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Characteristics of ambient 1-min $PM_{2.5}$ variation in Beijing

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Abstract One-minute $PM_{2.5}$ concentration was obtained with LD-5C pocket microcomputer laser dust instrument from Dec. 15th, 2005 to Jan. 16th, 2006 and Mar. 17th to Apr. 28th, 2006 in Beijing. The concentration of SO_2, NO_2, O_3, CO, and PM_{10} from Jan. 1st, 2001 to Dec. 31st, 2004 were obtained from the conversion of air pollution index. Results showed that all the pollutants showed cyclic characteristics. The longer yearly cycles was shown from SO_2, NO_2, O_3, CO, and PM_{10}, as the sampling time was 4-year long and daily collected. The shorter hourly and daily cycle was shown from 1-min $PM_{2.5}$, as the sampling time was about 1-month long and one collected at 1 min. The spectral density analysis confirmed this from the periodogram graphs. The longer yearly cycle (365, 180 days), the seasonal cycle (120, 60–90 days), and monthly cycle (21, 23, 27 days) of SO_2, NO_2, CO, O_3, and PM_{10} were obviously shown. In addition, the shorter weekly cycle of 5–7 days is obviously shown, too. The shorter hourly cycle (8–12, 4–6, 3, 1–2 h, 20 min) of 1-min $PM_{2.5}$ was also indicated from spectral density analysis. Two major factors contribute the 1-min $PM_{2.5}$ cycles, i.e., the meteorological factors and source effects. Both the relative humidity and dew point showed consistent variation with $PM_{2.5}$, but the wind speed showed inverse variations with $PM_{2.5}$. Furthermore, the spectral density analysis of the meteorological factors (4–5, 2–2.5, 1–1.5 days, 12, 6–8, 3 h) may partially explain the cycles of $PM_{2.5}$. As for the sources effects, it can be shown from the strong dust storm of April 16–18th, 2006. $PM_{2.5}$ constantly increased tens and even hundreds of times high concentration within a few minutes due to the intensity of the dust sources.

Keywords One–minute $PM_{2.5}$ · Meteorological effects · Dust storm · Cycles

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Introduction

Aerosol is now receiving worldwide attention, as it would have the potential impacts on the global

climate change and the effect on the health to human being all over the world. Aerosols also affect climate directly by scattering and absorbing radiation or indirectly by changing the depth and albedo of the clouds (Twomey 1974; Charlson et al. 1992). Atmospheric particulates, especially those fine particles, have been recognized to have serious health effects, and the level in ambient air are associated with increased cardiovascular and respiratory morbidity and mortality (Dockery et al. 1993). However, it is not yet known whether the health effects may be caused by the large number of particles (Penttinen et al. 2001) or perhaps due to some toxic components in the particulate matter. Thus, many studies of their physical and chemical characteristics are conducted to elucidate particle toxicity.

Aerosol particulate concentrations in urban areas are affected by several factors. Exhaust and suspension emission originating from local traffic are considered to be a major factor that contributes the short term and high concentration of the urban aerosol (Hussein et al. 2004). Besides vehicle-originated emissions, long-range transportation and coal burning especially in winter are also two most important contributors.

Studies on the characteristics of aerosol in Beijing have been conducted since 1980s of the twentieth century. Winchester and Bi (1984) have studied the fine and coarse particle compositions in Beijing in the late 1970s. Dust-soil, industry emission, coal burning, vehicle exhaust emission, and waste incineration were thought to be the major sources of particulate pollution at Beijing (Sun et al. 2004a). Chen et al. (1994) initiated the $PM_{2.5}$ (particles with diameters < 2.5) aerosol in Beijing, investigated the chemical compositions of $PM_{2.5}$, and observed that OC, EC, and sulfate, the three major components of $PM_{2.5}$, contributed 66% of the total fine particles, which had the yearly average of 89.7 $\mu g/m^3$ in Beijing. He et al. (2001) observed $PM_{2.5}$ concentrations of 37–357 $\mu g/m^3$ with significant seasonal variation and concluded that organic carbon was the most abundant species in $PM_{2.5}$ while sulfate, nitrate, and ammonium also were the major components. Yao et al. (2002) reported that sulfate, nitrate, and ammonium were the main ionic species in $PM_{2.5}$. Wang et al. (2005a) reported that temperature, relative humidity, rainwater frequency, and air mass origin might be the main factors regulating the $PM_{2.5}$ aerosol distribution by studying water soluble ions in different sites in Beijing.

However, most of these studies concentrated on the physicochemical species characterization of $PM_{2.5}$. Few studies have been conducted on the variation of aerosols during 1 day or shorter time. As a typical city of the mixing between coal burning and vehicle exhausts, Beijing is thought to be a good example of pollution. This study presents the characteristics of 1-min $PM_{2.5}$ for more than 1 month both in winter and in spring, with the special consideration of the influencing factors such as the sources effects and the meteorological information.

