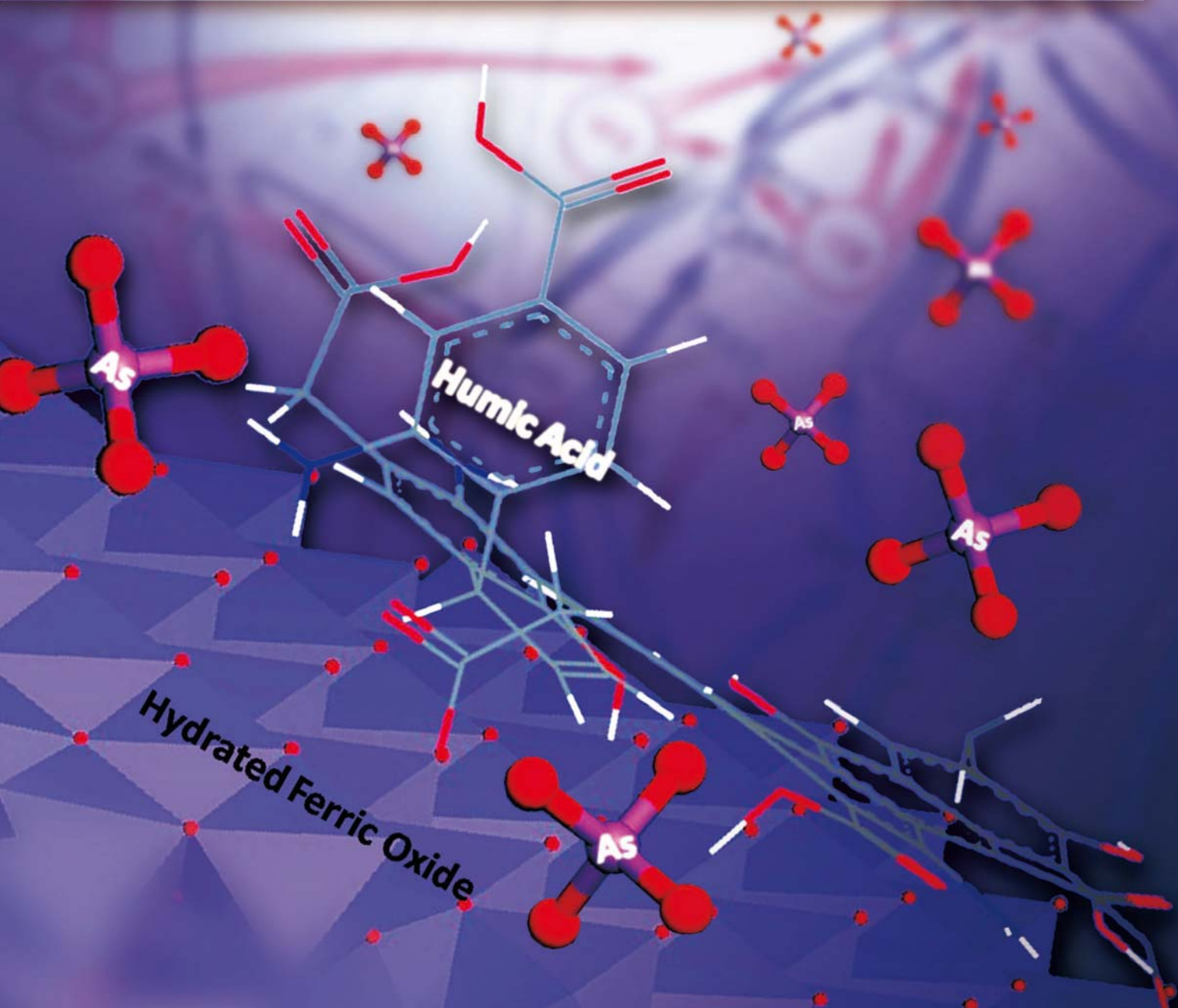


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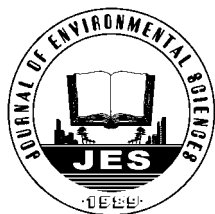
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## Online monitoring of water-soluble ionic composition of PM<sub>10</sub> during early summer over Lanzhou City

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### ABSTRACT

Lanzhou is one of the most aerosol-polluted cities in China. In this study, an online analyzer for Monitoring for AeRosols and GAses was deployed to measure major water-soluble inorganic ions in PM<sub>10</sub> at 1-hour time resolution, and 923 samples were obtained from Apr 1 to May 24, 2011. During the field campaign, air pollution days were encountered with Air Quality Index more than 100 and daily average concentration of PM<sub>10</sub> exceeding 150 μg/m<sup>3</sup>. Based on the variation of water-soluble ions and results of Positive Matrix Factorization 3.0 model execution, the air pollution days were classified as crustal species- or secondary aerosol-induced, and the different formation mechanisms of these two air pollution types were studied. During the crustal species pollution days, the content of Ca<sup>2+</sup> increased and was about 2.3 times higher than the average on clear days, and the air parcel back trajectory was used to analyze the sources of crustal species. Data on sulfate, trace gases and meteorological factors were used to reveal the formation mechanism of secondary aerosol pollution. The sulfur oxidation ratio (SOR) was derived from the 923 samples, and the SOR had high positive correlation with relative humidity in early summer in Lanzhou.

