



Fractional iron solubility of aerosol particles enhanced by biomass burning and ship emission in Shanghai, East China



H.B. Fu ^{a,*}, G.F. Shang ^a, J. Lin ^b, Y.J. Hu ^a, Q.Q. Hu ^a, L. Guo ^a, Y.C. Zhang ^a, J.M. Chen ^{a,*}

^a Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention, Department of Environmental Science & Engineering, Fudan University, Shanghai 200433, China

^b Key Laboratory of Nuclear Analysis Techniques, Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China

HIGHLIGHTS

- The major iron-bearing class is the aluminosilicate dust during DSE.
- The Fe-bearing aerosols are dominated by coal fly ash during NDS.
- Source-dependent composition of aerosol is a primary determinant for %Fe_s.
- %Fe_s is controlled by both biomass burning and oil ash from ship emission.

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ABSTRACT

In terms of understanding Fe mobilization from aerosol particles in East China, the PM_{2.5} particles were collected in spring at Shanghai. Combined with the backtrajectory analysis, the PM_{2.5}/PM₁₀ and Ca/Al ratios, a serious dust-storm episode (DSE) during the sampling was identified. The single-particle analysis showed that the major iron-bearing class is the aluminosilicate dust during DSE, while the Fe-bearing aerosols are dominated by coal fly ash, followed by a minority of iron oxides during the non-dust storm days (NDS). Chemical analyses of samples showed that the fractional Fe solubility (%Fe_s) is much higher during NDS than that during DSE, and a strong inverse relationship of $R^2 = 0.967$ between %Fe_s and total atmospheric iron loading were found, suggested that total Fe (Fe_T) is not controlling soluble Fe (Fe_s) during the sampling. Furthermore, no relationship between Fe_s and any of acidic species was established, suggesting that acidic process on aerosol surfaces are not involved in the trend of iron solubility. It was thus proposed that the source-dependent composition of aerosol particles is a primary determinant for %Fe_s. Specially, the Al/Fe ratio is poorly correlated ($R^2 = 0.113$) with %Fe_s, while the apparent relationship between %Fe_s and the calculated K_{BB}^+/Fe ratio ($R^2 = 0.888$) and the V/Fe ratio ($R^2 = 0.736$) were observed, reflecting that %Fe_s could be controlled by both biomass burning and oil ash from ship emission, rather than mineral particles and coal fly ash, although the latter two are the main contributors to the atmospheric Fe loading during the sampling. Such information can be useful improving our understanding on iron solubility on East China, which may further correlate with iron bioavailability to the ocean, as well as human health effects associated with exposure to fine Fe-rich particles in densely populated metropolis in China.

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1. Introduction

Iron is ubiquitous in the environment and plays a central role in many biological and chemical processes (Martin, 1990; Boyd and Ellwood, 2010). Iron is an important micronutrient, and its availability is critical in regulating primary productivity over the ocean. The supply of bioavailable Fe from atmospheric deposition can modulate CO₂ sequestration and thus influence the global carbon cycle and climate (Martin, 1990; Mahowald et al., 2009). It is generally believed that the soluble fraction of Fe is mainly considered as bioavailable for

phytoplankton (Chuang et al., 2005; Guieu et al., 2005). Given its abundance in the atmosphere, the health effects of iron have been investigated (Kim et al., 2010). Soluble iron in fine atmospheric particles has been identified as a public health concern by participating in reactions that generate reactive oxygen species (ROS) (Kim et al., 2010; Pattanaik et al., 2012). The amount of iron mobilized from particles has also been shown to correlate with measures of ROS generation (Kim et al., 2010; Pattanaik et al., 2012). Understanding iron mobilization from aerosol particles is important in achieving insight into the processes by which aerosol particles contribute to both phytoplankton productivity and health impacts (Oakes et al., 2012a).

A growing body of knowledge has emerged on various control factors that impact iron aerosols solubility (Bonnet and Guieu, 2004;

* Corresponding authors at Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention, Department of Environmental Science & Engineering, Fudan University, Shanghai 200433, China.

Paris et al., 2011; Shi et al., 2009; Li et al., 2012). Several studies have suggested that most of the soluble iron is linked to chemical processing of aerosol during transport and aging in the atmosphere. This “chemical processing” is generally thought to involve the reduction of Fe(III) to Fe(II), mediated by the presence of acidic species, sunlight, and cloudwater condensation and evaporation (Key et al., 2008; Okada et al., 2006; Paris et al., 2011; Shi et al., 2009; Cwiertyny et al., 2008; Li et al., 2012). On the other hand, the investigations conducted by Baker and Jickells (2006a) hypothesized that iron mobilization from aerosol particles is rather controlled by particle size than by chemical processes during their atmospheric transport. Oooki et al. (2009) reported a higher Fe solubility in finer aerosol collected in northern Japan during a dust event. In contrast, Buck et al. (2010) did not find an increasing trend in Fe solubility with the size of aerosol over the northwest Pacific Ocean. Shi et al. (2011) further pointed out that particle size alone cannot explain the increase in fractional iron solubility observed, suggesting that other processes may play a more dominant role or work synergistically with particle size to promote fractional iron solubility.