Sampling and analysis

The 1-min $PM_{2.5}$ concentration was obtained with LD-5C pocket microcomputer laser dust instrument (Beijing BINTA Green Technology Co., Ltd), based on the principle of laser light scattering method. Two sampling seasons were collected, i.e., from Dec. 15th, 2005 to Jan. 16th, 2006 for winter and from Mar. 17th to Apr. 28th, 2006 for spring in Beijing. The sampling site is located on the roof (~40 m high) of the 12th floor in the Building of Science and Technology of Beijing Normal University, located between the second and third Ring Roads.

SO_2, NO_2, O_3, CO, and PM_{10} data were obtained from air pollution index (API; API = 100 corresponds to Chinese air quality standard II) in the annual report of China EPA (http://www.bjepb.gov.cn). It was converted to be concentrations with the following formulas: $C = C_{low} + [(I - I_{low})/(I_{high} - I_{low})] \times (C_{high} - C_{low})$, where C is the concentration and I is the API value. I_{high} and I_{low}, the two values most approaching value I in the API grading limited value table, stand for the value higher and lower than I, respectively; C_{high} and C_{low} represent the concentrations corresponding to I_{high} and I_{low}, respectively. The detailed procedures were given elsewhere (Tang et al. 2005).

All the spectral density analysis was made by JMP 4.0. It can be made to show how long and how often about the cycles, i.e., the length and frequency of the cycles.

Results and discussion

The cyclic characteristics of pollutants

The longer-periodic cycle characteristics of SO_2, NO_2, O_3, CO, and PM_{10}

SO_2, NO_2, O_3, CO, and PM_{10} from Jan. 1st, 2001 to Dec. 31st, 2004 were obtained from the annual report of China EPA (http://www.bjepb.gov.cn). All of them but O_3 showed annually U-shaped cycles, i.e., higher in winter and spring, lower in summer and autumn, as Fig. 1 showed. As for ozone, it showed reverse tendency, i.e., highest concentration in summer and lowest in winter due to the sunshine density.

In addition, it is obviously from Fig. 1 that all the pollutants showed annual cycles. Take the SO_2 as an example: it showed higher concentration in winter and spring and lower in summer and autumn every year. This may be due to the stationary sources in different seasons, i.e., the coal burning in winter, dust storm in spring, and less sources in summer and autumn. In addition, the meteorological information may affect it much, which will

