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Short Communication

Severe haze episodes and seriously polluted fog water in Ji'nan, China

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HIGHLIGHTS

- Spatial distributions of $PM_{2.5}$ over China during server haze episodes were given.
- Fog water was seriously polluted due to longtime and large-scale haze pollution.
- Fog events reduced ambient air pollutants but difficult to cleanse the air.

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ABSTRACT

Haze episodes often hit urban cities in China recently. Here, we present several continuous haze episodes with extremely high $PM_{2.5}$ levels that occurred over several weeks in early 2013 and extended across most parts of the northern and eastern China—far exceeding the Beijing–Tianjin–Hebei region. Particularly, the haze episode covered ~1 million km^2 on January 14, 2013 and the daily averaged $PM_{2.5}$ concentration exceeded 360 μg m^{-3} in Ji'nan. The observed maximum hourly $PM_{2.5}$ concentration in urban Ji'nan reached 701 μg m^{-3} at 7:00 am (local time) in January 30. During these haze episodes, several fog events happened and the concurrent fog water was found to be seriously polluted. For the fog water collected in Ji'nan from 10:00 pm in January 14 to 11:00 am in January 15, sulfate, nitrate, and ammonium were the major ions with concentrations of 1.54×10^6, 8.98×10^5, and 1.75×10^6 μeq L^{-1}, respectively, leading to a low in-situ pH of 3.30. The sulfate content in the fog sample was more than 544 times as high as those observed in other areas. With examination of the simultaneously observed data on $PM_{2.5}$ and its chemical composition, the fog played a role in scavenging and removing fine particles from the atmosphere during haze episodes and thus was seriously contaminated. However, the effect was not sufficient to obviously cleanse air pollution and block haze episodes.

1. Introduction

Large quantities of pollutants have been emitted into the atmosphere along with rapid economic expansion in China (Tie et al., 2006). The annual average $PM_{2.5}$ concentration crept upward from approximately 60 μg m^{-3} in 1999–2000 to 90 μg m^{-3} in 2005–2006 in Shanghai (Ye et al., 2003; Feng et al., 2009). Carbonaceous materials and SO_4^{2-}, NO_3^-, and NH_4^+ are main components of $PM_{2.5}$ in haze episodes (Du et al., 2011; P. Li et al., 2011; W. Li et al., 2011; Yang et al., 2012; Sun et al., 2013). The concentrations of water-soluble ions in $PM_{2.5}$ during haze episodes can be 3.5 times higher than those observed in clear days (M. Zhang et al., 2013; Y. Zhang et al., 2013).

Carbonaceous materials and water-soluble ions in fog water are particularly interesting as they represent the result of hydrometeor interactions with ambient pollution (Herckes et al., 2006; P. Li et al., 2011; W. Li et al., 2011). From the 1980s to 1990s, fog events happened frequently in the Yangtze River Delta, the Sichuan basin, and the Gansu and Shanxi region (Wang et al., 2005; Niu et al., 2010). However, a significant decrease of fog days has been observed in the Gansu and Shanxi region (Wang et al., 2005) and precipitation at Mt. Hua in Shanxi province

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has decreased by 30% to 50% during hazy conditions (Rosenfeld et al., 2007). They suggest that the high level of fine particles in haze episodes plays an important role in effecting wet precipitation (Rosenfeld et al., 2008). While the content of pollutants in fog water is related to air quality due to the scavenging of fine particles and soluble gasses (P. Li et al., 2011; W. Li et al., 2011), little work has been reported on the fog chemistry and the capacity of fog for fine particle removal during severe haze episodes.