## Introduction

Atmospheric particulate matter, also known as particulates or particulate matter (PM), are tiny pieces of solid or liquid matter associated with the Earth's atmosphere. They are suspended in the atmosphere as atmospheric aerosol (Seinfeld and Spyros, 1998). PM<sub>10</sub> is the atmospheric aerosol with air dynamic equivalent diameter less than 10 μm; it can penetrate into the lower respiratory system of humans and increase mortality due to respiratory and cardiovascular disease (Bert and Stephen, 2002). Moreover, it affects the Earth's climate by changing incoming solar radiation and outgoing terrestrial long wave radiation retained in the Earth's system (Huang et al., 2008), and its climate effects are the largest source of uncertainty in future climate predictions (Solomon et al., 2007). From

the definition of air pollution (Juliann and Jim, 2001), PM<sub>10</sub> is an atmospheric pollutant and can create significant global health, welfare and ecological impacts. In the latest Ambient Air Quality Standards promulgated in Feb 29, 2012 by the Ministry of Environmental Protection of China (MEP-China, 2012a), the mass concentration of PM<sub>10</sub> is a basic monitoring item and a daily concentration lying above 150 μg/m<sup>3</sup> is used as an indicator of pollution days.

The Air Quality Index (AQI) is a quantitative method to profile air pollution level. It is determined by the maximum value of the Individual Air Quality Index (IAQI), which is calculated from mass concentrations of PM<sub>10</sub>, PM<sub>2.5</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO and O<sub>3</sub> in ambient air respectively; furthermore, the pollutant with maximum IAQI is called the primary pollutant. AQI is measured at monitoring stations throughout 120 cities and reported daily by MEP-China. More detail on the algorithm of AQI was presented in the Technical Regulation on Ambient Air Quality Index

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promulgated by MEP-China (MEP-China, 2012b). **Table 1** lists levels of AQI with corresponding health implications. Air is polluted and might impact human health when AQI is more than 100.

**Table 1** AQI levels with human health implications

AQI	Air Pollution Level	Health implications
0–50	Excellent	No health implications
51–100	Good	No health implications
101–150	Slightly polluted	Slight irritations may occur
151–200	Lightly polluted	Slight irritations may occur
201–250	Moderately polluted	Healthy people will be noticeably affected
251–300	Heavily polluted	Healthy people will be noticeably affected
300+	Severely polluted	Healthy people will experience problems

Lanzhou is located in a narrow and long valley basin in northwest China; this topographic characteristic causes low wind speed, and inversion layers occur frequently. Pollutants are difficult to disperse when encountering these meteorological conditions (Chu et al., 2008). It was already known that air pollution events occur frequently in Lanzhou in wintertime, and many works have investigated these wintertime pollution events (Zhang et al., 2008; Chu et al., 2008; An et al., 2005, 2007). In spring and summer, the formation mechanism of dust storms was the research focus (Wang et al., 2006; Liu et al., 2004; Zhang et al., 2003). However, no works have been carried out to measure water-soluble ions and trace gases in PM<sub>10</sub> during early summer by an online instrument with 1-hour resolution in Lanzhou, and then use the dataset to analyze the city's air pollution events.

In this article, we measured major water-soluble inorganic ions (NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>), gaseous precursors (HCl, HONO, SO<sub>2</sub>, HNO<sub>3</sub>, NH<sub>3</sub>) and daily PM<sub>10</sub> in Lanzhou, and used the Positive Matrix Factorization (PMF) model to apportion the sources of water-soluble ions. The purpose is to report the physical/chemical properties of PM<sub>10</sub> in Lanzhou in early summer, and to reveal the evolution and conversion of this aerosol during air pollution events.

## 1 Data and methods

The observation site (36.05°N, 103.86°E) was located on the roof of a two-story building belonging to the College of Atmospheric Science, about seven meters in height above ground, within the campus of Lanzhou University (**Fig. 5**). The site represented a mixed influence of residential, traffic and construction emissions in an urban district.

A model ADI 2080 online analyzer for Monitoring for Aerosols and Gases (MARGA, Applikon Analytical B.V., Netherlands) with a PM<sub>10</sub> sampling inlet was used to obtain an hourly integrated dataset from Apr 1 to May 24,

2011. The MARGA, which was developed and affirmed by the Energy Research Centre of the Netherlands, is a commercialized version of the GRAEGOR system (Thomas et al., 2009) and consists of sampling and analytical boxes (Jongejan et al., 1997). The two most important modules in the sampling box are the Wet Rotating Denuder (WRD) for collection of the precursor gases (Keuken et al., 1988) and Steam Jet Aerosol Collector (SJAC) for collection of the PM (Khlystov et al., 1995). The detection system in the analytical box has the capability to measure mass concentrations of major water-soluble inorganic ions in PM (NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>) and trace gases (HCl, HONO, SO<sub>2</sub>, HNO<sub>3</sub>, NH<sub>3</sub>) at 1-hour time resolution. To track changes in retention time and detector response for each sampling, the MARGA was continuously controlled by an internal calibration method using bromide for the anion chromatograph and lithium for the cation chromatograph over the whole observation period. Full details on the MARGA are given elsewhere (Trebs et al., 2004). Methodologies of MARGA operation and maintenance followed the manual provided by the manufacturer.