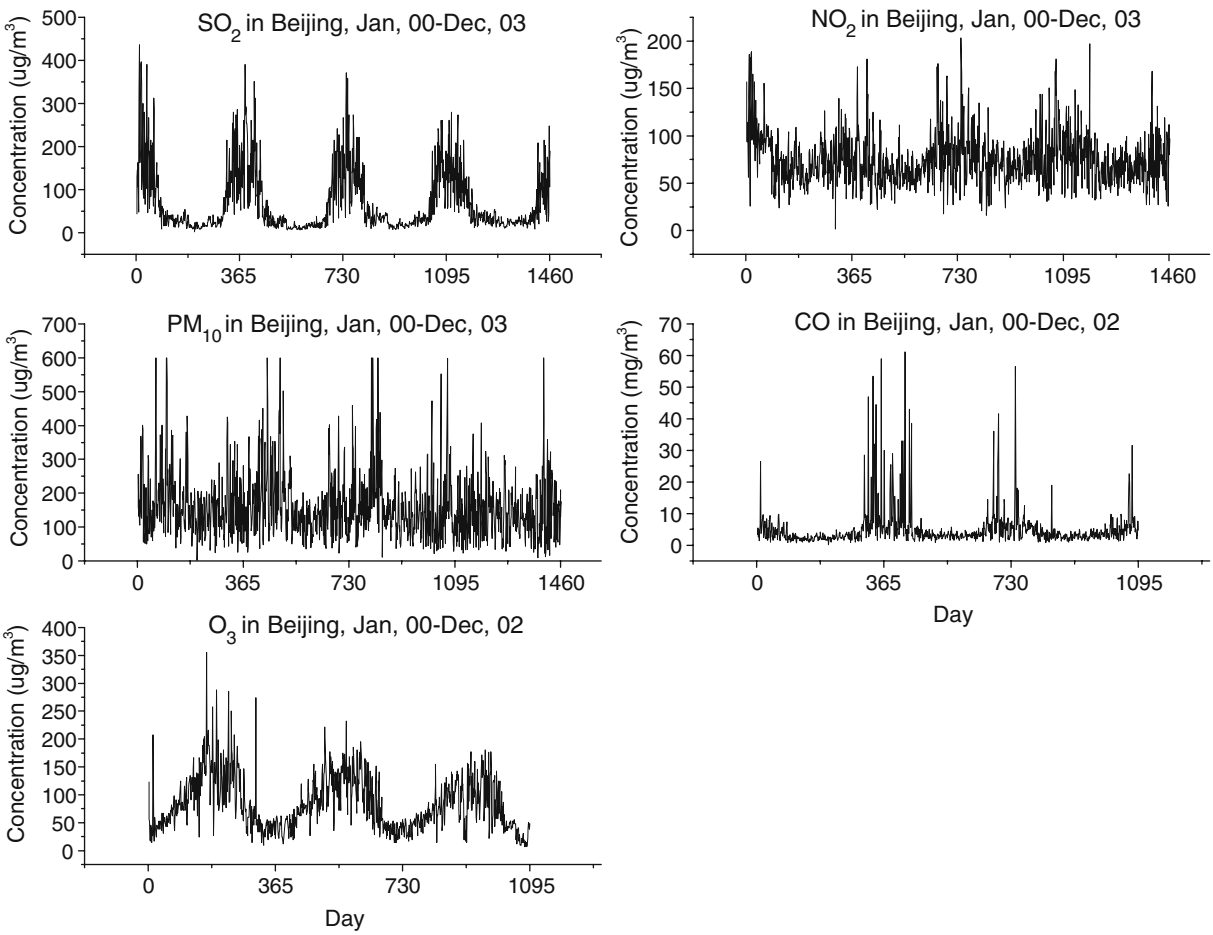

Fig. 1 The annual-cycle variation of SO_2, NO_2, O_3, CO, and PM_{10}

be discussed in detail in "The two possible factors that affect the cycle." It should be noted that the government control measures such as the use of anthracite coal and natural gases has helped much to control the SO_2 pollution, which can be shown from the steadily decrease of the concentration in heating period in wintertime.

However, as the pollutants' concentration was obtained one each day, we could not get the variation information of shorter time. How about it? Does it vary with cyclic characteristics? In order to get more information, we studied the 1-min $PM_{2.5}$ as following.

The shorter-periodic cycle characteristics of the $PM_{2.5}$

There seems a cyclic characteristic of 1-min $PM_{2.5}$, and the cycle is about 5–7 days. In general, $PM_{2.5}$ is lower and keeps a steady variation the first 2 to 3 days. It began to increase from the third night, and then it keeps the concentration until to the next night, amounts to about 10–15 times higher than the lowest, has a little decrease on the next daytime, and got the highest concentration on the fifth night. At last, it decreased sharply to the same level with the beginning. Then, next cycle begins.

Figure 2 gave two examples of the cycles of the ambient 1-min $PM_{2.5}$, both in wintertime and springtime. The Y axis referred to $PM_{2.5}$ concentration, which was read 1/min from the instrument. The X axis referred to the sampling time in that day, and the detailed sampling time was converted from day/hour/minute to numbers with the following formulas:

$$N = N_{\text{day}} + 1/1440 \times (60 \times T_{\text{hour}} + T_{\text{minute}})$$

Here, N refers to the beginning sampling time in the new form, N_{day} means the sampling day and

