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## Effects of oil pipeline explosion on ambient particulate matter and their associated polycyclic aromatic hydrocarbons

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## ABSTRACT

Effects of the oil pipeline explosion on PM<sub>2.5</sub>-associated polycyclic aromatic hydrocarbons (PAHs) and their substituted (alkylated, nitrated, oxygenated, hydroxyl and chlorinated) derivatives are assessed near the accident scene of Qingdao, China. Compared with those in TSP-PM<sub>2.5</sub>, gaseous phase, burn residue and unburned crude oil, eighty-nine PAHs in PM<sub>2.5</sub> are identified and quantified to investigate the composition, temporal and spatial distribution, and sources. The concentrations of PM<sub>2.5</sub>-associated parent PAHs increase approximately seven times from the non-explosion samples to the explosion samples (mean  $\pm$  standard deviation:  $112 \pm 2$  vs  $764 \pm 15$  ng/m<sup>3</sup>), while some substituted products (nitro- and oxy-) increase by two orders of magnitude ( $3117 \pm 156$  pg/m<sup>3</sup> vs  $740 \pm 37$  ng/m<sup>3</sup>). The toxicity evaluation indicates the BaP equivalent concentrations (based on the US EPA's toxicity factors) in PM<sub>2.5</sub> are much higher than those in the other phases, especially for a long duration after the tragic accident.

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## 1. Introduction

A catastrophic explosion of underground oil pipeline in Qingdao (Shandong Province, P.R. China) on 22 November 2013 resulted in crude oil leak and massive smoke, which raised toxic gases and particulate matter (PM) levels rapidly (Kakareka et al., 2005; Nwaichi et al., 2013). In the past few years, PM<sub>2.5</sub> (particulate matter below 2.5  $\mu$ m, fine fraction) has become a major environmental and ecological concern because of its adverse effects to human health, long range transfer, and low sedimentation rate (Chen et al., 2014). The adverse effects are thought to depend not only on the micron scale, but also on some toxic, carcinogenic and mutagenic organic compounds, such as polycyclic aromatic hydrocarbons (PAHs), sorbed onto the PM<sub>2.5</sub> surface (Masiol et al., 2012; Wang et al., 2008; Fang et al., 2013). The compositions and sources of PAHs in PM<sub>2.5</sub> or other PM and gaseous compounds have

been extensively studied (Kim et al., 2012; Gurley et al., 2013). Recently, increasing attention has been paid to the substituted products, which can be related to more serious health outcomes (Ventura et al., 2012; Manzano et al., 2013; Adetona et al., 2013). However, to our best knowledge, the studies on PM<sub>2.5</sub>-associated PAHs and their substituted derivatives due to explosion are limited, in respect that the opportunity to study air quality as the result of a tragic accident is rare.

PAHs, which formed during the incomplete combustion, have been widely accepted as a group of ubiquitous chemicals in previous studies on air pollution (Fujima et al., 2006; Yuan et al., 2014). PAHs associated with PM<sub>2.5</sub> can effectively penetrate into the respiratory system and be associated with systemic inflammation and many diseases (Chen and Schwartz, 2009; Kam et al., 2013). Thus, it is necessary to analyze the abundance, speciation, distributions, and potential sources of PM<sub>2.5</sub>-associated PAHs in order to efficiently control air pollution caused by particulates. In addition, the impact of PAHs is of concern due to their transport. PAHs are easily transported over long distances by wind, reacting with other pollutants or atmospheric gases (N<sub>x</sub>O<sub>x</sub>, SO<sub>x</sub>, Cl<sub>2</sub>, OH, and O<sub>3</sub>) or may undergo photolysis (Djordjevic et al., 2013; Wei et al., 2012). Therefore, these chemical reactions contribute to the degradation of parent PAHs in the atmosphere, nevertheless, to form more toxic

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substituted compounds such as nitro-, oxy-, chloro- or hydroxy-PAHs derivatives (Gogou et al., 1998; Ohura et al., 2008; Zimmermann et al., 2012). These substituted derivatives could also be emitted from incomplete combustion processes such as fossil fuel and biomass burning (Fujima et al., 2006; Kawanaka et al., 2007). Moreover, alkylated PAHs have been shown to contain higher concentrations and to be more toxic than parent PAHs in airborne samples (Zhao et al., 2014). This toxicological information stresses the importance of comprehensive analysis of not just the parent PAHs but, rather, their substituted products.

The objectives of this study were to determine the concentrations, compositions, temporal and spatial distributions, and sources of parent PAHs and their substituted products in PM<sub>2.5</sub> near the accident scene in Qingdao. Similarities of PAHs profiles among sampling sites and among five phases: PM<sub>2.5</sub>, TSP-PM<sub>2.5</sub> (total suspended particulates with 100  $\mu\text{m}$  > aerodynamic diameter > 2.5  $\mu\text{m}$ , coarse fraction), gaseous phase, burn residue, and unburned crude oil were compared, which indicated that remarkable differences in PAHs composition exist. Diagnostic ratios and factor analysis displayed the characterizations and sources of the explosion and non-explosion samples. Spatial and temporal variation evaluated the role of distance, time, meteorological condition and particle size in influencing the concentrations, profiles and toxicities of the measured PAHs. Their BaP equivalent concentrations (BaPeq, based on the US EPA's toxicity factors) were

calculated and compared to enable an understanding of environmental toxic effects of the above merging contaminants.

## 2. Material and methods

### 2.1. Background information of accident

Over 62 people were killed and 166 others injured in the explosion – one of the deadliest industrial accidents in China last year. On the 22 of November 2013, on the Zhoushandao Road and Zhaitangdao Road of the city's Huangdao District, the leaking underground pipeline caught fire and exploded at about 10:30. The incident happened as workers were repairing the pipeline, which had started to leak oil at around 3:00. The leaking branch line (the 176-km principle pipeline links oil depots in Qingdao with Dongying), which belongs to China Petrochemical Corporation (Sinopec Group, the country's largest oil refiner), links to several petrochemical plants. The company shut the pipeline down, but oil that leaked from the pipeline covered a ground area of about 1000 square meters and even flowed into the sea through a sewer pipe. Two containment booms were put up to prevent the spill from spreading. After the explosion Friday morning, dust and smoke clouded the air, making it hard to see much of these streets (Fig. S1). The fire produced enormous volumes of soot and burned oil residue, and was put out at about 13:00.



**Fig. 1.** Locations of the explosion point and the thirty-five sampling sites. 1, 2, 7, and 35 are campus sites, 3 and 4 are tunnel sites, 5 and 18 are port sites, 6, 28 and 29 are residential sites, 8–17 and 19–24 are roadside sites, 25 and 26 are ferry sites, 27 is a chemical plant site, 30–32 are coast sites, and 33 and 34 are bay bridge sites. The measured ambient PAHs concentrations were classified as explosion, affected and non-explosion samples based on the additional PAHs released from the burning of crude oil: 1, 2, 7 and 33–35 are the non-explosion sites, 3–6 and 29–32 are the affected sites, and 8–28 are the explosion sites.

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