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Identifying anthropogenic and natural influences on extreme pollution of respirable suspended particulates in Beijing using backward trajectory analysis

Yu Song, Weijie Miao, Bing Liu, Wei Dai, Xuhui Cai*

Department of Environmental Science, Peking University, Beijing 100871, China Received 27 July 2006; received in revised form 16 October 2007; accepted 16 October 2007 Available online 24 October 2007

Abstract

In China, daily respirable suspended particulate (RSP, particles with aerodynamic diameters less than $10 \,\mu$ m) concentrations exceeding 420 μ g m⁻³ are considered "hazardous" to health. These can lead to the premature onset of certain diseases and premature death of sick and elderly people; even healthy people are warned to avoid outdoor activity when RSP concentrations are high. Such high pollution levels are defined as extreme RSP pollution events. Recent epidemiological studies have shown that a distinct difference exists between the health effects caused by natural sources and anthropogenic sources, mandating knowledge of the source of extreme RSP pollution. Twenty-six extreme RSP pollution events were recorded in Beijing from January 2003 to December 2006. The HYSPLIT4 (Hybrid Single Particle Lagrangian Integrated Trajectory) model (Version 4) was used to discriminate the sources of these extreme RSP pollution events. The model found that twelve events were caused from natural sources (dust storms), nine events from anthropogenic sources (e.g., vehicles and industrial activities, etc.) under quasi-quiescent weather, and five events were from mixed causes. Identifying such events will be valuable in epidemiological studies on air pollution in Beijing. © 2007 Elsevier B.V. All rights reserved.

Keywords: Respirable suspended particulates; Extreme pollution; China; HYSPLIT4 model; Epidemiological study

1. Introduction

Although epidemiologic studies have found a strong association between fine particles and mortality [1,2], a recent systematic review based on existing studies showed that coarse particulate matter (PM; particles with an aerodynamic diameter >2.5 μ m and <10 μ m) has stronger, or as strong, short-term effects as fine PM in cases of chronic obstructive pulmonary disease, asthma, and respiratory hospital admissions [3]. This also reinforced many conclusions of the EPA in their final staff paper, and bolstered calls for the establishment of a new National Ambient Air Quality Standard for coarse PM [4]. Hence, further research on respirable suspended particulates (RSP, particles with aerodynamic diameters <10 μ m) is still needed and should be enhanced.

RSP pollution is a serious problem in Beijing. Monitoring data from the last decade indicated that annual RSP concentrations in Beijing can reach $140-160 \,\mu g \,m^{-3}$ [5]. The Chinese

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government provides an API (air pollution index) each day for the public. A pollution API exceeding 300 is defined as "hazardous"; all people should avoid outdoor activity at APIs between 300 and 400, and should remain indoors keeping windows and doors closed at APIs between 400 and 500. Such serious pollution events do occur in Beijing, although the frequency is less than 5%. Furthermore, in all such events, RSP had the highest API, or was the primary pollutant. A daily RSP concentration exceeding $420 \,\mu g \, m^{-3}$ is considered "hazardous". In this study, we defined this as an extreme pollution event of RSP (RSP-EPE). Schwartz [6] showed that PM pollution at extremely high concentrations, for example, London in 1952, the Meuse valley in 1930, and Donoroa, Pennsylvania in 1948, was strongly associated with deaths. PM can be emitted from anthropogenic sources (e.g., coal burning, vehicle exhausts, heavy oil combustion) and natural sources (e.g., dust, biomass burning). Recent epidemiological studies have shown that differences in the chemical composition of PM have significantly different health effects. Schwartz et al. [7] found no increase in total mortality on dust storm days; however, Kwon at al. [8] and Chen et al. [9] provided weak evidence that

^{*} Corresponding author. Tel.: +86 10 62751924; fax: +86 10 62751927. *E-mail address:* xhcai@pku.edu.cn (X. Cai).