In January, 2013, extremely severe haze episodes appeared in northern and eastern China, which attracted broad attentions from both domestic and international communities, partly due to the serious public health damage (M. Zhang et al., 2013; Y. Zhang et al., 2013). To understand the pollution characteristics, meteorological conditions, sources, and formation mechanism of the extraordinary haze episodes, several studies have been made and published recently. From ground-based measurements, it was found that the most polluted areas were Beijing–Tianjin–Hebei region, west Shandong, and north Henan provinces, and atmospheric aerosols mostly concentrated in boundary layer below 1500 m (H. Wang et al., 2014). Secondary sulfate, nitrate and organic aerosols were the major components of $PM_{2.5}$ during the haze episodes, accounting for 65.7% of the $PM_{2.5}$ in urban Shanghai (Zhou et al., 2013). Secondary aerosols mostly formed via heterogeneous reactions on particle surfaces, which changed the particle size, hygroscopicity and optical properties, causing large negative aerosol radiative forcing efficiency at surface and accelerating the formation of haze episodes (Che et al., 2014; Y. Wang et al., 2014). Modeling studies indicate the important role of pollutant transport to the regional haze (Z. Wang et al., 2014), and identify the major emission sources from industry, domesticity, and agriculture (Wang et al., 2013). Besides emissions and transformations of air pollutants, the unusual meteorological conditions, i.e., weak southerly winds in the middle and low troposphere, high pressure at 500 hPa, and inversion in near surface, were also responsible to the severe haze episodes (Y. Wang et al., 2014; Zhang et al., 2014).

During the severe haze episodes in January, 2013, several heavy fog events simultaneously happened within this region, which provide a unique opportunity to investigate the pollution characteristics of the fog and the capacity to remove the very high concentrations of fine particles.

In this study, we use the Geographic Information System (GIS) to reconstruct the severe haze episodes that occurred over a large region and an extended time period in January, 2013 in northern and eastern China. In the urban area of one of the most polluted cities, Ji'nan in Shandong province, on-line measurements of trace gases, $PM_{2.5}$ and the aerosol components were conducted during this period and the fog samples were collected. The pollutant levels in the seriously polluted fog water were analyzed and compared with those in other locations. The influence of fog on cleansing the atmosphere during the severer haze episodes was also investigated.

2. Materials and methods

2.1. Inversion of $PM_{2.5}$ concentrations

The $PM_{2.5}$ levels in provinces or cities in China shown in this study were mostly derived from the air pollution index (API) records from air quality daily reports published by the China National Environmental Monitoring Center (a small fraction from local air quality daily reports). Firstly, we calculated the daily PM_{10} concentrations from the API records by using the following equation: $C = [(I - I_{low})/(I_{high} - I_{low})] \times (C_{high} - C_{low}) + C_{low}$ based on the classification of API described by Bian et al. (2011). Here, C is the PM_{10} concentration; I is the API. I_{low} and I_{high} stand for API grading limits that are lower and larger than I, respectively; C_{high} and C_{low} represent the concentrations of PM_{10} corresponding to I_{high} and I_{low}, respectively. This derivation is a common method to obtain PM_{10} concentrations for a large number of cities in China (Wang et al., 2006; Qu et al., 2010). The daily $PM_{2.5}$ levels were then estimated from the obtained PM_{10} data with assumption of a $PM_{2.5}/PM_{10}$ ratio of 0.6 based on previous studies in China (Cao et al., 2003; Sun et al., 2006; Fu et al., 2008; Gu et al., 2010; Kong et al., 2010). Note that the $PM_{2.5}/PM_{10}$ ratio of 0.6 used here was almost the lowest limit. The $PM_{2.5}$ levels in the provincial capital or the neighboring city were used to represent the status of fine particles pollution over the whole province. Although uncertainty existed in the representativeness of $PM_{2.5}$ levels by API data due to the difference in $PM_{2.5}/PM_{10}$ ratio among cities, the derived $PM_{2.5}$ data were a good indication of the pollution degree and the concentration variation patterns on a large temporal or spatial scale.

2.2. Collection and chemical analyses of fog samples

Fog samples were collected using a CASCC2 fog/cloud collector (Demoz et al., 1996) with a lower droplet size cut of 3.5 μm on the rooftop of a three-story building on the campus of Shandong University in Ji'nan (36.67° N, 117.05° E). The pH and electrical conductivity were determined immediately after the sampling stopped. OC and EC concentrations in the fog water were measured using an OC/EC analyzer (Sunset Lab) after a 1:100 dilution, by applying 10 μL diluted sample on the quartz filter suspended in the quartz insert of the analyzer. The water-soluble ions were analyzed using ion chromatograph (Dionex, ICS-90) after a 1:100,000 dilution and subsequent filtration.

