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Full Length Article

## Effect of pool fire scale of heavy fuel oil on the characteristics of PAH emissions



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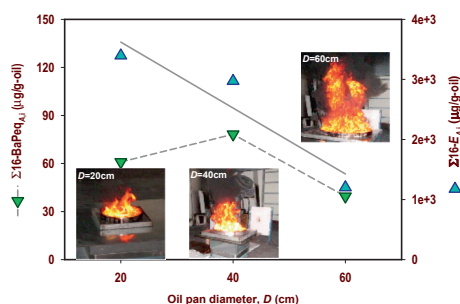
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### GRAPHICAL ABSTRACT



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### ABSTRACT

A series of pilot-scale pool test fires simulated an idealized fire accident in a storage tank containing low-sulfur No. 6 heavy fuel oil (LSFO6). This work demonstrates that the combustion characteristics of LSFO6 and shows that fire-related emission factors (EFs) of 16 USEPA PAHs during pool fires very much depend on the scale of oil pan diameter ( $D$ ). Over the range studied ( $D = 20\text{--}60$  cm), the combustion characteristics, (i) fuel mass loss rate, (ii) heat flux and (iii) ratio of  $CO/CO_2$  as time-weighted averages have positive linear relationships with  $D$ , but the combustion efficiency is a negative linear function of  $D$ . The sum of 16 PAH EFs has a negative linear function of  $D$ , while the overall vapor-particulate partitioning coefficient of 16 PAHs has a positive linear function of  $D$ . Most benzo(a)pyrene-toxic equivalents (BaPeq) for a given pool scale arise from high-molecular-weight PAHs. However, the maximum total BaPeq occurs at pool fire scale of  $D = 40$  cm. The scale of burning causes no significant variation in PAH source identifications in terms of the defined molecular diagnostic ratios.

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Further researches on large-scale pool test fires are needed to validate whether the applicability of pilot-scale studies will be acceptable to the relevant oil tank fires in industries.

## 1. Introduction

Heavy fuel oils (HFOs), the residue from the refining process of crude oil, are complex mixtures of high molecular weight compounds. Detailed information on the chemical composition of HFOs is limited due to the variety of crude oil sources and refinery processes. However, the typical compositions can include asphaltenes, naphthalene aromatics, polycyclic aromatic hydrocarbons (PAHs), aromatics, carbon residue, nitrogen, sulfur, ash and trace metals such as vanadium, nickel, iron, potassium, sodium, aluminum and silicon [1]. Thus, HFOs are generally thought to be more polluting fuels and they have been classified by International Agency for Research on Cancer (IARC) as a Group 2B carcinogen, meaning that they are possibly carcinogenic to humans [2]. Nevertheless, they are widely used and desirable as a fuel source to provide the energy needed for many industrial processes because of the relatively lower cost compared to distillate fuels on the market supply for energy demand. In 2016, the annual domestic consumption of fuel oils in Taiwan was 2.01 billion litre oil equivalent which accounted for nearly 2% of Taiwan's total energy use [3]. However, one of the risk concerns over the use HFOs can be linked to the potential negative effects on the atmosphere and the community health that would result from storage oil tank fires of fuel oils [4]. Such accidents are quite frequent in oil depots [5,6]. Accidental fires involving fuel oils represent a particular type of open burning. The emission factor of pollutants produced from open burning of fuel oils is higher than that from controlled burning because of the low combustion efficiency and the discharge of untreated pollutants [7–11]. The probable toxic chemicals produced from open burning of fuel oils include NO<sub>x</sub>, SO<sub>x</sub>, PAHs, volatile organic compounds (VOCs), particulate matter (PM), particulate trace metals, and persistent organic pollutants (POPs) [10,12]. Among these produced pollutants, PAHs are one of the most critical groups of chemicals because of a variety of potentially toxic and mutagenic impacts on both human and environment [13–17].