Table 1

The dates of API (averaged over the nine substations in urban areas) exceeding 300 from January 2003 to December 2006

Number	Date	API	Number	Date	API
1	1-11-2003	476	14	4-11-2005	334
2	19-2-2004	452	15	5-11-2005	469
3	10-3-2004	442	16	21-1-2006	308
4	28-3-2004	333	17	10-3-2006	407
5	7-10-2004	307	18	27-3-2006	332
6	8-10-2004	378	19	9-4-2006	500
7	10-10-2004	353	20	10-4-2006	489
8	3-12-2004	325	21	17-4-2006	500
9	15-12-2004	303	22	19-4-2006	314
10	5-4-2005	335	23	1-5-2006	435
11	6-4-2005	357	24	17-5-2006	490
12	28-4-2005	420	25	21-11-2006	416
13	3-11-2005	336	26	12-12-2006	499



Fig. 1. Nine monitoring substations (dots) of the eight districts in the main urban areas of Beijing: Xicheng (XC), Dongcheng (DC), Xuanwu (XW), Chongwen (CW), Haidian, Shijingshan (SJS), Chaoyang, and Fengtai.



Fig. 2. Emission amount (in tonnes year⁻¹) of anthropogenic RSP near Beijing. Regional data from Streets et al. [26], with 6-min resolution updated from the TRACE-P emissions inventory.

Asian dust storms were associated with increased risk of mortality in Seoul, Korea and Taipei, Taiwan. Anthropogenic PM may be increased after dust is transported long distances; for example, quantities of elements such as secondary aerosols and water-soluble ions increase during dust events [10]. More epidemiologic studies with source apportionment results showed that coal combustion, vehicle exhaust, and secondary sulfate in PM were strongly associated with respiratory deaths, but crustal particles and biomass burning were not [11–15]. Thus, distinguishing RSP-EPEs originating from natural source or from anthropogenic accumulations is necessary because of their distinct health effects.

A better way to identify RSP-EPEs is by using chemical compositions [16,17], as crustal elements are often dominant in dust, and sulfate, nitrate, and organic/elemental carbon

Table 2

Maximum and minimum wind speeds $(m s^{-1})$ averaged along the 72-h backward trajectories, starting at each hour from noon to noon, with the air pollution types

Date	Maximum speed	Minimum speed	Pollution type	Date	Maximum speed	Minimum speed	Pollution type
1-11-2003	3.1	1.8	2	4-11-2005	3.5	2.2	2
19-2-2004	7.7	3.9	1	5-11-2005	3.2	1.6	2
10-3-2004	21.7	4.2	1	21-1-2006	1.0	0.9	2
28-3-2004	16.9	3.0	3	10-3-2006	18.0	3.2	3
7-10-2004	3.1	2.3	2	27-3-2006	15.4	5.7	1
8-10-2004	2.9	2.1	2	9-4-2006	10.9	4.6	1
10-10-2004	1.8	1.7	2	10-4-2006	4.8	3.8	3
3-12-2004	3.3	1.7	2	17-4-2006	7.2	4.1	1
15-12-2004	11.4	2.6	1	19-4-2006	12.1	2.4	1
5-4-2005	6.3	3.0	1	1-5-2006	14.5	3.0	1
6-4-2005	6.1	4.4	3	17-5-2006	23.1	4.5	3
28-4-2005	14.6	6.3	1	21-11-2006	7.9	1.9	1
3-11-2005	3.5	2.6	2	12-12-2006	10.8	4.3	1

Note: 1 is a dust storm, 2 is local accumulation and 3 is a mixed case.

dominate anthropogenic emissions. Chemical data, however, are not always available and analysis is often time-consuming. Finding a simple way to identify sources from the available data for RSP-EPEs is needed. Although remote sensing data, especially MODIS (Moderate Resolution Imaging Spectroradiometer) aerosol data contain aerosol type values, and have been used to show the extent and types of PM pollution, especially for dust storms [18–20],



Fig. 3. Twelve cases of RSP-EPE caused by dust storms; 72-h backward trajectories were computed from noon (previous) to noon for each hour at 50 m AGL.



Fig. 3. (Continued).



Fig. 4. Nine cases of RSP-EPE caused by local accumulation; 72-h backward trajectories were computed from noon (previous) to noon for each hour at 50 m AGL.



Fig. 4. (Continued).

the data may be weak because they rely on polar orbital satellites that only provide snapshot data for a single time period. Daily RSP concentrations are often averaged over 24 h. Moreover, remote sensing data are not always available due to the obstructive effects of clouds.

In this study, we sought to find a procedure to identify the RSP-EPEs for 2003–2006 in Beijing, based on backward trajectory analysis.