Mass concentrations of daily PM<sub>10</sub> during the field campaign were acquired from the continuous monitoring using a TH-150C Automatic Medium Volume TSP Sampler (Wuhan Tianhong Instruments Co., Ltd., China) with TH-PM<sub>10</sub> Impactor and Whatman QMA. In order to avoid interaction between MARGA and TH-150C during the sampling time, the sampler was located seven meters away from MARGA. AQI of Lanzhou City was obtained from the website of MEP-China (<http://datacenter.mep.gov.cn/>). Three-hour meteorological data including mixed layer depth (MLD, m), relative humidity (RH, %), ambient temperature (K), rainfall (mm/hr) and downward solar radiation flux (W/m<sup>2</sup>) were obtained from the Global Data Assimilation System (GDAS, global, 2006–present) of NOAA Air Resources Laboratory.

## 2 Results and discussion

### 2.1 Air pollution events

In mainland China, AQI is viewed as a comprehensive index to measure levels of ambient air pollution. In this research, we considered the air to be polluted when intraday AQI > 100 and clear when AQI < 50 in Lanzhou City. On the basis of this method, Lanzhou experienced five air pollution events (sequences shown with light gray in **Fig. 1**) during the field campaign, and the primary pollutant for all of these events was inhalable particles (PM<sub>10</sub>). May 12 and 21 to 24 were clear days (sequences shown with light blue in **Fig. 1**).

**Figure 1** shows the time series of daily PM<sub>10</sub> mass concentrations and Three-hour precipitation from Apr 1



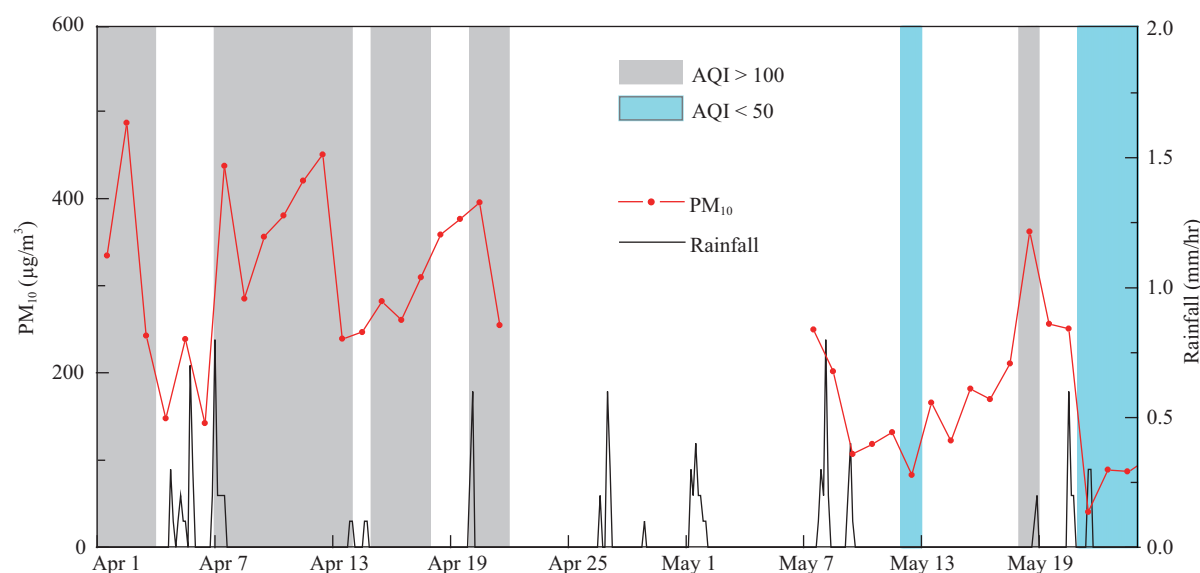


Fig. 1 Daily PM<sub>10</sub> mass concentration and 3-hour precipitation from Apr 1 to May 24.

to May 24. Viewing the variation of PM<sub>10</sub> for the whole period, the maximum daily concentration was 487 µg/m<sup>3</sup> on Apr 2, and these pollution events all exceeded the secondary standard (150 µg/m<sup>3</sup>) of Ambient Air Quality Standards (MEP-China, 2012a) in China; only 41 µg/m<sup>3</sup>, on May 21, was lower than the primary standard (50 µg/m<sup>3</sup>). Three-hour precipitation during the field campaign revealed that the pollution events could be referred to as continuous air contamination lasting throughout a free-rain period, and could be cleared by rainfall.

## 2.2 Water-soluble ions in PM<sub>10</sub>

Water-soluble ions like NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup> are important components in atmospheric particles. The sum of total water-soluble ions (TWSI) contributed a mean mass fraction of 16.03% in PM<sub>10</sub> for the field campaign, which was little higher than a previous observation in Beijing summertime (12.1%) (Cui et al., 2008) and much lower than in Lanzhou wintertime (27.34%) (Zhang et al., 2008). The average mass percentage of TWSI in PM<sub>10</sub> reached 14.4% during the five pollution events.

