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Ambient concentrations of aldehydes in relation to Beijing Olympic air pollution control measures

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Abstract

Aldehydes are ubiquitous constituents of the atmosphere. Their concentrations are elevated in polluted urban atmospheres. The present study was carried out to characterize three aldehydes of most health concern (formaldehyde, acetaldehyde, and acrolein) in a central Beijing site in the summer and early fall of 2008 (from June to October). Measurements were made before, during, and after the Beijing Olympics to examine whether the air pollution control measures implemented to improve Beijing's air quality during the Olympics had any impact on concentrations of the three aldehydes. Average concentrations of formaldehyde, acetaldehyde and acrolein were $29.34 \pm 15.12\,\mu g/m^3$, $27.09 \pm 15.74\,\mu g/m^3$ and $2.32 \pm 0.95\,\mu g/m^3$, respectively, for the entire period of measurements, all being the highest among the levels measured in cities around the world in photochemical smog seasons. Among the three measured aldehydes, only acetaldehyde had a substantially reduced mean concentration during the Olympic air pollution control period compared to the pre-Olympic period. Formaldehyde and acrolein followed the changing pattern of temperature and were each significantly correlated with ozone (a secondary product of photochemical reactions). In contrast, acetaldehyde was significantly correlated with several pollutants emitted mainly from local emission sources (e.g., NO_2, CO, and $PM_{2.5}$). These findings suggest that local direct emissions had a larger impact on acetaldehyde than formaldehyde and acrolein.

1 Introduction

Beijing is one of mega cities in the world with a population of over 17 million. The rapid economic growth in China places a high demand on energy consumption, resulting in massive fossil fuel emissions of pollutants, e.g. nitrogen oxides (NO_x=NO+NO_2), sulfur dioxide (SO_2), carbon monoxide (CO), volatile organic carbons (VOCs) and particulate matter (Tang, 2004). In recent years, the number of automobiles in Beijing increased rapidly at a rate of approximately 15% annually (Hao et al., 2006; Chan and Yao, 2008)

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and the car stock in Beijing grew up to 4 million by the end of 2009. Hence, a major source of air pollution in Beijing is automobile emissions (Streets et al., 2007). Beijing's air pollution has also been featured with its high atmospheric oxidation capacity due to photochemical reactions in the summertime (Tang, 2004; Streets et al., 2007).

The Chinese government implemented a series of aggressive air pollution control measures to improve the air quality during the Beijing Olympics and Paralympics. Control measures included the reduction of pollutant emission from factories and industrial facilities by installing or improving pollutant control devices, reducing the production capacity, or relocating factories. Most noticeably, the number of private cars was reduced by half through an odd/even plate number rule; and all construction projects were suspended during the Olympic period (Wang et al., 2009a). It is of interest to the public and the scientific community as to whether these control measures resulted in significant improvements of air quality.

Aldehydes are ubiquitous constituents of urban atmospheres (Seinfeld and Pandis, 1998; Finlayson-Pitts and Pitts, 1986). Aldehydes can be directly emitted into the atmosphere from the incomplete combustion of biomass and fossil fuels (Zhang and Smith, 1999; Schauer et al., 2001), and formed in the atmosphere as a result of photochemical oxidation of reactive hydrocarbons (Possanzini et al., 2002; Altshuller, 1993). Exposure of animals or humans to certain aldehydes results in adverse health effects (AkbarKhanzadeh and Mlynek, 1997; Benjebria et al., 1994; Blair et al., 1990; Cassee et al., 1996a, b). Consequently formaldehyde and acetaldehyde are regulated as hazardous air pollutants (HAPs) by the US EPA due to their toxicity (IARC, 1995, 1985). Acrolein, another HAP, is known as a potent irritant to the eyes and the respiratory systems of humans and animals (Brock et al., 1979) and a human carcinogen (Feng et al., 2006).

The aims of this study are to characterize atmospheric aldehydes before, during and after the Olympic and Paralympic Games at a central Beijing site, and to examine the impact of the control measures on aldehydes concentrations in Beijing's atmosphere. Target aldehydes in this study include formaldehyde, acetaldehyde, and acrolein. To

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help achieve the study aim, we analyze these three aldehydes in relation to other air pollutants (PM, CO, SO_2, NO, NO_2, NO_x and ozone) measured at the same monitoring site and in relation to meteorological conditions (temperature, relative humidity, wind speed, and wind direction).

