



Emission and profile characteristic of volatile organic compounds emitted from coke production, iron smelt, heating station and power plant in Liaoning Province, China



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HIGHLIGHTS

- 107 kinds of VOC species were analyzed for four types of stationary sources.
- VOC source profiles were established based on dilution sampling of stacks.
- OH loss rates of NMHCs were assessed.

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ABSTRACT

107 kinds of C₂–C₁₂ volatile organic compound (VOC) mass concentrations and profiles for four types of coal-fired stationary sources in Liaoning Province were studied by a dilution sampling system and GC–MS analysis method, which are of significant importance with regard to VOC emissions in northeast of China. The results showed that there were some differences among these VOC source profiles. The total mass concentrations of analyzed 107 VOC species varied from 10,917 to 19,652 μg m⁻³. Halogenated hydrocarbons exhibited higher mass percentages for the VOC source profiles of iron smelt (48.8%) and coke production plant (37.7%). Aromatic hydrocarbons were the most abundant in heating station plant (69.1%). Ketones, alcohols and acetates held 45.0% of total VOCs in thermal power plant. For non-methane hydrocarbons (NMHCs), which are demanded for photochemical assessment in the USA, toluene and n-hexane were the most abundant species in the iron smelt, coke production and thermal power plant, with the mass percentages of 64.8%, 52.7% and 38.6%, respectively. Trimethylbenzene, n-propylbenzene and o,m-ethyltoluene approximately accounted for 70.0% in heating station plant. NMHCs emitted from coke production, iron smelt, heating station and power plant listed above presented different chemical reactivities. The average OH loss rate of NMHCs from heating station, was 4 to 5.6 times higher than that of NMHCs from iron smelt, coke production and power plant, which implies that VOCs emitted from heating station in northeast of China should be controlled firstly to avoid photochemical ozone pollution and protect human health. There are significant variations in the ratios of benzene/toluene and m, p-xylene/ethylbenzene of these coal-fired source profiles. The representativeness of the coal-fired sources studied and the VOC samples collected should be more closely examined. The accuracy of VOC source profiles related to coal-fired processes is highly dependent on location and sampling method.

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1. Introduction

Volatile organic compounds (VOCs) are main air pollutants and play an important role in the earth system (Lothawornkitkul et al., 2009). In the atmosphere, they can react with NO_x, the ozone and OH radical at the presence of sunlight to generate photochemical oxidants (Atkinson,

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2000). They can significantly affect the oxidizing capacity and radioactive balance of the atmosphere according to their influence on stratosphere ozone depletion, tropospheric photochemical ozone formation, secondary organic aerosol (SOA) formation and the enhancement of the global greenhouse effect (Song et al., 2007; Guo, 2011). On the other hand, VOCs are proven to be carcinogenic and toxic and can impact human health directly or indirectly (Srivastava, 2004; Srivastava et al., 2005; Choi et al., 2011).

Fossil fuel combustion and industrial activities are considered as the major anthropogenic sources for organic compounds in the atmosphere (Zbigniew et al., 2002; Song et al., 2007; Ribeiro et al., 2012). However, the information lack of VOC emission from coal-fired sources (Fernandez-Martinez et al., 2001; Tsai et al., 2003). Lots of investigations have been undertaken for VOC emissions from vehicle (Olson et al., 2009; Liu et al., 2010), biomass burning (Schauer et al., 2001; Evtugina et al., 2012; Jumpponen et al., 2012), solvent (Niedojadlo et al., 2007; Yuan et al., 2010; Zheng et al., 2012), petrochemical (Chen et al., 2005, 2006a) and refinery (Cetin et al., 2003; Lin et al., 2004; Chen et al., 2006b).

Several studies have attempted to discuss the profile characteristic of VOCs from coal-fired industries. For example, Fernandez-Martinez et al. (2001) studied the distribution of 16 VOCs emitted from combustion processes of five Spanish coal-fired power stations, thermal desorption technique was employed to analyze VOC concentrations in gaseous emissions (trapped onto Carbotrap B sorbent) and found that aromatic, aliphatic hydrocarbons and chlorinated compounds are mainly formed during the combustion. Moreira dos Santos et al. (2004) sampled 36 VOCs with activated charcoal sorbent tubes from the thermoelectric stack of coal-fired power stations in Brazil, and found that benzene held the highest concentration. Tsai et al. (2008) measured the 68 VOCs from an integrated iron and steel plant located in southern Taiwan, including coke making, sintering, hot forming and cold forming. Exhaust gases were sampled from the stacks and analyzed by the US EPA Method 18 integrated bag method, and it was found that toluene, 1,2,4-trimethylbenzene, isopentane, m,p-xylene, 1-butene, ethylbenzene, and benzene were the predominant VOC species in these processes. Ciaparra et al. (2009) measured the aromatic hydrocarbons within the boundary of a coking plant in the UK, using differential optical absorption spectrometry. It was found that benzene concentrations determined in the vicinity of a coke plant were comparable to some urban locations and were 20-fold lower than in refineries. Obviously, aromatic hydrocarbons were the most abundant VOCs of the coal-fired stationary sources in these studies, but the significant variations among these VOC source profiles have been found, suggesting that the representativeness of these source profiles was not wide and might result in large uncertainty in the source apportionment of VOCs and thus further mislead the policy made for air pollution control. Therefore, it was necessary to develop VOC source profiles through scientific emission measurements from coal-fired stationary sources. In the present study, source profiles of VOCs from coal-fired industries in Liaoning were investigated by examining emissions from stacks of coke production, iron smelt, heating station and power plant.