Figure 2 presents a time series of these water-soluble ions in PM<sub>10</sub> from Apr 1 to May 24. Here we convert mass concentrations (µg/m<sup>3</sup>) to molar concentrations (nmol/L) for these water-soluble (Fig. 2), because molar concentrations can better reflect the quantitative relationships

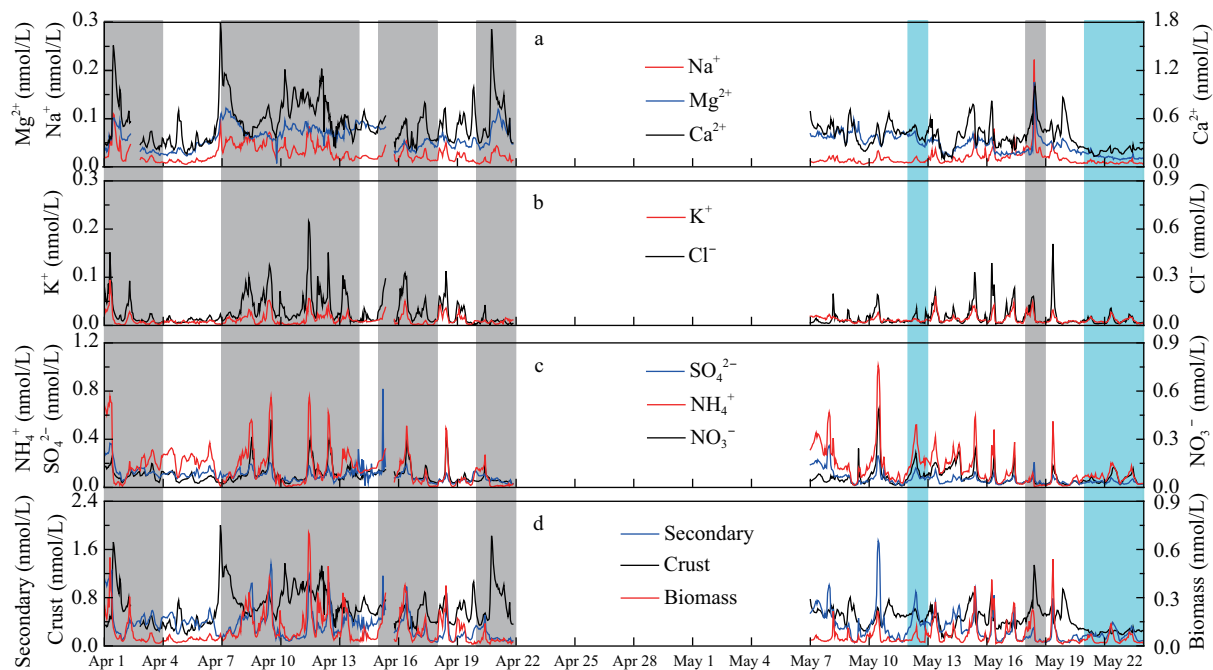
between water-soluble ions. Based on the average molar concentrations of water-soluble ions during field campaign, we ordered water-soluble ions from high to low: Ca<sup>2+</sup> > NH<sub>4</sub><sup>+</sup> > SO<sub>4</sub><sup>2-</sup> > NO<sub>3</sub><sup>-</sup> > Cl<sup>-</sup> > Mg<sup>2+</sup> > Na<sup>+</sup> > K<sup>+</sup>. Compared with the decreasing order: NH<sub>4</sub><sup>+</sup> > Ca<sup>2+</sup> > SO<sub>4</sub><sup>2-</sup> > Cl<sup>-</sup> > NO<sub>3</sub><sup>-</sup> > Na<sup>+</sup> > K<sup>+</sup> > Mg<sup>2+</sup> in winter listed in Table 2 (Zhang et al., 2008), contents of all water-soluble ions were roughly one- to nine-fold higher in winter, except Ca<sup>2+</sup>.

Na<sup>+</sup>, Mg<sup>2+</sup> and Ca<sup>2+</sup> in aerosols of northwest China are known to derive mainly from crustal sources, such as re-suspended soil particles, and are strongly influenced by dust storms in spring and early summer. SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> in aerosols are believed to originate from secondary particles produced by the transformation of their gaseous precursors: SO<sub>2</sub>, NO<sub>2</sub> and NH<sub>3</sub>, mainly emitted by anthropogenic sources and partially from natural sources (Wang et al., 2006). K<sup>+</sup> and Cl<sup>-</sup> are detected in biomass burning particles, and aerosol potassium has been regarded as a common useful tracer of biofuel combustion and biomass burning emissions (Dibb et al., 1995; Dabell et al., 2004; Liu et al., 2000; Hsu et al., 2009; Duan et al., 2004).

Data from the 923 samples obtained during field campaign were analyzed by IBM SPSS Statistics 19 to derive the Pearson correlation coefficients (PCC) among water-soluble ions. The PCCs are shown in Table 3 and Fig. 2; we can see high PCCs among Na<sup>+</sup>, Mg<sup>2+</sup> and Ca<sup>2+</sup>; NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>; K<sup>+</sup> and Cl<sup>-</sup>. Moreover, three predominant

Table 2 Comparison of water-soluble ions in PM<sub>10</sub> between summer and winter (unit: nmol/L)

	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>	Ca <sup>2+</sup>	Na <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>
Summer	0.087	0.072	0.070	0.160	0.460	0.022	0.012	0.053
Winter (Zhang et al., 2008)	0.354	0.229	0.333	1.055	0.395	0.192	0.106	0.070
Winter/Summer	4.076	3.183	4.775	6.598	0.859	8.545	8.923	1.324



**Fig. 2** Time series of levels of water-soluble ions, crust, biomass burning and secondary aerosol in PM<sub>10</sub> during whole field campaign. (MARGA offline for maintenance in Apr 22–May 6)