2.3. On-line measurements of air pollutants and meteorological parameters

Ambient concentrations of air pollutants and meteorological parameters were concurrently measured in real-time from January 18 to February 1, 2013 at the site on the campus. $PM_{2.5}$ levels were measured using a Beta attenuation and optical analyzer (Thermo Scientific, model 5030 SHARP monitor). Nine inorganic water-soluble ions in $PM_{2.5}$ were analyzed using two on-line ion chromatographs coupling with a wet rotating denuder and a steam jet aerosol collector (Applikon-ECN, MARGA ADI 2080). OC and EC in $PM_{2.5}$ were analyzed using a semi-continuous thermo-optical OC/EC analyzer (Sunset Lab). SO_2 was measured using a pulsed UV fluorescence analyzer (Thermo Scientific, model 43C), and O_3 was measured using a UV photometric analyzer (Thermo Scientific, model 49C). NO and NO_2 were measured by a commercial chemiluminescence analyzer equipped with a molybdenum oxide converter (Thermo Scientific, model 42C). The relative humidity was measured using an automatic meteorological station (JZYG, PC-4), and visibility was measured using a forward-scattering visibility sensor (Vaisala, PWD22).

3. Results and discussion

3.1. $PM_{2.5}$ distribution in eastern China

Based on the API records from air quality daily reports of 24 provinces or cities, the derived 24-h average concentration of $PM_{2.5}$ in Ji'nan exceeded 360 μg m^{-3} on January 14, 2013 (see Supplementary Material Table 1), 4.8 times higher than the 24-h average concentration limit of $PM_{2.5}$ (75 μg m^{-3}) of the Ambient Air Quality Standards of Class II of China. As shown in Fig. 1, the $PM_{2.5}$ concentration above 200 μg m^{-3} covered a large region in north and middle eastern China with an area of ~1 million km^2 on January 14. The super-regional fine particle pollution was also indicated by the Moderate Resolution Imaging Spectroradiometer true color images (see Supplementary Material Fig. 1), extending to East China Sea in the east, Mt. Hua in the west, Yangzte River in the south, and Heilungkiang River in the north. Based on the spatial distribution of $PM_{2.5}$, the area suffering from haze episode far exceeds the Beijing–Tianjin–Hebei region and the North China Plain reported by Che et al. (2014) and Wang et al. (2014a), also including the Northeast Plain, the Yangtze River