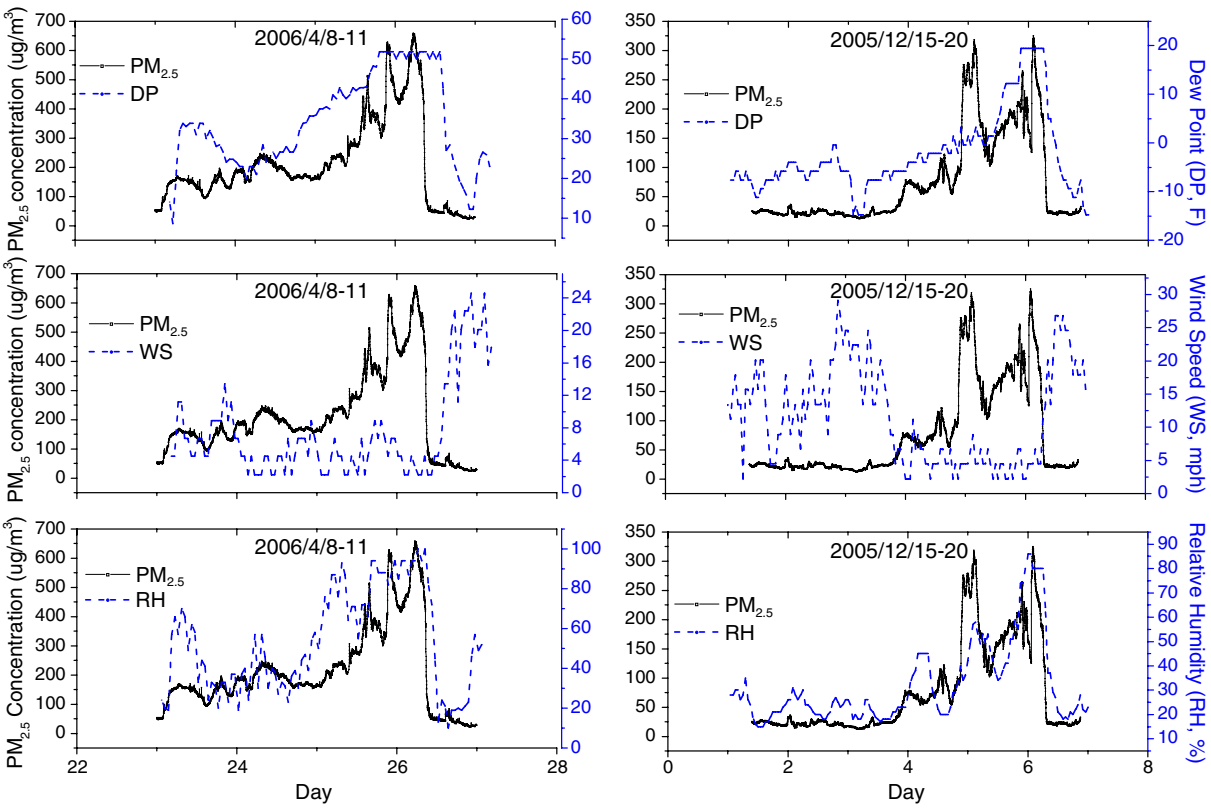

Fig. 2 $PM_{2.5}$ variation and correlation to meteorological information

1 from the beginning of sampling day. The number 1 is 2005/12/15 for wintertime and 2006/3/17 for springtime, and then other number follows in turn. T_{hour} and T_{minute} refer to the hour and minute time of the sampling $PM_{2.5}$ concentration. As every 1,440 min get 1 day, the 1/1,440 was multiplied before the total minute number.

It is obviously from the cycles in Fig. 2 that $PM_{2.5}$ varies much during the cycle. It increases slowly, then up to the highest concentration, then decreases sharply at the highest concentration like "a saw tooth" and was called a sawtooth cycle (Jia et al. 2008). In addition, it varies much during the sawtooth cycle, which may indicate even shorter cycle, which could be discussed more in the "periodograms" analysis and plots of spectral density in the "Proofs from the "periodograms" and spectral density analysis."

Proofs from the "periodograms" and spectral density analysis

In order to study how long the cycles of pollutants are, the periodogram graphs were made in Figs. 3 and 4. The spectral density analysis was shown in it. In general, the higher the peak of the cycle time is, the more often the cycle is. Thus, in Fig. 3, the longer yearly cycle (365, 180 days) of SO_2, NO_2, CO, O_3, and PM_{10} is obvious, which is consistent with what is shown in Fig. 1. Furthermore, the seasonal cycle (120, 60–90 days) and monthly cycle (21, 23, 27 days) are also evident. In addition,