There were several publications concerning the emission of VOCs from coal-fired stationary sources, whereas the sampling methods were mostly referred to be sampling with canister within the boundary of industries (Lin et al., 2004; Chen et al., 2005; Liu et al., 2008b), or sampling at the downwind of industrial plants with canister (Cetin et al., 2003; Liu et al., 2008a) or directly sampling with Teflon bubbler and Tenax tube from stacks (Kalabokas et al., 2001; Moreira dos Santos et al., 2004). Few attempts have yet been made to examine the VOC emissions from coal-fired stationary sources based on dilution sampling method (Zhang et al., 2007). For source dilution techniques, the hot exhaust gas was diluted with clean air or inert gas prior to VOC sampling to mimic atmosphere transformation of exhausts found in a plume immediately downwind of the stacks. It should be noted that direct measurement data from individual sources did not allow for normal

dilution and cooling that occurred in a plume, and they may not reflect the variability of actual emissions over time (Yang et al., 1998; Lee et al., 2008). Therefore, the dilution sampling may collect more representative and reliable samples for stationary sources.

China's new National Ambient Air Quality Standards (GB 3095-2012) was released in 2012, which has set limit values for atmospheric fine particles (PM_{2.5}) and ozone. As the precursors for PM_{2.5} and ozone, VOCs have drawn extensive attentions in China, and few studies have focused on the constituents of VOCs in the stack exhaust from coal-fired stationary sources. The field campaign to investigate 107 VOCs emission characteristic from four types of coal-fired stationary sources including coke production, iron smelt, heating station and thermal power plant based on dilution sampling method is quite necessary and useful. The objectives of this study were to: (1) obtain the mass concentrations of VOC species from coal-fired stationary sources; (2) establish VOC sources profiles based on dilution sampling method which could be used in receptor models; (3) assess the OH loss rate of NMHCs. The research will be useful and important for source apportionment studies of VOCs in various environmental media and establishing effective pollution control measures in northern China.

2. Materials and methods

2.1. Sampling site description

VOC samples from the stacks after desulfurization and dust-removing equipments of four types of coal-fired stationary sites were collected from August to December in 2009 in Liaoning Province, the traditional heavy industrial base in Northeast China. Detailed information on sampling sites was shown in Table 1.

2.2. Dilution sampling for VOCs from stacks

A self-designed dilution sampling system was adopted. Dilution sampling schematic diagram of the stack gas was shown in Fig. 1. All parts of the sampling system in contact with exhaust gas and diluted mixture were made of stainless steel. Teflon gaskets were used for seal at each connecting fraction to reduce contamination. Polytetrafluoroethylene pipes were used to connect each stainless steel part for convenient. The sampler consists of four stages, the dilution tunnel, the clean air generator, the residence chamber and the sampling system. Detailed structures and performance assessment of the system could be found in our former study (Kong et al., 2013).

VOCs were collected from the routine monitoring holes on the stacks set by local environmental protection departments. Before the test of another different source type, the dilution tunnel was cleaned thoroughly with deionized water and alcohol, then wrapped the whole dilution tunnel with heating blankets at 150 °C for at least 8 h. Meanwhile, ultra-pure N₂ kept supplying to the dilution tunnel. The vacuumed SUMMA canisters were cleaned in three pressure/evacuating cycles with humidified zero-grade nitrogen. The whole cleaning process was fulfilled automatically in a Model 3100A Automatic Canister Purging System (Entech Instrument Inc., USA) and pumped as vacuum state (the pressure less than 50 mTorr) finally. During sampling, the sampling probe was put into a proper stack sampling point and faced the stack gas flow (Ge et al., 2004). The transfer line was insulated and heated to sustain a surface temperature which is the same to the inlet temperature to avoid a gasification and condensation of particles in emissions. Dry and particle-free pressured air was introduced into the diluter. Prior to sampling, the whole sampling system was injected with smokes at a constant flow rate of 20 L min⁻¹ for 20 min, then a 3.2 L stainless steel vacuumed SUMMA canister (Entech Instrument Inc., USA) was installed downstream of the exhaust for collecting VOCs every 1 h. After instantaneous sampling, the canisters were stored in a dry, cool and well-ventilated place until analysis within 7 days.

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