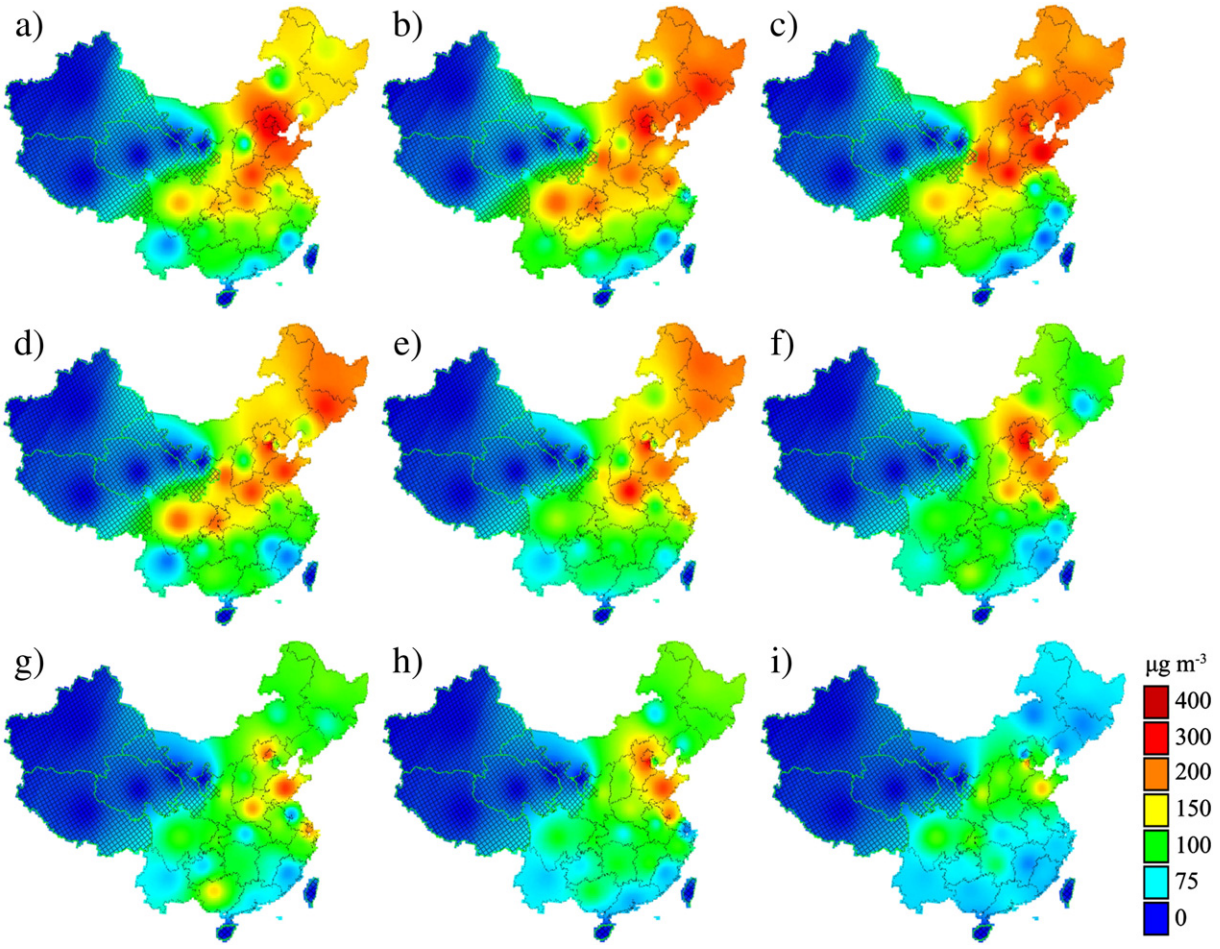

Fig. 1. Spatial distribution of $PM_{2.5}$ (μg m^{-3}) derived from API (24-h average) in China for days: (a) January 12, (b) 13, (c) 14, (d) 15, (e) 16, (f) 29, (g) 30, (h) 31, and (i) February 1. Grids in blue mean no data provided.

valley, and the Sichuan Basin. With examination of the emission distributions of aerosols and gas precursors in China (Zhang et al., 2009), the super-regional haze episodes exactly happened in the areas with intensive source emissions in northern and eastern China, which indicated that the fundamental cause of severe haze episodes was the emissions of massive amounts of air pollutants. However, of course, the unusual meteorological conditions in northern and eastern China promoted the accumulation of pollutions and the formation of secondary aerosols (Y. Wang et al., 2014; Zhang et al., 2014).

During the severe haze episodes in northern and eastern China in January, 2013, Ji'nan was among the top ten most polluted cities. The 24-h average concentration of $PM_{2.5}$ in Ji'nan was never less than 75 μg m^{-3} for a period of 28 days from January 5 to February 1, much longer than the average duration of 5 days for the "sawtooth circle" of fine particle concentration described for Beijing (Jia et al., 2008). The most polluted city was Shijiazhuang, the capital of Hebei province. The number of days with 24-h average $PM_{2.5}$ concentrations no less than 360 μg m^{-3} was 10 between January 5 and 29.

3.2. Carbonaceous materials and water-soluble ions in fog water

During the haze episodes, dense fog events occurred in Ji'nan and neighboring areas in the early morning of January 15, from the midday of January 29 to the morning of January 30, and from the night of January 30 to the morning of February 1. The occurrence of the fog events was mainly attributed to the weakened southerly surface winds and the reduced vertical shear of horizontal winds. The abnormally warm and humid air flow moved slow from the south and transported abundant water vapor to northern and eastern China (Zhang et al., 2014). The unusual meteorological conditions were favorable for the formations of both fog and haze. The detailed effects of the severe haze episodes on the occurrence of these fog events ere complex and remain unclear up to now. However, during the fog events the extremely high concentrations of air pollutants partly transferred into the fog droplets, significantly changing the physical and chemical properties of the fog water.