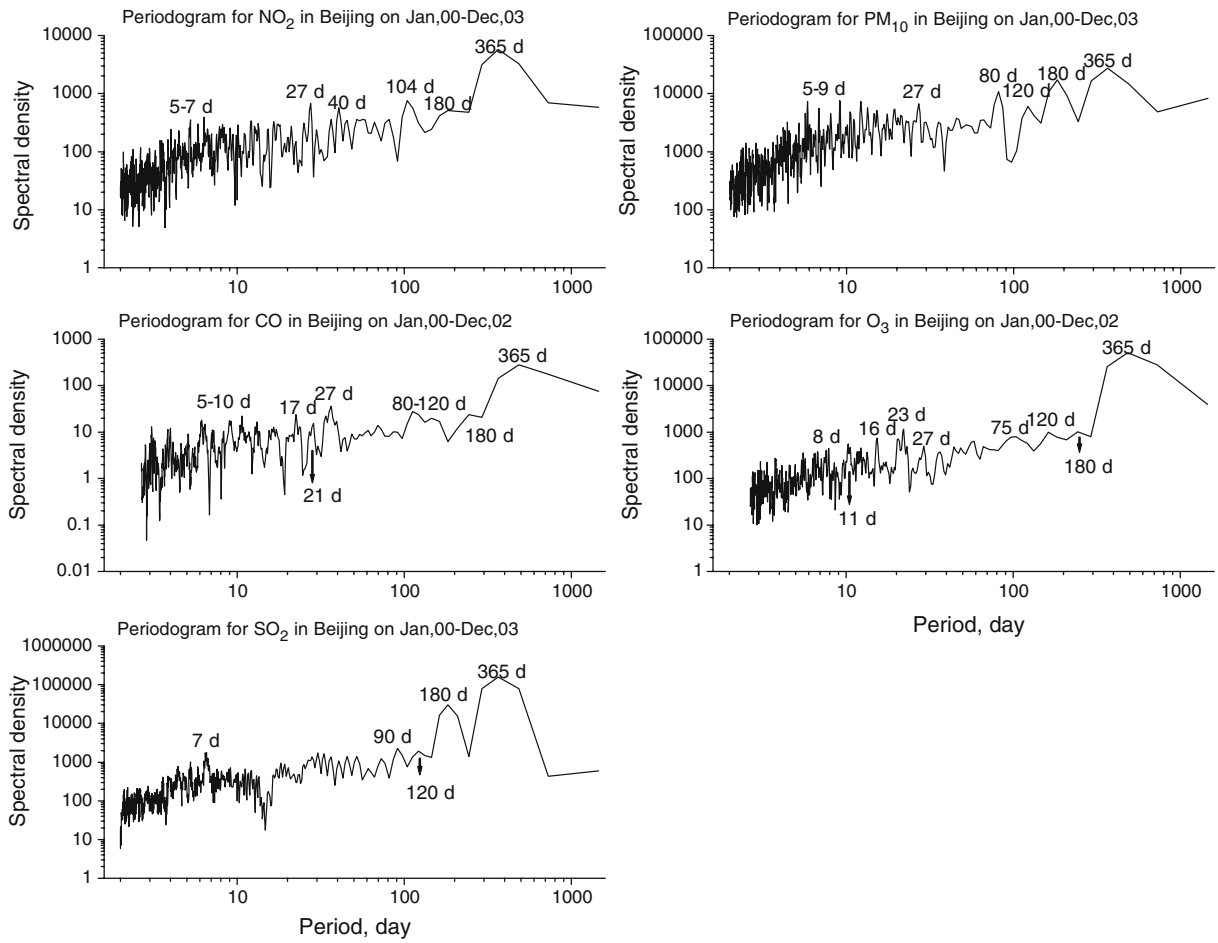

Fig. 3 The periodogram graphs of the longer-cycle pollutants by spectral density analysis

