Agricultural Fire Impacts on the Air Quality of Shanghai during Summer Harvesttime

Hongyu Li1,2, Zhiwei Han3, Tiantao Cheng2,3*, Huanhuan Du2, Lingdong Kong2, Jianmin Chen2, Renjian Zhang3, Weijie Wang2

1 Weather Modification Office, Beijing Meteorological Bureau, Beijing 100089, China
2 Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China
3 Key Laboratory of Region Climate-Environment Research for Temperate East Asia (TEA), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

ABSTRACT

Agricultural fire is an important source of atmospheric carbonaceous aerosols. To better understand biomass burning emission originating from fire over Asian agriculture areas and its transport into the downwind atmosphere, aerosols and major trace gases were measured continuously from 22 May to 30 June at Shanghai during the summer harvesttime of 2009. Water-soluble K material contained in aerosols showed a clear day-to-day pattern with an average of 1.25 ± 1.48 µg/m3. K ion loading and ratio of K ion to PM10 drastically increased during ‘K event’ days, accompanying with high PM10, SO2, and NO2 levels. MODIS remote sensing fire map revealed about 80% agricultural fires occurred in the agriculture areas of Anhui, Jiangsu, Shandong and Henan provinces. Four potential source areas of agricultural fires, identified as Shanghai, Zhejiang, Jiangsu and Anhui provinces, had significant contributions to worsen the air quality of Shanghai during the harvest season.

Keywords: Agricultural fire; Air quality; Potential source.

INTRODUCTION

Smoke aerosols contain a large amount of partially oxidized organic material and black carbon (BC) or soot, which may have significant climate implications (IPCC, 2007). Second only to CO2 in terms of direct climate forcing, BC is believed to be a principal component of global warming through absorption of solar radiation since its warming effect has been found to balance the net cooling effect of other anthropogenic aerosol constituents (Jacobson, 2001). Smoke aerosols originating from urban and industrial pollutions or some from wildfires have been shown to affect cloud microphysics, cloud reflection of sunlight to space, and the onset of precipitation (Andreae et al., 2004). Delays in the onset of precipitation can increase cloud lifetime and thereby increase cloud cover (Resenfeld et al., 2008). Most past researches on BC have been conducted in regions outside of Asia, but increasing recent studies within Asia show the importance of BC on that continent such as Asian Brown Cloud. Streets et al. (2001) estimated that BC emission in China (1342 Gg in 1995 and 1224 Gg in 2020), represents 30% of global anthropogenic emissions. Bond et al. (2004) have produced similar findings. Ohara et al. (2007) developed a new emission inventory for Asia for the period 1980–2020, and showed a rapid growth by 28% for BC and 30% for OC. The region is considered one of largest sources of smoke emission in the world.

Agricultural fire emits abundant gases and particles into the atmosphere, which causes serious local and regional air pollution during harvest seasons. In China, the field combustion of agricultural residues is a common way to eliminate waste after harvesting and a significant type of biomass burning. Measurements on gaseous and particulate emission factors from open burning of agricultural residues have been reported by Li et al. (2007) and Zhang et al. (2008). Some studies estimated that 17–25.6% of the total agricultural residue production, or 110–157.5 Tg of crop wastes were burned in the field in China every year (Cao et al., 2003, Yan et al., 2006). Cheng et al. (2006) carried out a continuous measurement of BC concentration from June 2004 to May 2005 at Hok Tsui, a site located downward of east Asia and China, and investigated the influence of southward outflow of continental pollution. Cao et al. (2003) profiled the characteristics of carbonaceous aerosols in the Pearl River Delta of China, and obtained the average

* Corresponding author. Tel.: (86)21-6564 2521; Fax: (86)21-6564 2080
E-mail address: ttcheng@fudan.edu.cn
concentrations of OC 14.7 µg/m³ and EC 6.1 µg/m³ in PM_{1.5} during winter 2001. Cao et al. (2005) also characterized carbonaceous aerosols and apportioned the sources of EC and OC during the fall and winter of 2003 in Xi’an, China. Other measurements on carbonaceous aerosols and their distributions have been conducted in sites of Waliguan (Tang et al., 1999), Beijing (Lou et al., 2005), Tongliao (Li et al., 2006) and Wusumu (Han et al., 2008), etc. Li et al. (2009) used the dataset of agricultural fire sites derived from satellite remote sensing to analyze the impacts of field straw burning on urban air quality during summer wheat-harvested period.