Fog water samples collected in Ji'nan were charcoal grey in color, indicating very high concentrations of carbonaceous materials. As shown in Fig. 2, elemental carbon (EC) concentrations were in the range of 0.8×10^3–1.25×10^4 mg L^{-1}. Organic carbon (OC) concentrations varied from 3.5×10^3 to 3.25×10^4 mg L^{-1}. The fog samples were also acidic, with in-situ pH values ranging from an extremely acidic value of 2.62 up to 4.20. The fog samples contained extremely high levels of water-soluble ions, with electrical conductivity values all above 2000 μS cm^{-1} (the upper limit of the Conductivity Meter). SO_4^{2-}, NO_3^-, and NH_4^+ were the most abundant water-soluble ions, with concentrations of 0.6×10^5–1.54×10^6, 0.4×10^5–8.98×10^5, and 0.9×10^5–1.75×10^6 μeq L^{-1} (μN), respectively. The contents of Cl^- and Ca^{2+} were also very rich, which exhibited maximum concentrations of 4.35×10^5 and 4.28×10^5 μN, respectively. Compared to fog collected at other sites in the world (see Supplementary Material Table 2), the major water-soluble ions in the Ji'nan fog samples are much higher, more than 22–544 times for SO_4^{2-}, 16–371 for NO_3^-, 23–437 for NH_4^+, 18–369 for Cl^-, and 10–207 for Ca^{2+}. The Cl^-/Na^+ ratios of collected fog samples were in the range of 3.4–11.8, suggesting that the pollutants in the fog are strongly influenced by

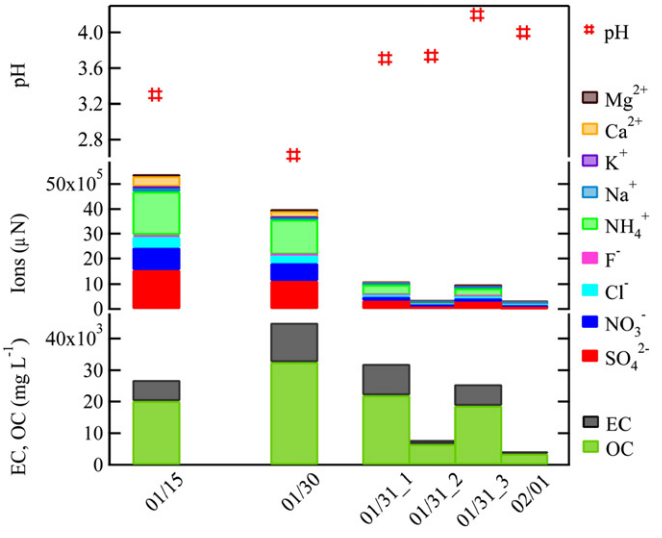

Fig. 2. The pH, water-soluble ions, EC and OC concentrations of fog water samples.

coal combustion in the local area and across the North China Plain (Ye et al., 2003; Mira-Salama et al., 2008).

3.3. Pollution tide during fog events

Pollutant timelines during the Ji'nan fog events (see Fig. 3) illustrated the positive effects that fog can have on air quality. The most impressive effect appeared during a fog event at the end of January. Beginning at 7:00 pm in the night of January 30 and extending until 8:00 am in the morning of February 1, this extended fog event was accompanied by substantial reductions, as shown in Fig. 3, in SO_2 and NO_x, and fine particles. CO mixing ratios also decreased obviously during the event; however, the reduction lagged when compared with those of SO_2, NO_x, and $PM_{2.5}$. The hourly $PM_{2.5}$ level (the $PM_{2.5}$ data recorded at the site of Shandong University in Ji'nan) was as high as 701 μg m^{-3} immediately before the fog event on January 31, and dropped to 115 μg m^{-3} in 36 h after the fog event. During the Ji'nan fog event, the percentage reduction of fine particle SO_4^{2-} was 90%. Reductions of NO_3^-, OC, and EC were 81%, 76%, and 71%, respectively. The overall $PM_{2.5}$ reduction was 84%, significantly higher than that of CO (~70%, as a benchmark to a certain degree considering the changes in air mass and mixing layer). This reduction indicated that during severe haze episodes fog had capacity to scavenge fine particles followed by wet deposition to the surface. While fog deposition is not characterized here, others (Collett et al., 2001) have demonstrated the efficient scavenging and deposition of inorganic pollutants in radiation fog in California. During the fog event from the midday of January 29 to the morning of January 30, apparent reductions in fine sulfate and SO_2 concentrations were also observed. We believe that the extremely high concentrations of carbonaceous materials and water-soluble ions in fog water came from the fog scavenging of fine particles and possibly some gas precursors. The role of fog in removing fine particles can also be seen in our previous studies in North China (Zhou et al., 2009; Wang et al., 2012).