**Table 3** Pearson correlation coefficients between major ions during whole field campaign

	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>
Cl <sup>-</sup>	1	0.556**	0.366**	0.629**	0.539**	0.766**	0.186**	0.274**
NO <sub>3</sub> <sup>-</sup>		1	0.437**	0.321**	0.768**	0.516**	0.050	0.016
SO <sub>4</sub> <sup>2-</sup>			1	0.319**	0.702**	0.442**	0.264**	0.170**
Na <sup>+</sup>				1	0.178**	0.465**	0.513**	0.609**
NH <sub>4</sub> <sup>+</sup>					1	0.607**	-0.011	-0.122
K <sup>+</sup>						1	0.073*	0.091**
Mg <sup>2+</sup>							1	0.752**
Ca <sup>2+</sup>								1

\* $p = 0.05$ ; \*\* $p = 0.01$

salts with their PCCs between cation and anion were: NH<sub>4</sub>NO<sub>3</sub> (0.768), KCl (0.766), (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>/NH<sub>4</sub>HSO<sub>4</sub> (0.702). Examining KCl, the reason why the PCC 0.766 was lower than the value of 0.923 reported by Du et al. (2011) in Shanghai, was caused by heavier biomass burning pollution in east China during summertime (Li et al., 2010) and the diversity of chlorides (KCl (0.766), NaCl (0.629), NH<sub>4</sub>Cl (0.539)) in PM<sub>10</sub> of Lanzhou as shown in **Table 3**.

PMF is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into two matrices, factor contributions and factor profiles, which then need to be interpreted by an analyst as to what source types are represented using measured source profile information, wind direction analysis, and emission inventories. The method is described in greater detail elsewhere (Paatero and Tapper, 1994; Paatero, 1997). We used EPA-PMF 3.0 to apportion sources of water-soluble in PM<sub>10</sub>. **Figure 3** shows the

model execution result with source profiles. The different sources were classified in three groups: representative of crust (Na<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>), secondary aerosol (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>) and biomass burning (K<sup>+</sup>, Cl<sup>-</sup>), and the mass contribution to PM<sub>10</sub> of each source was 7.3%, 5.7%, and 1.1% respectively, which resolved 14.1% of PM<sub>10</sub> mass concentration. It was remarkable that the biomass burning source (1.1%) was much lower than crust (7.3%) and secondary aerosol sources (5.7%) because the ground fire points, which were detected by the Moderate Resolution Imaging Spectroradiometer (MODIS) on Terra and Aqua during Apr 1 to May 24 (**Fig. 4**), were small compared with southeast China, where agriculture is much more advanced and influenced by biomass burning pollution in summer. (Li et al., 2010) As shown in **Fig. 2d**, the molar concentration of biomass burning species in PM<sub>10</sub> exhibited moderate variations compared with the other two, indicating that biomass burning pollution played an

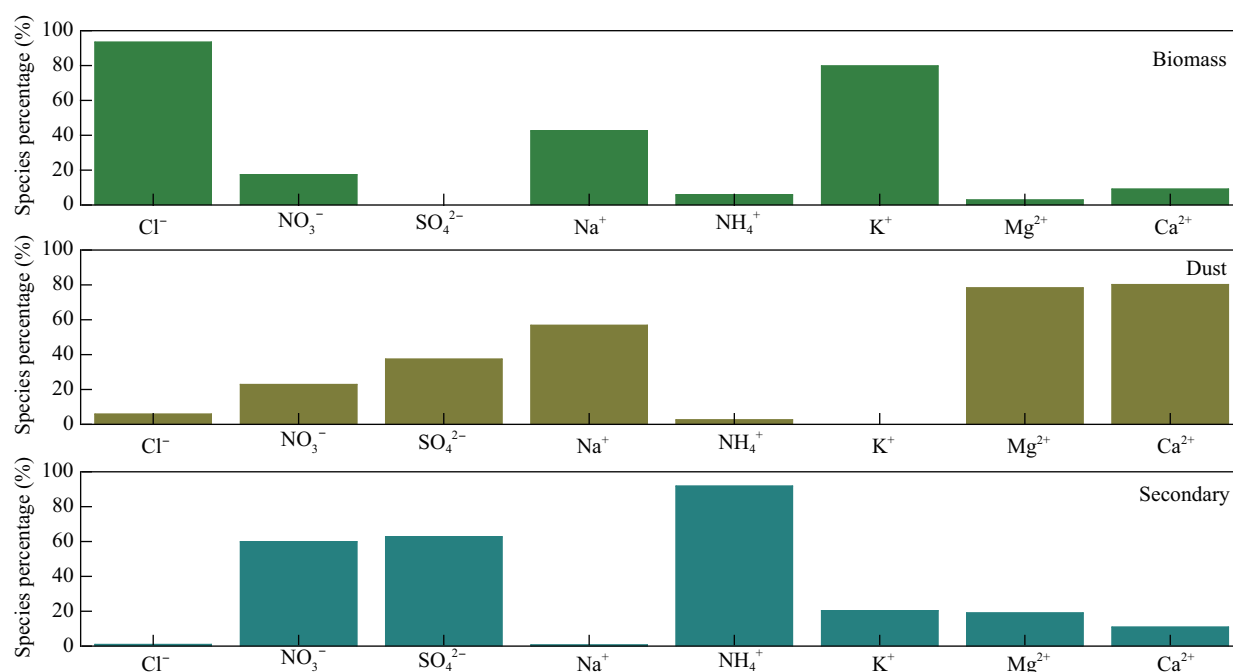


Fig. 3 Factor profiles from execution of EPA-PMF 3.0.

insignificant role in the formation of air pollution events in Lanzhou in early summertime.