2 Experimental methods

2.1 Study design

Based on the intensity of the air pollution control measures (Wang et al., 2009a), our study used three periods defined as follows: the pre-Olympic period (4 June–19 July) when some light controls were implemented, the during-Olympic period (20 July–19 September) when full-scale control measures were implemented, and the post-Olympic period (20 September–30 October) when the control measures were relaxed. At a more refined temporal scale, extra control measures were adopted during each of the Olympics (8 August–24 August) and the Paralympics (6 September–17 September). These extra controls included barring of additional 20% government-owned cars from traveling on the road, suspending outdoor construction work, and temporarily closing some gas stations. From this point of view, the during-Olympic period can be further divided into two sub-periods: sub-period 1 was the period with full-scale control measures (20 July–7 August and 24 August–5 September), and sub-period 2 was the period with the full-scale control measures and the extra actions described above (8 August–23 August and 6–17 September). In order to examine if the air pollution control measures led to reduction in ambient concentrations of aldehydes, we measured the three aldehydes within a 1-month time window for each of the pre-Olympic, during-Olympic, and post-Olympic periods. The measurement scheme is shown in Fig. 1. Note that in the during-Olympic period, aldehydes were measured in both sub-periods 1 and 2.

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All samples were collected on the top of a 7-story building located in central Beijing, within the 2nd Ring Road and about 3 km northwest of the Tiananmen Square. This building was suited in the center of a hospital campus surrounded by streets with high densities of motor vehicle, pedestrian, and bicycle traffic.

2.2 Aldehydes measurement method

We used the PAKS method described previously to measure aldehydes (Herrington et al., 2005; Zhang et al., 2000). Briefly, this method uses a passive sampling technique and an HPLC-Fluorescence analytical technique. Samples were collected through a C_{18} cartridge (LC-18, 0.5 g/4.5 mL, Supelco Inc. US) coated with dansylhydrazine (DNSH) with a sampling duration of 24 h. Samples and field controls were eluted with acetonitrile; and aliquots of extracts were analyzed using an HPLC system with fluorescent detection. A Nova-Pak C_{18} column was used along with a mobile phase program described as follows: mobile phase A was composed of 80% water, 10% acetonitrile, and 10% tetrahydrofuran containing 0.68 g/L of KH_2PO_4 and 3.48 g/L of K_2HPO_4; mobile phase B was composed of 30% water, 40% acetonitrile, and 30% tetrahydrofuran containing 0.68 g/L of KH_2PO_4 and 3.48 g/L of K_2HPO_4. The excitation and emission wavelengths used for detecting aldehyde–DNSH derivatives were 250 nm and 525 nm, respectively. The collection efficiencies for ambient formaldehyde, acetaldehyde and acrolein of this method were 115.5% ± 11.0%, 105.8% ± 9.1%, and 87.5% ± 4.7% (mean ± SD, $N = 30$), respectively. The analytical detection limits of the method were 0.98 ng, 0.86 ng and 1.15 ng per cartridge and the analytical precision, determined as relative standard deviations (RSDs) of replicate samples, were 7.72%, 1.84% and 4.56% ($N = 8$) for formaldehyde, acetaldehyde and acrolein, respectively.

2.3 Other pollutants and meteorological data

Other pollutants, including O_3, CO, SO_2, NO, NO_2, NO_x and fine particles ($PM_{2.5}$), were measured simultaneously with the aldehydes (i.e. at the same site and on the

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same dates). $PM_{2.5}$ was collected onto Teflon filters using a Quad Channel Ambient Particulate Sampler (TH-16A, Tianhong Inc. China) at a flow rate of 16.7 L/min. $PM_{2.5}$ mass concentrations were then determined gravimetrically. Gaseous pollutants were measured using instruments from Ecotech Ltd., Australia, including an EC9810B ozone analyzer, an EC9830 CO analyzer, an EC9841B $NO/NO_2/NO_x$ analyzer, and an EC9850B SO_2 analyzer. Meteorological data (temperature, relative humidity, wind speed, and wind direction) were also collected at the same site. Wind speed and wind direction were monitored using a RM Young 05103V wind monitor (NexSens Technology, Inc); and temperature and relative humidity were monitored using a Met One Meteorology system.

3 Results

3.1 Concentrations of atmospheric aldehydes

Throughout the entire sampling period, 78 aldehyde samples were collected in total, including 28, 26, and 24 samples for the pre-, during-, and post-Olympic periods, respectively. No samples were collected on rainy days. One field control and one duplicate sample were collected every 3 to 5 days for quality control purposes. Sample concentrations were corrected with the average field blank concentrations. All the samples had detectable concentrations of aldehydes. Mean, standard deviation, minimum and maximum values of aldehydes concentrations throughout the entire period and in the three specific periods are given in Table 1. Results show that the period-mean concentration of formaldehyde increased by 1.55 μg/m^3 (4%, $p = 0.576$) from the pre- to the during-Olympic period and decreased by 23.37 μg/m^3 (63%, $p < .0001$) from the during- to the post-Olympic period. Period-specific mean concentration of acetaldehyde decreased by 11.45 μg/m^3 (33%, $p = 0.0074$) from the pre- to the during-Olympic period and continued to decrease by 3.12 μg/m^3 (13%, $p = 0.483$) from the during- to the post-Olympic period. Period-specific mean concentration of acrolein increased by