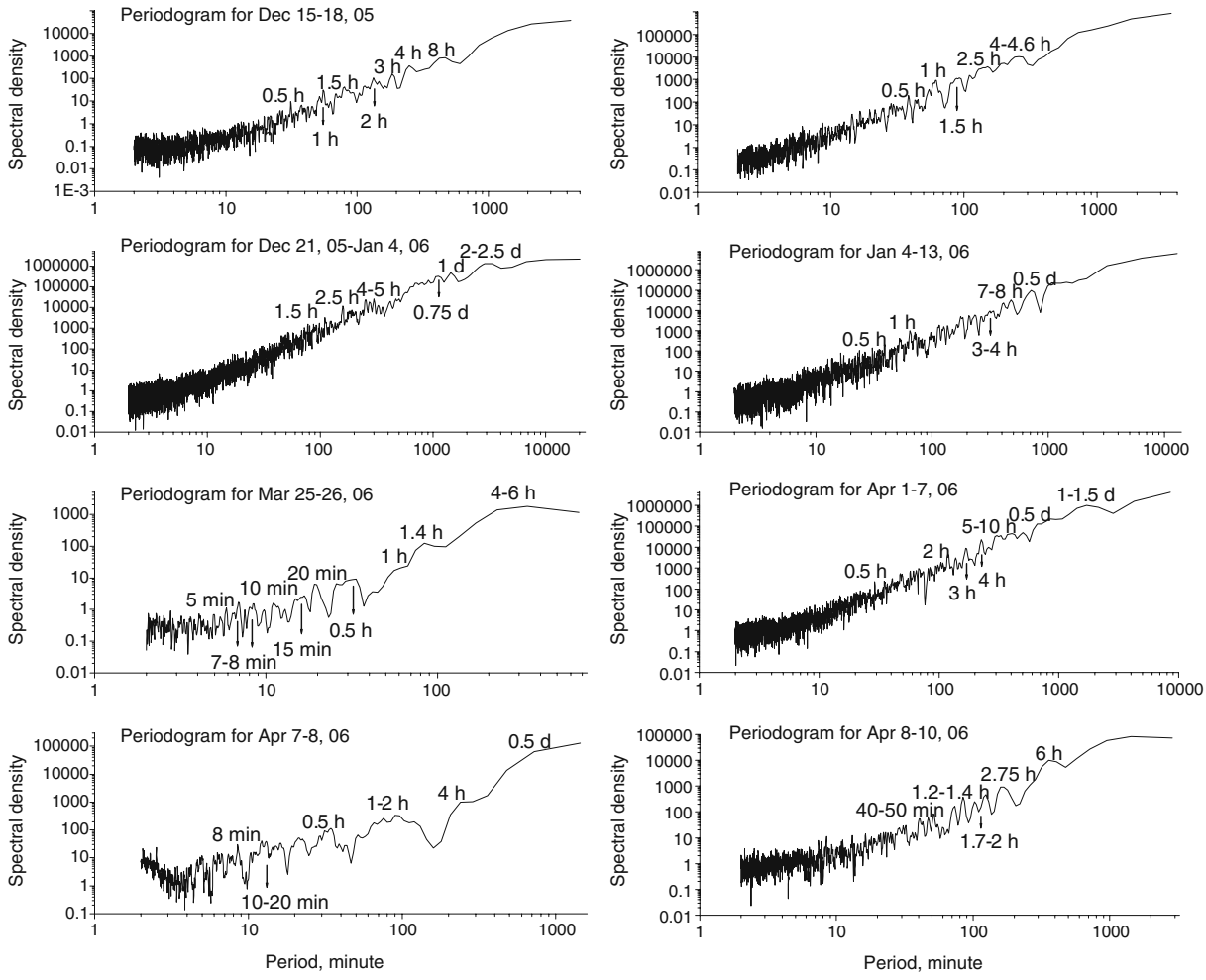

Fig. 4 The periodogram graphs of the shorter-cycle 1-min $PM_{2.5}$ by spectral density analysis

the shorter weekly cycle of 5–7 days is obviously shown, too. This may be due to the influences of meteorological synoptic cycles.

As for the 1-min $PM_{2.5}$, the cycle is even shorter, i.e., 8–12, 4–6, 3, 1–2 h, 20 min, as is shown in Fig. 4. This may partly explain the small cycle of 1-min $PM_{2.5}$ in Fig. 2 and gave the detailed information of the shorter cycles.

However, why do the pollutants show the cyclic characteristics? What are the factors influence it? We gave the investigation about 1-min $PM_{2.5}$, with the consideration of meteorological factors and the sources' effects, as the following "The two possible factors that affect the cycle."

The two possible factors that affect the cycle

The meteorological factors—synoptic cycle effects

Figure 2 showed the 1-min $PM_{2.5}$ and its correlation with the meteorological information. It is obvious that both the dew point (DP) and the relative humidity (RH) showed consistent variation with $PM_{2.5}$, i.e., gradual increase with the increase of $PM_{2.5}$ and sharp decreased at the highest point, which is similar with the $PM_{2.5}$ variation. While the wind speed showed reverse variation with $PM_{2.5}$, i.e., less variation and with short swings

with the $PM_{2.5}$ variation and decreased to lowest point with the increase of $PM_{2.5}$ to the highest peak, and then sharp increase with the sharp decrease of $PM_{2.5}$.

It is obvious from Fig. 2 that the meteorological factors influence greatly the $PM_{2.5}$. The spectral density analysis of the meteorological factors was made in Fig. 5 during the sampling time of Dec. 15th, 2005 to Jan. 16th, 2006 for winter and Mar. 17th to Apr. 28th, 2006 for spring. The meteorological cycle is consistent with the pollutants, as the cycle of the pressure, relative humidity, temperature, dew point, and wind speed is 4–5, 2–2.5, 1–1.5 days, 12, 6–8, 3 h, and so on. It should be noted that the sampling time of meteorological information is only about 1 month, and it could not explain the longer cycles. In addition, the meteorological data we got were 1/h, and it could not explain the shorter cycle, too.

Furthermore, there seems much difference about the meteorological factors between spring and winter. It is obvious that a 3-h cycle was evident in spring in Fig. 5 but not so clear in wintertime. This may be due to the diverse variation of meteorological factors such as more winds in spring may help to the diffusion of pollutants. However, the relative steadiness of the meteorological factors in winter and the inverse layer effects may help to the accumulation of pollutants and make the cycle longer. This can also be seen in the 1-min $PM_{2.5}$ cycle in Fig. 4.

The special sources effects

In addition to the meteorological factors, different source is another major factor that contributes the pollutants, that is why so many studies have been conducted to investigate the sources