2. Methods

2.1. Air pollution data

The BJEPB records hourly RSP mass concentration using TEOM particulate monitors. A pump, controlled by a programmable timer, is turned off and on at preset times, generating an airflow rate of 16.7 L/min. API data (calculated from RSP concentrations from noon to noon) were obtained from the BJEPB Web site (http://www.bjepb.gov.cn/). Table 1 lists API values of RSP-EPEs, averaged from the nine monitoring substations located in the center of Beijing (Fig. 1), from 4 years, 2003–2006. Twenty-six RSP-EPEs were found. Although the API frequency was only 1.8% in the 4 years, it would be much higher if the criteria were relaxed to include the API exceeding 300 from any one of the nine substations. The total numbers of RSP-EPEs were 13, 3, and 10 in spring, autumn, and winter, respectively. No events were found in summer because this is the rainy season, and most PM in ambient air are washed out.

2.2. The HYSPLIT4 model

Backward trajectories to show the pathways of air parcels transporting particles were calculated from the HYSPLIT4 (Hybrid Single Particle Lagrangian Integrated Trajectory, Version 4) model [21] with FNL meteorological data. This model was developed by NOAA/ARL (US National Oceanic and Atmospheric Administration/Air Resources Laboratory). The back trajectories were started from Beijing (39.8°N, 116.47°E). Although initial heights of 500 m AGL (above ground level) provide a good representation of airflow of the atmospheric boundary layer (typical height, 1000 m AGL), we found that backward trajectories starting from 50 m AGL within the surface layer better reflect the movement pathways of local or regional pollutants. Twenty-four backward trajectories were computed each hour, from noon to noon, starting at 50 m AGL.

3. Results and discussion

3.1. Dust storms

Many recorders have shown that dust storms can lead to high RSP pollution in Beijing [5]. The dust source region consists of deserts, including the Gobi in northern China and southern Mongolia. This source region comprises two parts: the Mongolian Plateau system, including deserts and the Gobi desert on the Mongolian Plateau and its south extension, the Ordos and Alxa plateaus; and the Tarim Basin system, including deserts and the Gobi desert in the Tarim Basin, Junggar Basin, and areas to the east [22,23]. In late winter and spring, when strong frontal winds, or cold waves from Siberia frequently flow through the dust area, a huge amount of dust is lifted from the dry surfaces, forming severe dust storms, and some of these arrive in Beijing [22]. Sand saltation before entering the ambient air requires momentum provided by the wind, or the dust raised above a threshold wind speed or a threshold friction velocity (e.g., In and Park [24]). The threshold wind speed in Asia is $6-10 \text{ m s}^{-1}$ [25]. In this study, we computed the average wind speeds along the 24 trajectories. The maximum wind speed is an indicator of whether a dust storm will occur.

Note that the effects of anthropogenic emissions can be neglected because the local pollution is removed by strong advection when a dust storm passes Beijing. Moreover, the emission inventory showed that the amount of RSP emissions located along the pathways from dust sources to Beijing (e.g., Inner Mongolia) was small (Fig. 2). Previous research found that during dust storms, gaseous pollutants had low concentrations and PM contributions from anthropogenic emissions; that is, emissions from traffic exhaust, coal combustion, and secondary products were small [16].

Identifying a RSP-EPE caused by a dust storm is relatively simple. The backward trajectories of a dust storm event should pass through the dust sources (north or northwest of Beijing) and the wind speeds along the trajectories should be very large. Twelve RSP-EPEs could be identified as caused by dust storms (Table 2 and Fig. 3). All of these passed the dust sources at high wind speeds. Under such strong winds, as noted above, local pollutants emitted near Beijing are blown away. As the wind flows were realistically complex, some dust storms did not sweep directly into Beijing, but turned into Beijing when they met airflows from southern or eastern directions, e.g., on February 19, 2004, April 4, 2006, and April 17, 2006.

3.2. Accumulation under quasi-quiescent weather

Nine RSP-EPEs were caused by local/regional accumulation under quasi-stagnant air (Table 2 and Fig. 4). They could be identified for two reasons. First, the wind speeds along the backward trajectories in the nine cases (Table 2) did not reach the threshold value to raise the dust from the surface, and the pathways did not pass through the dust sources; thus, these nine cases were excluded from the dust storm data. Second, the minimum wind speeds along the 72-h backward trajectories were small, which implied weak advection. All of the pathways passed from the local/regional regions, especially from south of Beijing, where high anthropogenic emissions were located (Fig. 2). During the nine RSP-EPEs, except for the event on December 3, 2004, we found that the high RSP concentration gradually accumulated in two stages.