The kinetic pathways of PAH synthesis in the burning process are very complex and questions still remain [18–21]. However, an understanding is an essential aspect of anthropogenic production of PAHs from oil fires. In general, the emission yields of both PAHs and CO increase with the decrease of excess air ratio [22], although there is a large variation in total PAHs yields with different types of materials burned in full-scale test fires [11]. According to the results of crude oil test fires with different pan size, it should be noted that both the emission ratio of monoxide to carbon dioxide (CO/CO<sub>2</sub>) and the smoke yield increase with the pan diameter [23,24]. Meanwhile, the PAH compounds formed and their distribution profiles during incomplete combustion of fuel oils are largely dependent on combustion conditions [9,25]. Pool burning tests of fuel oils are frequently conducted using oil pans to simulate realistically scenarios of accidental fires at oil storage tanks [23,26,27]. The dynamic behaviour of pool fires is commonly characterized using the parameters measured during the burning test such as burning rate, heat release rate, flame height, flame temperature, smoke emission and the emission ratio of CO/CO<sub>2</sub> [26]. Factors which affect these measurable quantities of liquid fuels in pool fires include: pool diameter, pool depth, pool lip, open wind speed and fuel type [26,28], in which the pool diameter is the most significant parameter to provide an explicit link with the pool fire behavior. The smoke yield and the emission ratio of CO/CO<sub>2</sub> are generally recognized as the indicators of incomplete combustion where the higher both measured values will give the poorer combustion efficiency. In addition, the early, but well-known analysis of Hottel's compiled from diffusive burning data shows that pool fire mode can be categorized as two regimes in

terms of heat-transfer mechanism: the convectively controlled burning for pool diameter of  $\leq 20$  cm as well as the radiatively controlled burning for pool diameter of  $\geq 20$  cm [29].

Based on this literature review, it may be presumed that PAH emission characteristics of liquid fuels during uncontrolled fires may vary very much depend on the scale of the fire. However, when compared with other sources of PAH emissions from open burning of biomass and anthropogenic fuels [10,14,30], few studies of the emission characteristics of PAHs from *in-situ* burning of HFOs have been systematically undertaken. Lack of critical information about the emission characterization of target pollutants is difficult to develop potential solutions and identify the environmental liability of a polluting party which, in turn, invariably leads to public nuisance disputes [31,32]. Therefore, this study aims to explore the relationships between PAH emission factors from HFOs and pool fire size by conducting a series of pilot-scale pool test fires. Here the potential health impacts of PAH emissions related to burning scale are assessed in terms of equivalent toxicity as benzo[a]pyrene (BaP) and the source finger printing of HFOs are identified by the burn-related PAH molecular indices.

## 2. Materials and methods

### 2.1. Pool fire experiments

Low-sulfur No. 6 heavy fuel oil (LSFO6) with CAS No.: 68553-00-4 obtained from the Chinese Petroleum Corporation of Taiwan was employed as the selected test specimen since it is widely used in industry. Pool test fires using three different sized stainless-steel circular pans with diameters of 20, 40 and 60 cm (all with the same pan height of 6 cm filled with the test specimen) were lit under a cone calorimeter in a full-scale ISO 9705 test facility. During the burning process, the oil pan was placed in a water tray to minimize the effect of heat conduction through the surrounding rim of pool pan and an online load cell was located under the oil pan to monitor fuel oil weight loss. The heat flux,  $Q_F$  (kW/m<sup>2</sup>), was simultaneously measured using a cone calorimeter, while the gas concentrations of carbon monoxide and carbon dioxide in the exhaust stream were determined using a flue gas analyzer probe [33]. A K-type thermometer was used to determine the flame temperature,  $T_f$  (°C), 20 cm above the oil surface, in the centre of oil pan [27]. The cone calorimeter, the gas analyser and the thermometer were connected to a personal computer based data acquisition system for recording online data during a period of pool test fires. The flame height was determined by analysing the flame images captured with a digital video camera.

### 2.2. PAHs sampling

The combustion stream was collected in an exhaust hood using the sampling system of XC-572 (Apex Instruments Inc., USA). In this work, the 16 PAHs on the USEPA priority pollutant list were determined, i.e., Naphthalene (Nap), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pyr), Benzo[a]anthracene (BaA), Chrysene (Chr), Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Benzo[a]pyrene (BaP), Indeno[1,2,3-cd]pyrene (IcP), Dibenz[a,h]anthracene (DhA), and Benzo[ghi]perylene (BgP). The exhaust materials from the LSFO6 pool fires collected at the stack consist of both vapor and particulate phases. The sampling and analysis of PAH emissions from the stack were determined with the California Air Resources Board (CARB) method 429 [34]. PAHs from the vapor phase were adsorbed on XAD-2

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