the data from previous burn measurements as shown in Table 3.

PAHs. Crude oil burns result in PAH downwind of the fire, but the concentration on the particulate matter is often an order-of-magnitude less the concentration in the starting oil. Diesel contains low levels of PAHs with smaller molecular size, but results in more PAHs of larger molecular sizes. Larger PAHs are either created or concentrated by the fire. Larger PAHs, some of which are not even detectable in the diesel fuel, are found both in the soot and in the residue (Wang *et al.*, 1999). The concentrations of these larger PAHs are, however, low and often just above detection limits. Overall, the fires destroy PAHs. Table 4 shows the averaged calculated values of PAHs from crude and diesel burns for a burn area of 250 m².

VOCs. One hundred and forty-eight VOCs were measured from samples taken in Summa canisters and some on carbon absorption tubes. The concentrations of VOCs are about the same in a crude or diesel burn. Concentrations appear to be under human health limits even at the closest monitoring station. VOC concentrations are about three times higher when the oil is not burning and is just evaporating. Unfortunately, this is difficult to measure at all burns.

Dioxins and dibenzofurans. Particulates precipitated downwind and oil residue were analyzed for dioxins and dibenzofurans. The levels of these toxic compounds were at background levels indicating no production by either crude or diesel fires.

Carbonyls. Oil burns produce low amounts of the small aldehydes (formaldehyde, acetaldehyde, etc.) and ketones (acetone, etc.). These would not be a health concern, even close to the source fire. Carbonyls from crude oil fires are at very low concentrations. Table 5 shows the typical carbonyl levels from crude and diesel burns for a burn area of 250 m².

Carbon dioxide. Carbon dioxide is the end result of combustion and is found in increased concentrations around a

Table 3. Particulates from fires.

Particulate type	Crude oil burns		Diesel burns	
	Concentration (mg/m ³)	Safe distance (m)	Concentration (mg/m ³)	Safe distance (m)
Total particulates	0.005	100	0.05	370
PM-10	0.02	200	0.21	800
PM-2.5	0.004	200	0.04	530

Note: Calculated by regression from empirical results for a burn area of 250 m².

Table 4. PAHs on soot.

PAH compound	Crude oil burns		Diesel burns	
	Concentration ($\mu\text{g}/\text{m}^3$)	Safe distance (m)	Concentration ($\mu\text{g}/\text{m}^3$)	Safe distance (m)
1-Methylnaphthalene	< 1	0	< 1	0
1-Methylphenanthrene	< 1	0	< 1	40
2,3,5-Trimethylnaphthalene	< 1	0	< 1	10
2,6-Dimethylnaphthalene	< 1	0	< 1	0
2-Methylnaphthalene	< 1	0	< 1	0
Acenaphthene	< 1	0	< 1	10
Acenaphthylene	< 1	0	< 1	70
Anthracene	< 1	10	< 1	110
Benz(a)anthracene	< 1	0	< 1	20
Benzo(a)pyrene	< 1	30	< 1	30
Benzo(b)fluoranthene	< 1	0	< 1	40
Benzo(k)fluoranthene			< 1	0
Benzo(e)pyrene	< 1	0	< 1	10
Benzo(g,h,i)perylene	< 1	20	< 1	50
Biphenyl	< 1	0	< 1	20
Chrysene	< 1	0	< 1	20
Dibenz(a,h)anthracene	< 1	0	< 1	30
Dimethylnaphthalene	< 1	0	< 1	0
Fluoranthene	< 1	70	< 1	90
Fluorene	< 1	0	< 1	0
Idenol(1,2,3-cd)pyrene	< 1	0	< 1	10
Methylphenanthrene	< 1	0	< 1	10
Naphthalene	< 1	0	< 1	0
Perylene	< 1	0	< 1	0
Phenanthrene	< 1	100	< 1	90
Pyrene	< 1	30	< 1	90
Trimethylnaphthalene	< 1	0	< 1	0

Note: Calculated by regression from empirical results for a burn area of 250 m².

Table 5. Carbonyls from fires.