Despite the apparent cleaning effect on the atmosphere by fog scavenging and deposition, the $PM_{2.5}$ level never fell to 75 μg m^{-3} in the fog events in this study. $PM_{2.5}$ concentration sharply increased from 115 to 195 μg m^{-3} within 4 h after the fog event in the morning of February 1. These results showed that air pollutants were efficiently removed by fog, but it could hardly clean up air pollutants down to good air quality during such severe haze episodes.

4. Summary and conclusions

In summary, from January 1 to February 1, 2013, north and middle areas of eastern China experienced continuous severe fine particle pollution. The super-regional haze episodes covered an area of ~1 million km^2,

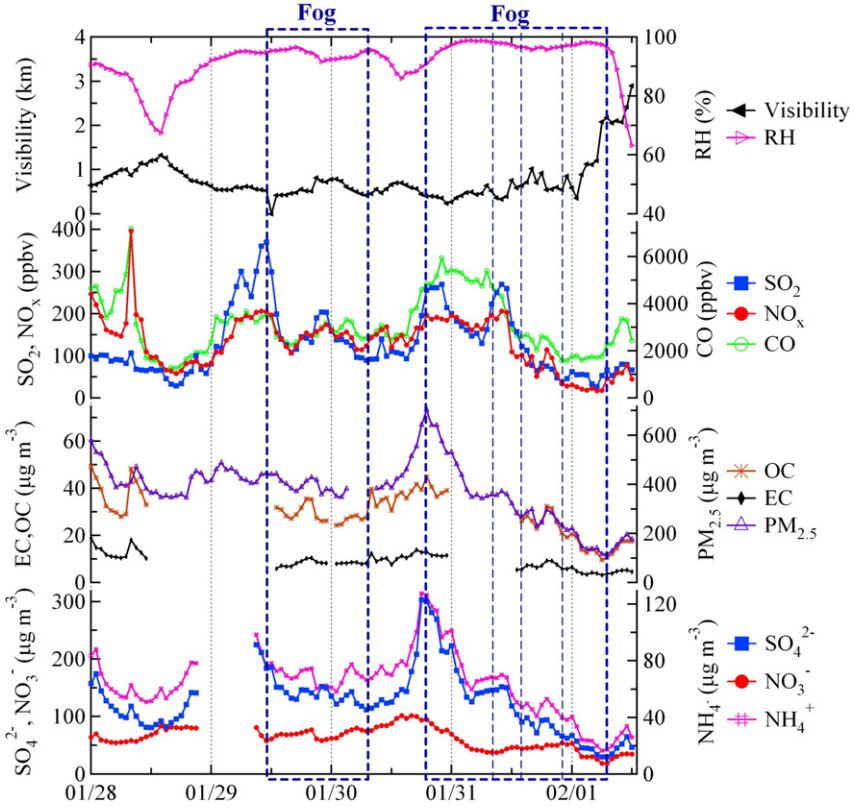

Fig. 3. Hourly average concentrations of $PM_{2.5}$, OC, EC, SO_4^{2-}, NO_3^-, NH_4^+, SO_2, NO_x, CO, visibility, and RH during fog events.

far beyond the Beijing–Tianjin–Hebei region. In particular on January 14, the daily average concentrations of $PM_{2.5}$ in most of cities in this region were above 200 μg m^{-3}. The severe haze episodes happened in China in early 2013 sound the alarm again to the government and researchers. They emphasize the urgent need to find approaches to control $PM_{2.5}$. During the severe haze episodes, several fog events occurred. The fog scavenged a large proportion of the fine particles in atmosphere and thus contained extremely high levels of carbonaceous materials and water-soluble ions. Nevertheless, it was difficult for fog to clean up fine particles to eliminate the regional haze episodes. The incorporation of pollutants in fog is also a concern as the deposited pollutants may damage ecosystems and the capture of light absorbing black carbon in the regional fog could significantly alter the optical properties and thus impact climate change (Jones et al., 2011; Allen et al., 2012). Further studies are required to give a comprehensive evaluation.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.05.135.

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