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0.47 µg/m^3 (20%, $p = 0.038$) from the pre- to the during-Olympic period and decreased by 1.50 µg/m^3 (52%, $p < .0001$) from the during- to the post-Olympic period. P-value of mean comparison between two periods was calculated based on two-tailed t test. The uncertainty of aldehydes difference between periods can be estimated using twice of RSDs for formaldehyde, acetaldehyde and acrolein which were 15.44%, 3.68% and 9.12%, respectively. It is, hence, clear that the difference of formaldehyde from the pre- to the during-Olympic period was very small and it might result from the measuring uncertainty.

Sixteen and ten samples were collected in the sub-period 1 and 2, respectively. Average concentrations of aldehydes in the sub-period 1 and in the sub-period 2 were 37.73 ± 10.72 µg/m^3 and 36.48 ± 12.42 µg/m^3 for formaldehyde, 26.32 ± 15.93 µg/m^3 and 19.88 ± 12.32 µg/m^3 for acetaldehyde, and 2.99 ± 0.81 µg/m^3 and 2.70 ± 0.75 µg/m^3 for acrolein. Hence, the reduction in aldehydes concentrations in the sub-period 2 in reference to in the sub-period 1 was 3%, 24%, and 10% for formaldehyde, acetaldehyde and acrolein, respectively. It is notable that acetaldehyde has the largest reduction between the two sub periods. Comparing concentrations in the two sub periods with those in the pre-Olympic period, we found that formaldehyde concentration was increased by 6% in the sub-period 1, and increased by 2% in the sub-period 2; acetaldehyde decreased by 25% in the sub-period 1 and 44% in the sub-period 2, and acrolein increased by 24% in the sub-period 1 and 12% in the sub-period 2.

3.2 Other air pollutants and meteorological condition

As shown in Table 2 period specific mean concentrations (derived from 24-hour or daily average concentrations) of SO_2, NO, NO_2, NO_x, CO and $PM_{2.5}$ decreased by 38% ($p < .0001$), 43% ($p = 0.0006$), 27% ($p = 0.0001$), 30% ($p < .0001$), 48% ($p < .0001$), and 26% ($p = 0.0079$) from the pre- to the during-Olympic period, respectively, whereas period-specific mean concentrations of daily average O_3 (24-h average) and

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daily maximum O_3 (1-h maximum within a day) increased by 23% ($p = 0.066$) and by 17% ($p = 0.036$), respectively. Another study also observed 16% increase in ozone from the pre- to the during-Olympic period (Wang et al., 2010b). From the during- to the post-Olympic period, SO_2, NO, NO_2, NO_x, and CO increased by 27% ($p = 0.053$), 852% ($p < .0001$), 102% ($p < .0001$), 186% ($p < .0001$), and 19% ($p = 0.126$), whereas daily average of O_3, daily maximum of O_3 and $PM_{2.5}$ decreased by 63% ($p < .0001$), 51% ($p < .0001$), and 5% ($p = 0.697$). Period-specific mean concentration of daily average photooxidant, approximated as the sum of O_3 and NO_2, remained in the same level from the pre- to the during-Olympic period and decreased by 8% ($p = 0.143$) from the during- to the post-Olympic period. Period-specific mean concentration of daily maximum photooxidant (the sum of maximum O_3 and NO_2) increased by 6% ($p = 0.336$) from the pre- to the during-Olympic period and decreased by 26% ($p < .0001$) from the during- to the post-Olympic period.

Figure 2 depicts the prevailing wind direction in summertime of Beijing as S-SSW-SW and was consistent among the three different sampling periods. A similar result was also reported in a previous paper reporting meteorological data measured in Beijing surrounding the 2008 Beijing Olympics (Wang et al., 2009b). The average temperature, relative humidity and wind speed in the three sampling periods are summarized in Table 2. The average temperature increased by less than 2 °C (7%, $p = 0.072$) from the pre-Olympic period to the during-Olympic period, but decreased by about 10 °C (36%, $p < .0001$) from the during-Olympic period to the post-Olympic period. Relative humidity (RH) decreased by about 4% (5%, $p = 0.194$) from the pre- to the during-Olympic period, and decreased by 15% (20%, $p < .0001$) from the during- to the post-Olympic period. Wind speed increased 0.07 m/s (4%, $p = 0.99$) from the pre- to the during-Olympic period, and increased by 0.18 m/s (11%, $p = 0.174$) from the during- to the post-Olympic period. There was no significant change in temperature, RH or wind speed between the pre- and the during-Olympic period. In contrast, the change between the during- and the post-Olympic periods was much larger for all three parameters.