Past studies have sufficiently portrayed the characteristics and role of anthropogenic aerosols in the atmosphere of Shanghai (Wang et al., 2006), however, to our knowledge few has focused on the influence of carbonaceous aerosols from widespread rural biomass burning in Chinese agricultural areas. The purpose of this paper is to report agricultural fire impacts on the air quality of Shanghai using satellite remote sensing fire site data, and to determine the potential source contribution through tracing major outflow pathways.

MEASUREMENT

The atmospheric research station (31.3°N, 121.5°E) was established in 2005, and instruments were placed on the roof of one build about 20 m height above the ground in the campus of Fudan University in Shanghai of China.

A model ADI 2080 online analyzer of Monitoring of AeRosols and GAses (MARGA, Applikon Analytical B. V. Corp., Netherlands) with a PM_{10} inlet was used to measure the mass concentrations of major water-soluble aerosol inorganic ions and trace gases at the time resolution of 1 hour from 22 May to 30 June 2009. MARGA consists of a sampling box and an analytical box, with a particle collection efficiency of 99.7%. Ambient air is absorbed into the sampling box (flow 1 m³/h) through the inlet, and then particles in residual airflow go through the supersaturated steam (0.0035% H₂O₂, 120–140°C) formed by one Wet Rotating Denuder (WRD), and then particles in residual airflow go through the supersaturated steam (0.0035% H₂O₂, 120–140°C) erupted from one Steam Jet Aerosol collector (SJAC) to be pooled into its collector. These two liquid samples flow through glass filters respectively, and are stored in syringe pumps of the analytical box ready for analyzing by Ion Chromatography (IC). MARGA has the capability of measuring the hourly average concentrations of NH₄⁺, Na⁺, K⁺, Ca²⁺, Mg²⁺, SO₄²⁻, NO₃⁻, Cl⁻, HCl, HNO₂, SO₂, HNO₃, NH₃ in the atmosphere. During the field campaign, MARGA was calibrated using internal standard solution (LiBr) every week to verify accurate detecting limits and to ensure data quality. The daily average concentration of K ion was used here to identify field fire events as a proper indicator of biomass burning emission (Chow, 1995).

The daily fire site data during harvest seasons over Chinese agricultural areas between 19 May and 30 June 2009 were derived from the Ministry of Environmental Protection of China (Table 1), which was screened using ground land-use information on the basis of MODIS daily 1-km level 3 hotspot/fire products. Meteorological factors in Shanghai, including temperature, relative humidity (RH), wind speed and visibility, were downloaded from internet site http://www.wunderground.com. Daily average concentrations of SO₂, NO₂ and PM_{10} from the Shanghai Environmental Protection Bureau (http://www.envir.gov.cn/airnews). To identify the influence of agricultural fire emissions from different regions on air quality at the sampling site, air mass trajectories were calculated using the HYSPLIT 4 model of the Air Resources Laboratory of NOAA (http://www.arl.noaa.gov/ready/hysplit4.html).

RESULTS AND DISCUSSION

Variation of Air Quality

It was known that water-soluble K ion is a proper indicator to be used in estimating biomass burning emission and tracing carbonaceous aerosol long-range transport in the atmosphere (Chow, 1995). The daily means of K ion concentrations for this study were evaluated from hourly average K⁺ data collected by MARGA with PM_{10} sampler. Measured from 22 May to 30 June (Fig. 1), average K⁺ concentration at Shanghai was 1.25 ± 1.48 µg/m³. This is comparable to previous studies conducted at Beijing suburb by Zheng et al. (2005), with a yearly mean of 1.21 ± 0.97 µg/m³, and at Beijing by Pu and Zhao (2006), with a autumnal mean of 1.49 µg/m³. Average ratio