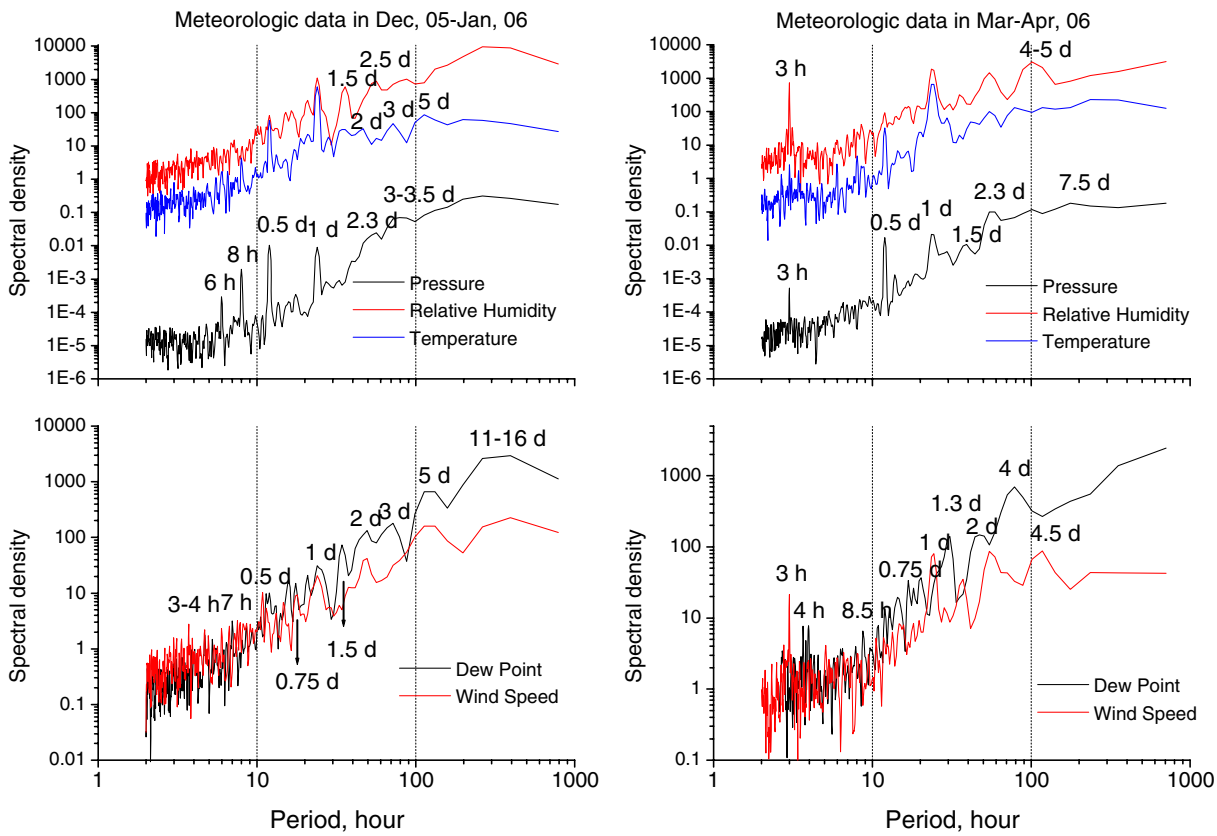

Fig. 5 The periodogram graphs of the meteorological information by spectral density analysis

identification. In order to study the contribution of sources to $PM_{2.5}$, one special and strong source is needed to study. As the mixing of vehicle exhausts, industry emission, coal burning, and dust soils constitute the species of $PM_{2.5}$ and shows even variation in the aerosol; it is not easy to study the source contribution in normal days.

However, the strong dust storm in April 16–18, 2006 may help to understand this. The dust storm attacked Beijing quietly at night, and people only found outside a "yellow" world in the morning of Apr. 17th, 2006, and the visibility even amounts to less than 3 km at 8:00 A.M. in Apr 17th. It brought ~300,000 tons of soil to Beijing and even influenced more than 300,000 km in China. Figure 6 gave the variation of $PM_{2.5}$ concentration and related meteorological information during the dust storm time. The dust events were generally characterized cold fronts and relative dry conditions, i.e., low relative humidity, and often with higher wind speed (Wang et al. 2005a), which can be seen in the blue dotted rectangle area in Fig. 6. However, the $PM_{2.5}$ concentration did not increase at this area; this can be explained by the coarse characteristics of this dust storm. The dust storm brought coarse particles to Beijing at first, which made the city to be a "yellow" world and most of the particle sizes were more than