Compound	Crude oil burns		Diesel burns	
	Concentration ($\mu\text{g}/\text{m}^3$)	Safe distance (m)	Concentration ($\mu\text{g}/\text{m}^3$)	Safe distance (m)
Acetaldehyde	< 1	0	< 1	0
Acetone	< 1	0	5	0
2-butanone	< 1	0	103	0
Butyraldehyde	< 1	0	< 1	0
Formaldehyde	< 1	0	5	0
Propionaldehyde	< 1	0	0	0

Note: A safe distance of 0 implies that the compound is of little concern.

burn. Normal atmospheric levels are about 300 ppm and levels near a burn can be around 500 ppm. There is no human danger at this level. The three-dimensional distributions of carbon dioxide around a burn have been measured. Concentrations of carbon dioxide are highest at the 1-m level and fall to background levels at the 4-m level. Concentrations at ground level are as high as 10 times that of the plume. Distribution along the ground is broader than for particulates. Carbon dioxide emissions are not of human health concern.

Carbon monoxide. Carbon monoxide levels are usually at or below the lowest detection levels of the instruments and thus do not pose any hazard to humans. The gas has only been measured

when the burn appears to be inefficient, such as when water is sprayed into the fire. Carbon monoxide appears to be distributed in the same way as carbon dioxide. Carbon monoxide appears to be of little concern with respect to human health.

Sulfur dioxide. Sulfur dioxide, *per se*, is usually not detected at significant levels or sometimes not even at measurable levels. Sulfuric acid, or sulfur dioxide that has reacted with water, is detected at fires and levels, although not of concern, appear to correspond to the sulfur contents of the oil.

Other gases. Attempts were made to measure oxides of nitrogen and other fixed gases. None were measured in about ten experiments.

Data synthesis and calculation

Sufficient data are now available to assemble these and then correlate the results with spatial and burn parameters. The extensive work is described in the literature (Fingas and Punt, 2000; Fingas *et al.*, 1999b). Many correlations were tried, however, it was found that atmospheric emissions correlated relatively well with distance from the fire and the area of the fire. This fact then was used to develop prediction equations for each pollutant, using the data gathered from the 40 experiments conducted to date. Sufficient data were available to calculate estimation equations for over 150 individual compounds and for all the major groups. The data are, however, in some cases, insufficient to yield high correlation coefficients and low errors. The result of the correlation will provide several significant advances in the understanding of *in situ* burning: assessment of the importance of specific emissions and classes; capability of predicting a "safe" distance; and capability of predicting concentrations at a given point. Predictions from this type of empirical evaluation are far more accurate than that from models because they are based solely on actual data.

These calculations confirm that the greatest concern lies with the particulate matter, secondly with the PAHs on the particulate matter, and next with the total VOCs. Manipulation of the VOCs shows these to be close to being a matter of concern; however, it should be noted that the level of VOCs is much higher (as much as three as measured in some tests) when oil is evaporating in the absence of burning than the level of VOCs emitted when burning. There is no concern for fixed gases such as carbon dioxide, and carbon monoxide.

Overall findings

The measurement of emissions and calculation using equations developed from emission data, have revealed several facts about the fate, behavior, and quantity of the basic emissions from burning.

Particulate matter/soot. Particulate matter at ground level is a matter of concern (greater than occupational health criteria values) close to the fire and under the plume. The concentration of particulates in the smoke plume may not be a concern past about 500 meters for crude oil burns. The level of respirable particulates, those which have a size less than 10 μm , or 2.5 μm is not understood well, but follows the trends noted for TSP. Diesel fuel burns result in significantly more soot production and safe distances are much further.

PAHs. *In situ* oil fires do not produce additional polyaromatic hydrocarbons. Oils contain significant quantities of PAHs. These are largely destroyed in combustion. The PAH concentrations in the smoke, both in the plume and the particulate precipitation at ground level, are much less than the starting oil. The burn residue does, however, show a slight increase in the concentration of multi-ringed PAHs. However, when considering the mass balance of the burn, the fire destroys most of the 5- and 6-ringed PAHs.

Gases. Combustion gases, such as carbon dioxide and carbon monoxide, are significantly under any concern level.

VOCs. Fires emit many volatile organic compounds; however, in lesser quantity than when the oil is not burning. VOCs are not a concern, but can rise to close to concern levels very near a fire (<100 m).

Carbonyls. Oil fires create carbonyls, such as aldehydes and ketones, but do not exceed health exposure limits even very near fires. Overall, emissions are now understood to the extent that fires of various sizes and types can be evaluated for emission

levels and safe distances. A standard crude oil fire would not exceed exposure limits for emissions beyond about 500 m.

Biography

Mervin F. Fingas is Chief of the Emergencies Science Division in Environment Canada. Dr. Fingas' specialty is research in the analysis and behavior of oil spills in the environment. He manages 20 other scientists and staff studying various aspects of oil and chemical spills. He has devoted the last 27 years of his life to spill research and has over 450 papers and publications in the field.

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