



Influence of sulfur in fuel on the properties of diffusion flame soot



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HIGHLIGHTS

- Lean flame soot was influenced more by sulfur in fuel than rich flame soot.
- Sulfate species were mainly formed on the surface of soot.
- The diameter of primary lean flame soot from high sulfur content fuel increased.
- The hygroscopicity of lean flame soot from high sulfur content fuel increased.

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ABSTRACT

Previous studies indicate that sulfur in fuel affects the hygroscopicity of soot. However, the issue of the effect of sulfur in fuel on soot properties is not fully understood. Here, the properties of soot prepared from fuel with a variable sulfur content were investigated under lean and rich flame conditions. Lean flame soot was influenced more by sulfur in fuel than rich flame soot. The majority of sulfur in fuel in lean flame was converted to gaseous SO₂, while a small fraction appeared as sulfate and bisulfate (referred to as sulfate species) in soot. As the sulfur content in fuel increased, sulfate species in lean flame soot increased nonlinearly, while sulfate species on the surface of lean flame soot increased linearly. The hygroscopicity of lean flame soot from sulfur-containing fuel was enhanced mainly due to sulfate species. Meanwhile, more alkynes were formed in lean flame. The diameter of primary lean flame soot particles increased and accumulation mode particle number concentrations of lean flame soot from sulfur-containing fuel increased as a result of more alkynes. Because the potential effects of soot particles on air pollution development greatly depend on the soot properties, which are related to both chemical aging and combustion conditions, this work will aid in understanding the impacts of soot on air quality and climate.

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1. Introduction

Soot particles, which are produced from incomplete combustion of fossil fuels and biomass, are ubiquitous in the atmosphere and comprise 10–50% of the total tropospheric particulate matter (Jacobson, 2001; Lary et al., 1999). They have an important influence on global and regional radiative balance and climate by directly absorbing solar radiation as well as by indirectly scattering

solar radiation as cloud condensation nuclei (CCN) (Chameides and Bergin, 2002; Menon et al., 2002). It has been reported that the contribution of soot to global warming may be second to that of CO₂ (Jacobson, 2001; Moffet and Prather, 2009). Soot also poses a health risk by causing and enhancing respiratory, cardiovascular and allergic diseases (Sydbom et al., 2001). In addition, when emitted into the atmosphere, soot particles are subjected to several aging processes, including adsorption or condensation of gaseous species, coagulation with other preexisting aerosols and oxidation (Han et al., 2013a, 2013b, 2012c; Lelièvre et al., 2004; Saathoff et al., 2003; Zhang and Zhang, 2005; Zhang et al., 2008). These aging processes not only significantly affect the chemical composition of the atmosphere, but also induce modification of soot properties, such as morphology, hygroscopicity, and optical properties, thus

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influencing the climatic and health effects of soot (Peng et al., 2016; Zhang et al., 2008).

Sulfur-containing compounds are widely distributed in fuels. Sulfur in fuel can be converted to toxic sulfur oxides (SO₂) through combustion that result in air pollution and poison the oxidation catalysts in emission control systems (Stanislaus et al., 2010). Although the environmental regulations on fuels sulfur content are currently becoming more and more stringent all around the world, the sulfur content in fuels remains exceedingly high in the developing countries, such as China and Brazil (http://www.dieselnet.com/standards/br/fuel_automotive.php#anp_2009, 2015; Zhang et al., 2010).

Many previous studies imply that fuel properties show a strong influence on soot properties including functional groups, morphology, and chemical composition as well as microstructure (Daly and Horn, 2009; Jansma et al., 2012; Lu et al., 2012). Therefore, sulfur-containing compounds widely distributed in fuels may affect soot properties. At present, a large number of studies have been performed on combustion of sulfur-containing fuels in diesel engines (Arnold et al., 2006, 2012; Giechaskiel et al., 2005; Jansma et al., 2012; Lu et al., 2012; Maricq et al., 2002; Olfert et al., 2007; Ronkko et al., 2013; Saiyasitpanich et al., 2005; Shi and Harrison, 1999; Weingartner et al., 1997) or diffusion flames (GÜLder and GlavinČevski, 1991). The majority of these studies have mainly showed that sulfur in fuel promoted exhaust particle formation (Arnold et al., 2006, 2012; Ronkko et al., 2013; Shi and Harrison, 1999) or emission (Saiyasitpanich et al., 2005), especially for ultrafine particles. However, the way by which sulfur in fuel promoted particle formation or emission has been not clearly understood. Meanwhile, only a few studies pay attention to the effect of sulfur in fuel on the properties of soot (Jansma et al., 2012; Lu et al., 2012; Olfert et al., 2007; Weingartner et al., 1997). For example, little is known about the effects of sulfur in fuel on soot morphology, and no experiments have been undertaken so far to investigate the quantity of sulfate adsorbed in soot. Therefore, the influence of sulfur in fuel on soot particle properties has not been clearly understood. In this study, diffusion flame soot prepared under well-controlled combustion conditions was investigated in laboratory experiments. By combining several measurements, characteristics of soot such as functional groups, composition, morphology and hygroscopicity were analyzed as a function of the sulfur content in fuel. The results will increase understanding of the effect of sulfur in fuel on soot properties and may also help assess the environmental effects of soot.