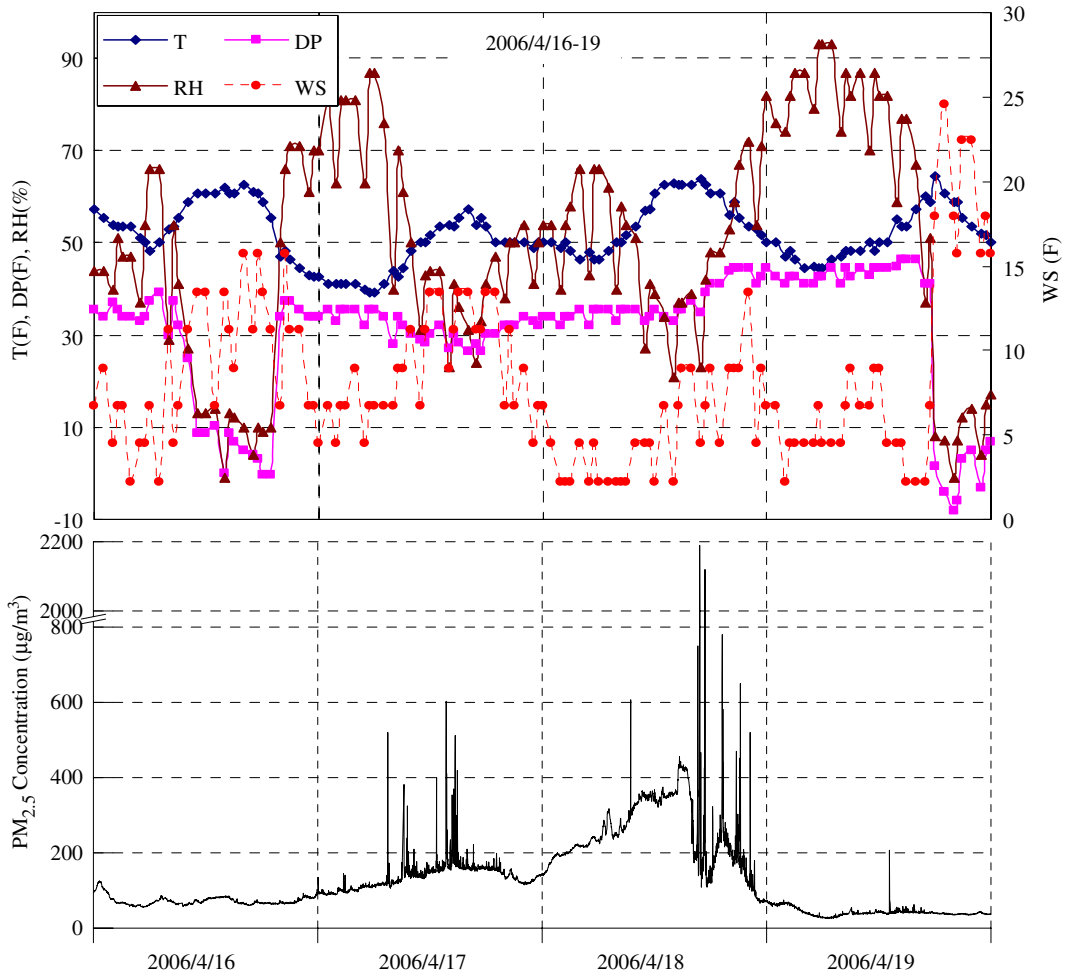

Fig. 6 The variation of $PM_{2.5}$ concentration and related meteorological information during the dust storm time (*T*, *DP*, *RH*, and *WS* means the temperature, dew point, relative humidity, and wind speed)

PM_{10} (http://www.cma.gov.cn). That is why $PM_{2.5}$ showed steady variation at the beginning of the dust storm.

Different from the dust storms before (Wang et al. 2005b; Sun et al. 2004b), the dust storm this time was characterized by the coarse particles at first, and $PM_{2.5}$ showed little increases. However, the later high relative humidity, lower wind speed, and other better meteorological factors help the creation and accumulation of the fine particles. That is why the $PM_{2.5}$ increases sharply and varies much especially during the daytime of April 17th and 18th. It is obviously that the meteorological information could not explain the diversity of $PM_{2.5}$ in Fig. 6, and it constantly increased tens even hundreds of times higher concentration within few minutes. This may be due to the intensity of the sources, i.e., the great amounts of fine particles was created due to the friction of coarse particles, resuspension of fine particles, and so on. Thus, it is concluded that the sources intensity could contribute $PM_{2.5}$ most especially at special time.

Conclusion

Characteristics of 1-min $PM_{2.5}$ were investigated, and the cyclic characteristics were shown in this paper. Several conclusions were obtained as following:

1. The variation of pollutants such as SO_2, NO_2, O_3, CO, and PM_{10} showed longer cycles, while the short-time 1-min $PM_{2.5}$ also showed shorter cycles.
2. The spectral density analysis confirmed this from the periodogram graphs, as the longer yearly cycle (365, 180 days), seasonal cycle (120, 60–90 days), monthly cycle (21, 23, 27 days), and weekly cycle (5–7 days) dominate most to SO_2, NO_2, CO, O_3, PM_{10}, and shorter hourly cycle (8–12, 4–6, 3, 1–2 h, 20 min) of 1-min $PM_{2.5}$ dominate most to 1-min $PM_{2.5}$ in this study.
3. Two important factors were considered to the cyclic characteristics of 1-min $PM_{2.5}$, the meteorological factors, and sources. Both the RH and the dew point showed consistent variation with $PM_{2.5}$, but the wind speed showed reverse variation with $PM_{2.5}$. Also, the coincidence of the meteorological cycles (4–5, 2–2.5, 1–1.5 days, 12, 6–8, 3 h) with 1-min $PM_{2.5}$ cycle may explain the pollutants cycles. The strong dust storm of April 16–18th, 2006 was taken as an example of sources effects. The greatly increase of $PM_{2.5}$ concentration within few minutes may show the great impact of sources, which may affect the cyclic characteristics much.

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