| Date       | Fire number | Date       | Fire number | Date       | Fire number | Date       | Fire number |
|------------|-------------|------------|-------------|------------|-------------|------------|-------------|
| 19 May     | 4           | 10 June    | 226         | 21 June    | 31          |
| 20 May     | 39          | 11 June    | 16          | 22 June    | 128         |
| 21 May     | 64          | 12 June    | 372         | 23 June    | 11          |
| 22 May     | 0           | 13 June    | 434         | 24 June    | 32          |
| 23 May     | 109         | 14 June    | 274         | 25 June    | 2           |
| 24 May     | 1           | 15 June    | 153         | 26 June    | 9           |
| 25 May     | 83          | 16 June    | 13          | 27 June    | 11          |
| 26 May     | 2           | 17 June    | 37          | 28 June    | 3           |
| 27 May     | 268         | 18 June    | 2           | 29 June    | 17          |
| 28 May     | 56          | 19 June    | 35          | 30 June    | 4           |
| 29 May     | 175         | 20 June    | 4           |            |             |
of K+ concentration to PM10 was 0.0125, less than K/TSP 0.0728 measured at Beijing in the autumn of 2004 (Pu and Zhao, 2006). A day-to-day variation of K+ concentration was obvious, with the highest daily average on 30 May (8.19 µg/m³) and the lowest value on 19 June (0.043 µg/m³) (Fig. 1).

In order to profile the impacts of biomass burning emission from agricultural fire on air quality, we first scanned the daily K+ loading data to pick up the ‘K event’, which was defined as periods when daily average K+ concentration exceeded background value. Based on three criteria of K+ concentration lower than average (1.25 µg/m³), no agricultural fire occurrence in Shanghai suburb and visibility exceeding 10 km, the days disturbed by low carbonaceous aerosol loading were sieved out for clear periods. The average K+ concentration of these selected clear days, in a magnitude of 0.3 µg/m³, was served as the background against the ‘K event’, indicating contributions of other sources such as industrial and vehicle emissions in Shanghai, and remote biomass burning in domestic cooking and heating from upwind. As seen in Fig. 1, K ion loading and ratio K+/PM10 drastically increased during event days. These criteria resulted in a somewhat subjective selection of events from the available records, but we expect that this will make the ‘K event’ more representative. Since there are no stable sources continuously emitting carbonaceous aerosols near the sampling location, except for traffic emissions, those events should reflect the local and long-range transport pollution from biomass burning related to agricultural fires.

Table 2 shows mean PM10 chemical and meteorological conditions in various K+ mass concentrations. High averages of PM10, K+, SO2, NO2 concentrations and K+/PM10 appeared during the events compared to the background. For example, the average K+ concentration (3.96 µg/m³) of severe event days within 2.0–8.5 µg/m³ was more than 24 times that of noevent days in 0.0–0.3 µg/m³ range. In contrast, meteorological parameters of temperature, RH, wind speed and visibility almost reduced with K loading promoting. Wind speeds were usually low, with an average of 2.61 m/s for all event days, compared to the average 3.24 for noevent days. Meanwhile, visibility generally showed lower values during events than noevents. All of these are evidence of poor dispersion condition during carbonaceous aerosol events.

### Agricultural Fire Event

To investigate outflows of biomass burning emission related to agricultural fire, the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) was used to trace 5-day back trajectories using K+ as a marker for carbonaceous aerosols transported from agricultural regions. Fig. 2 shows 5-day back trajectories arriving at 150 m above the Shanghai sampling site for each day in high K+ loading exceeding the background level during the summer harvest season of 2009. It was notable that the agricultural areas covered by the 5-day back trajectories arriving at the sampling site during daytime were similar with those during nighttime. As Fig. 2 shown, air parcel outflows arriving at the sampling site, prevailed in northerly and southerly pathways, had traveled a long distance over the Asian continent, and then toward coastal areas in eastern China, or across Fujian, Zhejiang provinces and even the Yangtze River Delta, and then southward to Shanghai. Streets et al. (2001) reported that BC emissions in China were concentrated in a west-to-east region across the agricultural heartland from Sichuan to Hebei provinces. In comparison with the BC emission inventory developed for China by Streets et al. (2001; 2003), most air masses arriving at Shanghai did not travel over those major BC emission source areas.
Table 2. Summary of chemical and meteorological conditions in various K⁺ mass concentrations.