According to the source apportionment result (Fig. 3) and time series shown in Fig. 2d, the primary pollutant (PM<sub>10</sub>) in the five air pollution events (AQI > 100) was classified into crustal species- and secondary aerosol-induced. The average molar concentrations of water-soluble ions in PM<sub>10</sub> during these two types of pollution events are presented in Table 4. The content of each water-soluble ion per air volume in the periods of air pollution days was almost always higher than on clear days; as shown in Table 4, molar concentrations of water-soluble ions showed drastic changes of roughly one- to three-fold between air pollution and clear days.

## 2.3 Formation mechanism of pollution events

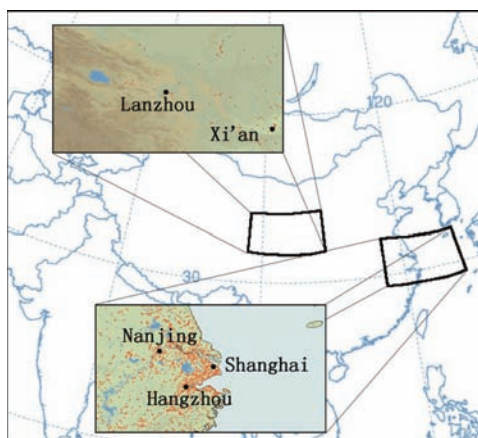
### 2.3.1 Crustal species-induced pollution

Lanzhou is located in an arid and semi-arid area of China where Taklamakan, Gurbantunggut, Tsaidam Basin, Kumutage, Badain Juran, Tengger, Ulan Buh, Hobq, Mu Us and other smaller deserts are distributed from west to east (Fig. 5). In spring and early summer, sand-dust weather influenced Lanzhou severely, and crustal species flew up in the air to reduce visibility (Wang et al., 1999; Liu et al., 2004; Ta et al., 2004). Na<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup> content in PM<sub>10</sub> are excellent indicators to use in estimating crustal species emission and tracing long-range transport of aerosol from natural sources in the atmosphere (Arimoto et al., 2004).

Table 4 Mean concentrations of water-soluble ions in PM<sub>10</sub> during the air pollution events and clear days (unit: nmol/L)

	Date	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>
Crust	7 Apr	0.056	0.045	0.110	0.047	0.114	0.010	0.103	0.842
	10 Apr	0.068	0.042	0.060	0.034	0.021	0.003	0.079	0.840
	20–21 Apr	0.027	0.047	0.065	0.019	0.060	0.008	0.070	0.634
	18 May	0.067	0.033	0.057	0.049	0.038	0.018	0.060	0.574
Secondary	1–3 Apr	0.079	0.084	0.152	0.028	0.227	0.012	0.050	0.494
	8–9 Apr	0.170	0.125	0.116	0.048	0.262	0.017	0.064	0.520
	11–13 Apr	0.151	0.109	0.101	0.032	0.199	0.017	0.075	0.665
	15–17 Apr	0.108	0.088	0.102	0.023	0.134	0.011	0.048	0.445
Ave	–	0.091	0.072	0.095	0.035	0.132	0.012	0.069	0.627
Clear days (CD)	12 May; 21–24 May	0.036	0.079	0.068	0.011	0.185	0.010	0.038	0.314
Ave/CD	–	2.528	0.912	1.394	3.198	0.713	1.232	1.784	1.993

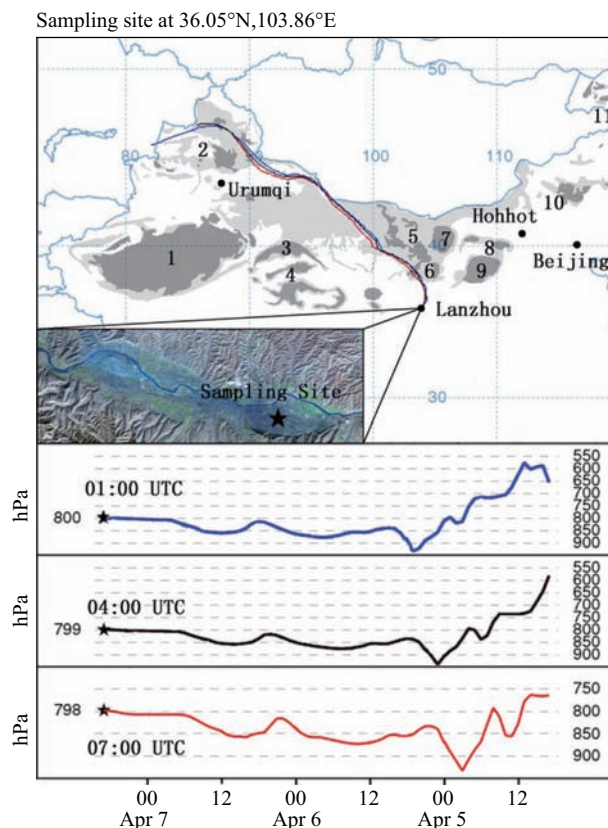




**Fig. 4** Comparison of ground fire points between Lanzhou and Yangtze River delta. (Orange dots represent fire points on the ground detected by MODIS on Terra and Aqua).