Several recent studies have emphasized that aerosol mineralogy, especially iron chemical from in aerosols, could be a critical factor for Fe mobilizing (Cwiertyny et al., 2008; Fu et al., 2012a; Oakes et al., 2012b). Journet et al. (2008) supposed that iron solubility can be linked to the chemical specificity of the bonds in which iron is coordinated in the different minerals. Schroth et al. (2009) have documented very high solubility of iron in oil fly ashes, in which iron speciation is mainly as ferric salts. Fu et al. (2012a) reported that the biomass burning particles yielded much higher fractions of Fe solubility relative to mineral particles because such particles hosted more labile Fe forms. Several lines of evidence from field measurements on the North Atlantic, India Ocean and East Asia supported the hypothesis that combustion-derived aerosols could be a significant source of soluble Fe (Chuang et al., 2005; Guieu et al., 2005; Trapp et al., 2010; Sholkovit et al., 2009; Sedwick et al., 2007; Kumar et al., 2010). A modeling study by Luo et al. (2008) estimated that iron particles from combustion can represent more than 50% of the total iron deposited in areas near combustion sources. Also, Fe mobilization is closely correlated with other components within the aerosols. Oakes et al. (2012a) proposed that sulfate content of iron-containing particles is an important proxy for iron solubility.

Combined field measurements and modeling for East China, typically indicated that coal combustion, industry, biomass burning, and vehicles represent the most important sources of aerosols (Moffet et al., 2012). Shanghai is the biggest city in China, and located in the eastern coast of China. In the springtime, large scale dust storms frequently occur in the western China and a large amount of dusts can be transported long distances over Shanghai (Huang et al., 2012a, 2012b). Also, biomass burning becomes increasingly important due to intensive agricultural activities in spring (Wang et al., 2009). The dusts passing through the highly populated and industrialized regions in eastern megacities of China have ample opportunity to be subject to influence of pollutants, which may enhance iron mobilization from dust particles, assuming that anthropogenic emission of SO₂ and NO_x has some effects in Fe dissolution from mineral dust (Solmon et al., 2009; Li et al., 2012).

Giving the complexity of aerosol-phase iron chemistry and the limited field data, more work should be done to refine the assessment of the solubility and source of iron in this region. We focused this study on Shanghai due to the coexistence of dust and pollution, which could be transported over the north Pacific region, identified as one of two main high-nutrient low-chlorophyll (HNLC) regions of the globe (Mahowald et al., 2009). In this paper, we reported estimates of aerosol iron solubility based on samples collected in Shanghai. Additional measurements by electron microscopes provided information regarding the chemical and physical characteristics of the single particles. These data represent valuable information for

elucidation of the source and origin of Fe-containing particles, as well as their solubility. Analysis of single iron-bearing particles combined with bulk iron solubility measurements in a variety of aerosol particles will provide unique insight to factors corresponding to iron solubility. To our knowledge, reports on identifying sources of Fe-bearing aerosols, and observing the controlled factors on Fe solubility in East China were infrequent.

2. Experimental section

2.1. Sampling

The sampling site (31.406°N, 121.285°E) is located on the rooftop of a four-storey building of Shanghai Institute of Applied Physics in Jiading district of Shanghai (Fig. S1), which is off ground about 18 m, and almost no high buildings are around this sampling site. The sampling site is about 28 km away to the northwest of downtown of Shanghai (The People's Square) and is approximately 15 km inland from the East China Sea. An incineration facility (Jiangqiao) with a capacity of 1500 t d⁻¹ is situated 10 km southeast. Baosteel factory and Jiangnan Shipyard is 15 km and 25 km east of the site, respectively. Apart from the point sources, vehicular traffic may also have affected the measurements made at the study site. There is a local busy road (Jialuo road) 300 m south of the observation site, and a more heavily trafficked road (Liuxiang road) is about 350 m east of the site. The site can be treated as representative suburb site influenced by mixture of residential, traffic, agricultural, and industrial sources, but not dominated by one of these.

Aerosol samples were collected during the period of 16th April 2011 to 16th May 2011. Two median-volume samplers (Model-2, Beijing Geological Instrument Factory, China) were used to collect PM_{2.5} in parallel on both the D90 Quartz filter (Millipore, USA) and polycarbonate membrane (0.2 μm, 90 mm, Millipore, USA). Every sample was integrated for a time-period ranging from 20 to 24 h to collect adequate aerosol mass on the filters. The airflow rate of each sampler was calibrated and controlled uniformly at 70 L min⁻¹, and the volumes were recorded by an airflow meter for quality assurance. PM_{2.5} masses were determined by gravimetry. Before and after the sampling, the polycarbonate membranes were weighed on an analytical balance with a reading precision of 10 μg (Mettler Toledo). The samples were balanced to achieve constant temperature and humidity for 24 h before weighing. All of quartz-fiber filters were pre-baked at 450 °C for 6 h before sampling to eliminate organic impurities. The relative temperature and humidity in the weighing room were 20 ± 2 °C and 40 ± 1%, respectively. The filters were then stored in individual zip-lock bags and frozen for analysis. In parallel with aerosols sampling, we estimated the contamination of the method with the use of “blank filters”. All of the procedures were strictly quality controlled to avoid any possible contamination of the samples. Typically, some particle agglomeration was observed near the center of the impaction region. Care was taken to analyze portions of the substrate that were not overloaded with particles.

For the TEM, the sampler has a collection efficiency of 100% at 0.5 μm aerodynamic diameter if the density of the particles is 2 g cm⁻³. More information about the sampler can be found elsewhere (Fu et al., 2012b). The grids contain fiber-like carbon substrates that minimize particle overlap. Sampling periods ranged between 30 and 180 s, depending on particle loading. The collected samples were put in plastic carriers, sealed, and stored in a desiccator to minimize exposure to ambient air and preserve it for further analysis.

Measurements of wind speed/direction, relative humidity, and ambient temperature were automatically recorded by a Kestral 4000 Pocket Weather Tracker (Nielsen-Kellermann Inc., USA). The trace gases of O₃, NO_x, SO₂ and PM₁₀ were friendly provided by Shanghai Meteorological Bureau. O₃ was measured by pulsed UV fluorescence (Thermo Fisher Scientific, Co., Ltd, Model 49i). NO_x was measured by a molybdenum NO₂-to-NO converter using the

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