| K⁺ conc. (µg/m³) | PM₁₀ (mg/m³) | SO₂ (mg/m³) | NO₂ (mg/m³) | Temp. (°C) | RH (%) | Wind speed (m/s) | Visibility (km) | K/PM₁₀ |
|------------------|--------------|-------------|-------------|------------|--------|-----------------|-----------------|--------|
| 0.0–0.3          | 0.049        | 0.022       | 0.038       | 27.38      | 63.55  | 3.24            | 11.65           | 0.003  |
| 0.3–1.0          | 0.086        | 0.031       | 0.050       | 25.00      | 65.88  | 2.91            | 10.47           | 0.009  |
| 1.0–2.0          | 0.105        | 0.040       | 0.061       | 23.56      | 61.32  | 2.48            | 8.91            | 0.014  |
| 2.0–8.5          | 0.140        | 0.048       | 0.070       | 23.80      | 47.17  | 2.44            | 8.78            | 0.033  |
| total            | 0.082        | 0.032       | 0.050       | 25.55      | 60.95  | 2.93            | 10.55           | 0.012  |

Fig. 2. Five-day air mass back trajectories at local time (a) 00:00 and (b) 12:00 during event days from 19 May to 30 June 2009.

Fig. 3 gives a map of fire distributions derived from MODIS remote sensing products over primary Chinese agricultural areas from 19 May and 30 June 2009. These daily-summed fire sites scattered in regions partially corresponding with areas covered by the calculated air outflows during the summer harvest season (Fig. 2). Fig. 4 shows a statistics for all fire-site records collected in the whole harvest season. As can be seen, about 80% of agricultural fires occurred in Anhui, Jiangsu, Shandong and Henan provinces, and only less than 10% fires were found in Zhejiang, Shanghai, Hebei and other provinces, respectively.

Potential Source Contribution

In order to evaluate the potential source contribution of agricultural fires to air pollution in the atmosphere of Shanghai, correlation coefficient values were calculated between daily-summed fire numbers and daily-averaged K⁺
mass concentrations. Moreover, back-to-fire interval, a period that biomass burning emission transported long range from source areas arriving over the sampling site along air mass outflows, was considered in these correlativity analyses. According to the results of relationship analysis (Table 3), four potential source areas of agricultural fires were identified as having important contributions to air quality at the sampling site. Shanghai, Zhejiang, Jiangsu and Anhui provinces constituted the four important source areas.

The biomass burning emission from agricultural fires, travelling for a period following air mass outflows, arrived at the sampling site to affect carbonaceous aerosol loading. As seen in table 3, for all event days with K⁺ concentrations exceeding the background 0.3 µg/m³, only the fire site records of Shanghai and Zhejiang provinces exhibited a high relativity with the K⁺ ion loadings in intraday and 1-day intervals. The fire site records of Jiangsu province were high correlated to the K⁺ ion loadings exceeding 1.0 µg/m³ in 3-day interval, and the K⁺ ion loadings exceeding 3.0 µg/m³ in intraday and 1-day intervals. Similar results were found in the fire site records of Anhui, Shandong, Guangdong, and Jiangxi provinces. All these indicated that agricultural fires which occurred in Shanghai suburb and Zhejiang province were the most important sources to rapidly and easily impact the air quality of Shanghai. Only during moderate and heavy events, agricultural fires which occurred in Jiangsu, Anhui, Shandong, Henan, and Guangdong provinces would force biomass burning emission to transport a long-range distance into the atmosphere of Shanghai. The potential source contribution of agricultural fires to Shanghai air quality is primary Shanghai and Zhejiang provinces, secondary Jiangsu and Anhui provinces, and final other provinces.