Pollution days caused by crustal species are listed in **Table 4**. Molar concentrations of crustal species occurred on Apr 7 with a value of 0.991 nmol/L, higher than secondary aerosol pollution (0.269 nmol/L) and biomass burning (0.067 nmol/L); this was a typical sand-dust day caused by long-range transport. The NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model and meteorological data GDAS were used to study the formation mechanism of this typical event. More information about HYSPLIT and GDAS can be found at the website: <http://ready.arl.noaa.gov/hysplit-bin/trajasc.pl>. The HYSPLIT model was applied to trace the air parcel 3-days back, and 300 m (above ground level) was selected as the simulation altitude because air pollution at this height was mainly influenced by large-scale long-range transport. Moreover, 300 meters also represented the air flow near the ground, but reduced the influence of ground surface friction (Wang et al., 2009). The backward trajectory for the receptor site of Lanzhou is shown in **Fig. 5**. In order to track the whole process of this typical sand-dust weather event, the trajectory end time was set to 01:00 UTC Apr 7 (09:00 local time, LT hereafter), 04:00 UTC Apr 7 (12:00 LT) and 07:00 UTC Apr 7 (15:00 LT) respectively. As shown in **Fig. 5**, the air parcel outflowed in a northwesterly pathway, traveled a distance over the Gobi desert, and passed through Hexi Corridor arriving at the sampling site, showing the features of a sand-dust event that was induced by long-range transport. The result was similar with a previous observation by Tao et al. (2007) in Lanzhou, who indicated that the Hexi Corridor was the major passageway for sand and dust from northwest to south and east.

Besides long-range transport, sand-dust pollution could also be induced by local emission. Construction sites in urban districts are the main local sources of crustal species. Strong wind could combine long-range transport and local emission, increasing the adverse impact on the city environment (Ye et al., 2000).

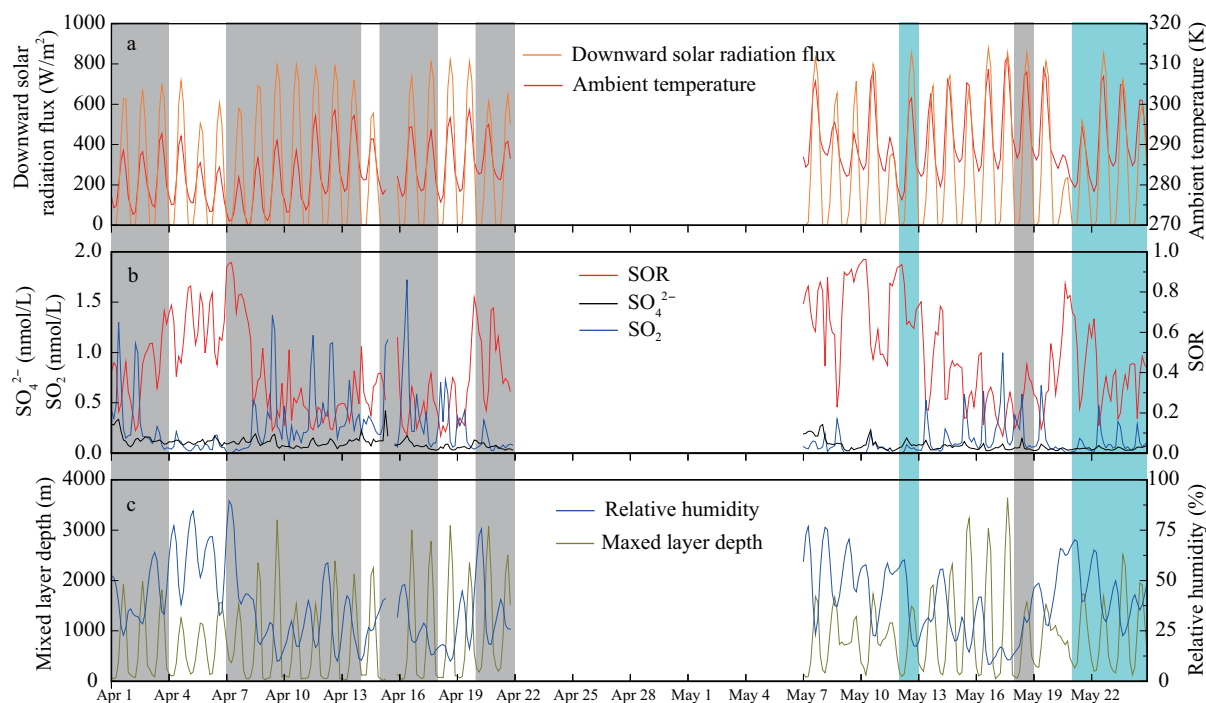


**Fig. 5** NOAA HYSPLIT Model result with GDAS meteorological data. Back trajectory ending at 01:00, 04:00, 07:00 UTC Apr 7. (Dark gray: 1. Taklimakan; 2. Gurban Tonggut; 3. Kumtag; 4. Qaidam; 5. Badain Jaran; 6. Tengger; 7. Ulan Buh; 8. Qubqi; 9. Mu Us; 10. Ortindag; 11. Hulun Buir. Light gray: Gobi desert)