In the early stage, weak synoptic systems dominated north of China. The weather in northern China is often dominated by continental dry and cold air masses; in particular, cold Siberian highs frequently intrude into northern China. Usually, after a cold front passes this area, the continental air becomes denaturalized and the weather is controlled by quasi-stationary highs. At this stage, quasi-quiescent weather accompanied by low wind speeds can lead to weak advection and a greater accumulation of pollutants, especially as local emissions begin to accumulate.

Subsequently, the weather in most of the North China Plain is dominated by weak, southerly or southwesterly winds, when the vast areas, including most of the North China Plain, lie in front of a succeeding cold front. Such a cold front is usually part of a mid-latitude cyclone that forms in the northwest of China and moves east to Beijing. Under this condition, the air near Beijing is often relatively warm and humid. The persistent weak winds bring the anthropogenic pollutants from the south or southwest (Fig. 2) into Beijing.

However, the case of December 3, 2004 is quite special (Fig. 4). During the early stage (from December 1 to 2), Bei-



Fig. 5. The 72-h backward trajectories commencing at each hour from 13:00, November 2 to 12:00, November 3, 2005 in Beijing (39.8° N, 116.47° E) at 50 m (solid line) and 500 m (dash line) AGL.

jing was at the rear end of a high pressure system located in the east. Beijing was dominated by weak westerly winds when the rear of the high pressure swept the area. After it passed, typhoon Nanmadol arrived from the east of the South China Sea, and Beijing was at its northern edge. The dominant winds in Beijing were from the east. The high RSP pollution was caused by local accumulation under the weak winds.

Fig. 6. Five mixed cases of RSP-EPE; 72-h backward trajectories were computed from noon to noon for each hour at 50 m AGL.

Note that an initiating height of 50 m AGL characterizing the lower atmospheric flow may better represent the accumulation of air pollutants. Fig. 5 shows backward trajectories starting from 50 and 500 m AGL for the RSP-EPE on November 3, 2005. The airflows in the upper boundary layer were blown from the west at relatively higher speeds, which benefited pollution dispersion; however, the flows within the surface layer were from the southwest and at much lower speeds, which helped to transport the anthropogenic emissions (Fig. 2) from the southwest to Beijing. The pollutants were gradually transported and accumulated in Beijing.

3.3. The mixed case

The daily RSP concentrations (from noon to noon) were averaged from 24-h concentrations. When a sandstorm sweeps Beijing, part or all of the hourly records can reflect its effects. Moreover, the lifetime of a sandstorm remaining over China is often 2-3 days. Beijing may repeatedly suffer from the same sandstorm if it returns through the circumfluence, in which the wind speeds are often small. Thus, the mixed RSP-EPEs contain two types: Type I, in which the 24-h concentrations record part of a sandstorm and part of the anthropogenic effect; Type II, in which the sandstorm returns to Beijing, and it experiences high anthropogenic emissions. Three Type 1 events occurred on March 28, 2004, March 10, 2006, and May 15, 2006 (Fig. 6). Two Type II events occurred on April 6, 2005 and April 10, 2006 because sandstorms appeared in Beijing on the 2 previous days and returned from the south and northeast of Beijing, respectively (Fig. 6). Both regions had high RSP emissions (Fig. 2).

4. Conclusion

Although extreme RSP pollution events (API > 300) seldom occur in Beijing, they may cause premature deaths of sick and elderly people. Both dust events, especially sandstorms, and PM accumulation from local or regional anthropogenic sources under calm conditions can lead to extreme pollution events. However, they have distinctly dissimilar effects on human health due to differences in chemical composition. In our study, we collected 26 extreme RSP pollution events in Beijing for the 2003-2006 period and computed the 72-h backward trajectories staring at 50 m AGL using the HYSPLIT4 model. After analyzing the pathways of backward trajectories, dust source locations and anthropogenic emission inventory results, and wind speeds estimated along the trajectories, we found that 12 events were caused by dust storms, nine events were due to accumulation of anthropogenic emissions, and five events were mixed cases. These results could be helpful in epidemiological studies of air pollution, especially using the case-crossover method.

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