CONCLUSIONS

Measurements of major water-soluble inorganic ions contained in aerosols and trace gases were conducted at Shanghai from 22 May to 30 June during the summer harvesttime of 2009. The mean K⁺ mass concentration during the whole campaign was 1.25 ± 1.48 µg/m³. Throughout the sampling period, the minimum daily mean K⁺ ion loading was 0.043 µg/m³ on 19 June, and the highest value occurred on 30 May (8.19 µg/m³). Additional, outflow back trajectories clearly revealed that carbonaceous aerosols from biomass burning emission related to agricultural fires had significant impacts on the air quality of Shanghai.
Table 3. Correlation coefficients between daily average K+ mass concentration and 1-day fire site number remote-sensed by satellite. The back-to-fire interval is the days from PM10 observation back to fire occurrence.

| K+ conc. (µg/m³) | Back-to-fire interval | Shanghai | Zhejiang | Jiangsu | Anhui | Henan | Shandong | Guangdong-Jiangxi |
|------------------|-----------------------|----------|----------|---------|-------|-------|----------|------------------|
| 0.3–8.5          | 0 day                 | 0.60     | 0.55     | 0.02    | −0.09 | −0.15 | 0.04     | −0.02            |
|                  | 1 day                 | 0.79     | 0.82     | 0.18    | −0.03 | −0.13 | −0.16    | −0.03            |
|                  | 2 days                | 0.17     | 0.26     | 0.14    | 0.01  | −0.13 | −0.10    | −0.12            |
| 1.0–8.5          | 0 day                 | 0.60     | 0.57     | −0.02   | −0.25 | −0.33 | −0.05    | −0.11            |
|                  | 1 day                 | 0.82     | 0.91     | 0.34    | −0.12 | −0.24 | −0.19    | −0.14            |
|                  | 2 days                | 0.04     | 0.21     | 0.32    | −0.07 | −0.19 | −0.22    | −0.17            |
|                  | 3 days                | −0.13    | 0.15     | 0.75    | −0.06 | −0.13 | 0.24     | 0.18             |
| 2.0–8.5          | 0 day                 | 0.55     | 0.54     | −0.08   | −0.03 | −0.35 | −0.12    | −0.30            |
|                  | 1 day                 | 0.84     | 0.90     | 0.21    | −0.26 | −0.45 | −0.20    | −0.23            |
|                  | 2 days                | −0.13    | 0.01     | 0.23    | 0.75  | −0.44 | −0.05    | 0.24             |
|                  | 3 days                | −0.24    | 0.03     | 0.77    | −0.07 | −0.41 | 0.11     | 0.86             |
| 3.0–8.5          | 0 day                 | 0.50     | 0.50     | 0.65    | 0.63  | 0.87  | −0.09    | −0.49            |
|                  | 1 day                 | 0.84     | 0.89     | 0.83    | −0.05 | −0.49 | −0.16    | −0.47            |
|                  | 2 days                | −0.19    | −0.06    | 0.20    | 0.85  | −0.55 | −0.11    | 0.22             |
|                  | 3 days                | −0.30    | −0.02    | 0.81    | 0.14  | −0.33 | 0.89     | 0.89             |

Using back trajectories for the days with high K+ concentrations, four potential source areas of agricultural fires of Shanghai suburb, Zhejiang, Jiangsu and Anhui provinces were identified as having important contributions to air quality at the sampling site. Most air masses arriving at Shanghai did not travel over the agricultural heartland in China. The biomass burning emission of agricultural fires in source areas of Shanghai suburb and Zhejiang provinces had more influence on K material in aerosols than source areas of Jiangsu, Anhui and other provinces.

ACKNOWLEDGMENTS

This research was supported by the National Natural Science Foundation of China (40605001), the National 973 Project of China (2006CB403703), the Shanghai Dawn Project (08SG07), the Hundred Talents Program of the Chinese Academy of Sciences, and the Key Laboratory of Region Climate-Environment Research for Temperate East Asia of the Chinese Academy of Sciences.

REFERENCES

Andreae, M.O., Rosenfeld, D., Artaxo P., Costa, A.A., Frank, G.P., Longo, K.M. and Silva-Dias, M.A.F. (2004). Smoking Rain Clouds over the Amazon. Science 303: 1337–1342.