### 2.3.2 Secondary aerosol pollution

High concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  were representative of the air pollution days due to secondary aerosol pollution, covering the periods of Apr 1 to 3, Apr 7 to 8, Apr 11 to 13 and Apr 15 to 17 listed in **Table 4**. Three kinds of salts with their PCCs between cation and anion were  $\text{NH}_4\text{NO}_3$  (0.768),  $(\text{NH}_4)_2\text{SO}_4/\text{NH}_4\text{HSO}_4$  (0.702) and  $\text{NH}_4\text{Cl}$  (0.539) in  $\text{PM}_{10}$  as shown in **Table 3**. Semi-volatile  $\text{NH}_4\text{NO}_3$  and  $\text{NH}_4\text{Cl}$  are formed via reversible phase equilibrium with  $\text{NH}_3$ ,  $\text{HNO}_3$ , and  $\text{HCl}$  (Pio and Harrison, 1987) and the main form of  $\text{NO}_3^-$  in the fine mode was  $\text{NH}_4\text{NO}_3$  (Pierson and Brachaczek, 1988).  $\text{NH}_3$  was emitted from livestock and ammonia synthesis; more importantly,  $\text{NH}_3$  in the atmosphere is a major gas-phase neutralizing agent (Krupa, 2003). At lower concentrations,  $\text{NH}_3$  would primarily react with  $\text{H}_2\text{SO}_4$  and generate  $(\text{NH}_4)_2\text{SO}_4/\text{NH}_4\text{HSO}_4$ .

$\text{SO}_2$  was emitted by fossil fuel combustion, and the formation of  $\text{SO}_4^{2-}$  from  $\text{SO}_2$  usually occurred by aqueous- and gas-phase oxidation reaction with OH radical (Meng and Seinfeld, 1994). Therefore, the chemical formation of  $\text{SO}_4^{2-}$  from  $\text{SO}_2$  should not be neglected in these secondary aerosol pollution days. The sulfur oxidation ratio ( $\text{SOR} =$



**Fig. 6** Variation of meteorological factors and sulfur oxidation ratio (SOR) during the field campaign.

$n\text{-SO}_4^{2-}/(n\text{-SO}_4^{2-}+n\text{-SO}_2)$  while,  $n$  is molar concentration) is an available indicator to quantitatively characterize the secondary transformation reactions of  $\text{SO}_2$ . The time variation of SOR calculated from the 923 samples and meteorological factors during the field campaign is shown in **Fig. 6**. The average value of SOR was  $0.30 \pm 0.02$  during the secondary aerosol pollution days, compared with previous observation by Fu et al. (2008) and Yao et al. (2002). SOR above 0.10 implied sulfate was mainly produced through the transformation of  $\text{SO}_2$  by oxidation. As seen in **Fig. 6**, the average value of SOR in Apr 1 to 3, which were typical secondary pollution days, was 0.40, lower than on clear days (0.57) and crust species pollution days (0.84). This result indicated that SOR could not appear alone, instead, the concentrations of  $\text{SO}_4^{2-}$  and  $\text{SO}_2$  must be presented with SOR simultaneously. For example, although SOR (0.40) was lower on secondary aerosol pollution days than on clear days (0.57), the molar concentrations of  $\text{SO}_4^{2-}$  and  $\text{SO}_2$  during those days were 3 and 4 times higher than on clear days. High  $\text{SO}_2$  concentration and high SOR directly resulted in a large amount of secondary sulfate particles in the atmosphere. Meanwhile, meteorological factors also played a key role in the formation processes of these secondary aerosol pollutants. From **Fig. 6**, it was remarkable that high positive correlation occurred between RH and SOR, and we could conclude that higher RH, lower MLD and lower ambient temperature were conducive to the formation of secondary pollution. Low MLD was favorable for gaseous and particulate pollutants to accumulate in the atmospheric boundary layer and persist for a long time, and higher RH could

offer the proper conditions for heterogeneous reaction on pre-existing particles. Furthermore, high downward solar radiation flux could promote the photolysis of HONO by the equation:  $\text{HONO} + h\nu = \text{OH} + \text{NO}$  ( $300 \text{ nm} < \lambda < 405 \text{ nm}$ ) (Alicke et al., 2003), and OH radical provided strong atmospheric oxidation ability and accelerated gas-to-particle transformation.

### 3 Conclusions

Urban pollution events ( $\text{AQI} > 100$ ) were monitored in Lanzhou during early summer from Apr 1 to May 24, 2011. An online instrument (MARGA) was deployed to measure hourly concentrations of major water-soluble inorganic ions ( $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ) in aerosols and trace gases (HCl, HONO,  $\text{SO}_2$ ,  $\text{HNO}_3$ ,  $\text{NH}_3$ ) in the atmosphere. Compared with Lanzhou wintertime, water-soluble ions were roughly one- to nine-fold higher except for  $\text{Ca}^{2+}$ , revealing that Lanzhou is more influenced by sand-dust during early summer and heavy secondary aerosol pollution in winter. Based on different concentration levels of water-soluble ions and the result of EPA-PMF 3.0 modeling, the pollution events were classified as crustal species-induced and secondary aerosol-induced pollution. Air parcels containing crustal species outflowing from northwest of Lanzhou traveled a distance over the Hexi corridor and elevated concentrations of  $\text{Ca}^{2+}$ ,  $\text{Na}^+$  and  $\text{Mg}^{2+}$  in  $\text{PM}_{10}$  during a typical sand-dust day. Meteorological data and concentrations

of  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{SO}_2$  were analyzed to reveal the formation mechanism of secondary aerosol pollution. SOR was derived from the concentration of  $\text{SO}_4^{2-}$  and  $\text{SO}_2$ ; it had strong positive correlation with relative humidity, indicating that the processes of gas-to-particle transformation were mainly influenced by the relative humidity in the Lanzhou early summer. Moreover,  $\text{NH}_4\text{NO}_3$ ,  $(\text{NH}_4)_2\text{SO}_4/\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{Cl}$  were common salts in secondary aerosols.

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