2. Experimental section

2.1. Soot production

Soot samples were produced in a co-flow homemade burner system as described in detail in our previous studies (Han et al., 2012a, 2012b, 2013a, 2013b). The co-flow burner consisted of a diffusion flame maintained in a flow of synthetic air, which was controlled by mass flow meters to regulate the fuel/oxygen ratio. The fuel was fed by a cotton wick extending into the liquid fuel reservoir. The combustion conditions were expressed as the molar ratio of the consumed fuel (measured by the mass of consumed n-hexane) to the introduced oxygen (obtained from the entrained airflow volume) during the combustion process. Thus, soot samples obtained from a relatively high fuel/oxygen ratio of about 0.16 and a relatively low fuel/oxygen ratio of about 0.12 were identified as “rich” flame soot and “lean” flame soot, respectively, as used in our previous studies (Han et al., 2013a, 2012a, 2012c).

N-hexane (AR, Sinopharm Chemical Reagent Co., Ltd.) was used as model liquid fuel, and thiophene (AR, Sinopharm Chemical

Reagent Co., Ltd.), a major sulfur-containing compound in fossil fuel (Dai et al., 2008), was added to n-hexane to vary the sulfur content in fuel as used in previous studies (Bladt et al., 2012; Dai et al., 2008; GÜLder and GlavinČevski, 1991; Hernandez-Maldonado and Yang, 2003).

2.2. Characterization of soot

The functional groups of soot samples were characterized using a Fourier transform infrared spectrometer (FT-IR, NEXUS 6700, Thermo Nicolet Instrument Corp.) equipped with a high-sensitivity mercury–cadmium–telluride (MCT) detector cooled by liquid N₂ and an ATR-IR cell. The spectra of soot were recorded (100 scans, 4 cm⁻¹ resolution) using the blank ZnSe ATR crystal as reference in the spectral range from 4000 to 650 cm⁻¹. Alkynes on soot were further identified using Lindlar catalyst, which can selectively catalyze conversion of alkynes to alkenes (Lindlar, 1952; Rajaram et al., 1983; Vile et al., 2014). A portion of soot generated from fuel containing 3420 ppm S was extracted in toluene by ultrasonication. The extract was hydrogenated in the presence of Lindlar catalyst according to the literature (Rajaram et al., 1983). Alkynes in the toluene extracts of soot from fuel containing 3420 ppm S before and after reaction were characterized in an ATR-IR cell after evaporation of toluene, as described above. Elemental analysis was performed on a Vario EL Analyzer (Elementar Analysensysteme GmbH, Hanau, Germany).

Sulfate species in soot were detected with ion chromatography (IC). A portion of soot collected on polytetrafluoroethylene membranes (PTFE, Pall corporation) was extracted in ultrapure water by ultrasonication for 30 min in a water bath, where ice was regularly added to avoid heating. Then, the extract was filtered through a nylon syringe filter (0.22 μm). The obtained sulfate solution was analyzed with IC using a Metrohm 820 IC separation center (with a Metrosep A Supp5-150 column and an 819 IC conductivity detector). An eluent of 9.0 mM Na₂CO₃ was used. An auto-sampler was used where 50 μL of each sample was introduced into the eluent, which was kept at a flow rate of 1.0 mL min⁻¹. Meanwhile, the presence of ammonium ion (NH₄⁺) in soot was also examined with IC.

Gaseous sulfur, mainly SO₂, was examined with IC. The exhaust gas containing SO₂ filtered through a PTFE membrane was directly bubbled into an adsorption solution containing excessive sodium hydroxide, and SO₂ was adsorbed as sulfite. Then, a large amount of ozone (O₃) was bubbled into adsorption solution, through which sulfite was transformed to sulfate. The obtained sulfate solution was analyzed with IC as mentioned above.

The morphology and particle size of the soot were examined using a TEM (H-7500, Hitachi). Soot produced in a diffusion flame was directly deposited onto a Cu microgrid. The acceleration voltage was set to 80 kV for the measurements. Image J 1.41 software was used to analyze the diameter of primary soot particles.

The particle number concentration and size distribution of lean flame soot were measured with a scanning mobility particle sizer (SMPS, TSI Model 3081) equipped with DMA 3080 (TSI Inc.) and CPC 3776 (TSI Inc.). An electrical Low Pressure Impactor (ELPI, Dekati Inc.) was used to control the stability of the particle emission during the SMPS scans. Before passing through the SMPS, the exhaust gas was diluted by zero air, and the dilution ratio was 7.5. The length of the sampling line from the diluter to the particle instruments was minimized. Each experiment was repeated five times and the results were found to agree with each other within the 95% confidence level.

The hygroscopicity of soot was investigated qualitatively using ATR-IR. Fresh lean flame soot particles were directly deposited on the ZnSe crystal of the ATR-IR cell, which was then sealed with a

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