Bond, T.C., Streets, D.G., Yarber, K.F., Nelson, S.M., Woo, J.H. and Klimont, Z. (2004). A Technology-Based Global Inventory of Black and Organic Carbon Emissions from Combustion. J. Geophys. Res. 109: D14203

Cao, J.J., Lee, S.C., Ho, K.F., Zhang, X.Y., Zou, S.C., Fung, K., Chow, J.C. and Watson, J.G. (2003). Characteristics of Carbonaceous Aerosol in Pearl River Delta Region, China during 2001 Winter Period. Atmos. Environ. 37: 1451–1460.
Li, X.H, Wang, S.X., Duan, L., Hao, J.M., Li, C., Chen, Y.S. and Yang, L. (2007). Particulate and Trace Gas Emissions from Open Burning of Wheat Straw and Corn Stover in China. *Environ. Sci. Technol.* 41: 6052–6058.

Li, X.X., Shen, Z.X., Cao, J.J., Liu, S.X., Zhu, C.S. and Zhang, T., (2006). Distribution of Carbonaceous Aerosol during Spring 2005 over the Horqin Sandland in Northern China. *China Particuology* 4: 316–322.

Lou, S.J., Mao, J.T. and Wang, M.H. (2005). Observational Study of Black Carbon Aerosol in Beijing. *Acta Scien. Circum.* 25: 17–22 (Chinese).

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X. and Hayasaka, T. (2007). An Asian Emission Inventory of Anthropogenic Emission Sources for the Period 1980–2020. *Atmos. Chem. Phys.* 7: 4419–4444.

Pu, Y.F. and Wu, R.X. (2006). Research on Chemical Composition and Sources of Atmospheric Particles in Autumn of Beijing in 2004. *Clim. Environ. Res.* 11: 739–744 (Chinese).

Rosenfeld, D., Lohmann, U., Raga, G.B., O’Dowd, C.D., Kulmala, M., Fuzzi, S., Reissell, A. and Andreae, M.O. (2008). Flood and Drought: How Do Aerosols Affect Precipitation? *Science* 321: 1309–1313.

Streets, D.G., Gupta, S., Waldhoff, S.T., Wang, M.Q., Bond, T.C. and Bo, Y.Y., (2001). Black Carbon Emissions in China. *Atmos. Environ.* 35: 4281–4296.

Streets, D.G., Bond, T.C., Carmichael, G.R., Fernandes, S.D., Fu, Q., He, D., Klimont, Z., Nelson, S.M., Tsui, N.Y., Wang, M.Q., Woo, J.H. and Yarber, K.F. (2003). An Inventory of Gaseous and Primary Aerosol Emissions in Asia in the Year 2000. *J. Geophys. Res.* 108: 8809.

Tang, J., Wen, Y.P., Zhou, L.X., Qi, D.L., Zheng, M., Trivett, N. and Wallgren, E. (1999). Observational Study of Black Carbon in Clean Air Area of Western China. *Q. J. Appl. Meteor.* 10: 160–170 (Chinese).

Wang, Y., Zhuang, G., Zhang, X., Huang, K., Xu, C., Tang, A., Chen, J. And An, Z. (2006). The Ion Chemistry, Seasonal Cycle, and Sources of PM$_2.5$ and TSP Aerosol in Shanghai. *Atmos. Environ.* 40: 2935–2952.

Yan, X., Ohara, T. and Akimoto, H. (2006). Bottom-up Estimate of Biomass Burning in Mainland China. *Atmos. Environ.* 40:5262–5273.

Zhang, H.F., Ye, X.N., Cheng, T.T., Chen, J.M., Yang, X., Wang, L. and Zhang, R.Y. (2008). A Laboratory Study of Agricultural Crop Residue Combustion in China: Emission Factor and Emission Inventory. *Atmos. Environ.* 42: 8432–8441.

Zheng, X.Y., Liu, X.D., Zhao, F.H., Duan, F.K., Yu, T. and Cachier, H. (2005). The Season Characteristics of Biomass Burning Emission Contribution to Aerosols at Beijing. *China Sci.* 35: 346–352 (Chinese).

Received for review, August 3, 2009
Accepted, November 4, 2009