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3.3 Correlation among aldehydes and other air pollutants

Since some of the air pollutants, e.g. $PM_{2.5}$, NO and NO_2, did not satisfy the normality distribution assumption, the Spearman rank correlation was used to examine the association between pollutants. The Spearman correlation coefficients among aldehydes and other air pollutants are shown in Table 3. The p-value for each coefficient was calculated using permutation test and the significance level of each correlation coefficient is indicated in Table 3 as well. Formaldehyde, acetaldehyde and acrolein were significantly correlated with each other. Formaldehyde was significantly correlated with oxides of nitrogen (NO, NO_2 and NO_x) in the negative direction, and was significantly correlated with each of daily average O_3, daily maximum O_3, CO and $PM_{2.5}$ in the positive direction. Acetaldehyde was significantly and positively correlated with SO_2, NO_2, CO and $PM_{2.5}$. Acrolein was significantly correlated with oxides of nitrogen in the negative direction and significantly correlated with ozone and $PM_{2.5}$ in the positive direction. No significant correlation was found for any of the three aldehydes with daily average photooxidant. However, daily maximum photooxidant was significantly correlated with formaldehyde ($p = 0.023$) and acrolein ($p = 0.053$), respectively. Both formaldehyde and acrolein were significantly correlated with temperature and RH. Acetaldehyde was significantly and positively correlated with RH but not with temperature.

4 Discussion

As shown in Table 4, formaldehyde and acetaldehyde concentrations in the summer time of Beijing were substantially higher than those reported for other cities during photochemical seasons. For example, Milan and Rome in Italy, the downtown area in Savannah of Georgia in the US, Rio de Janeiro in Brazil, and Guangzhou in China all had lower formaldehyde and acetaldehyde concentrations in the atmosphere than Beijing during photochemical seasons (Andreini et al., 2000; Baez et al., 1995; Feng et al., 2005, 2004; Grosjean et al., 2002; MacIntosh et al., 2000; Possanzini et al., 1996; Zhang et al., 1994).

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Aldehydes in the atmosphere are generated primarily from direct emissions from industrial and/or traffic sources and secondarily from the photochemical reactions in the atmosphere. Both of these sources might have contributed to the high concentration of atmospheric aldehydes in Beijing during the summer of 2008 when the present study was conducted. Our monitoring site was located in central Beijing and was surrounded by streets with high densities of motor vehicles. Hence, we think the mobile source was an important contributor to the high aldehyde concentrations we measured. In addition to directly emitting aldehydes, the mobile source emits a large amount of NO_x and VOCs, both of which are precursors of photochemical smog products including aldehydes.

Another important factor resulting in high concentration of ambient aldehydes could be meteorological conditions affecting air quality during summer months in Beijing (Streets et al., 2007). Beijing is located at 39°56′ N and 116°20′ E on the northwest border of the Great North China Plain. It is located in a warm temperate zone and has a typical continental monsoon climate (Chan and Yao, 2008). The air quality of Beijing in the summertime is largely determined by the meteorology (Streets et al., 2007), as, for example, temperature as well solar radiation are key factors that control the photochemistry processes (Wang et al., 2009b). Wind direction is associated with the origin of air masses transported from the surrounding areas of Beijing; and wind speed controls the dispersion of air pollution. In summer months, Beijing typically has high temperature (mean: 27 °C) and high RH (mean: 64%), both of which favor the photochemical reactions. In the summer, Beijing also has few windy days, which is unfavorable for atmospheric dispersion of air pollutants.

Due to the fact that the traffic source might be an important contributor to ambient aldehydes in Beijing, the adoption of intensive air pollution control measures, including the aggressive traffic restrains, in the during-Olympic period relative to the pre-Olympic period might lead to lower ambient concentrations of aldehydes in the during-Olympic. As described earlier, the changing patterns for formaldehyde, acetaldehyde and acrolein between the pre- and the during-Olympics periods were

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different: formaldehyde and acrolein increased from the pre- to the during-Olympic period, whereas acetaldehyde decreased. In terms of the sub periods, because of the extra air pollution control measures implemented in the sub-period 2 , more reduction in aldehydes concentrations were expected in the sub-period 2 than in the sub-period 1. Data showed that the reduction in acetaldehyde concentration (44%) from the pre-Olympics to the sub-period 2 was markedly larger than the reduction from the pre-Olympics to the sub-period 1 (25%). These results suggest a direct association between the adoption of air pollution control measures and the reduction in ambient concentration of acetaldehyde.

Comparing formaldehyde concentrations between the two sub periods, although the concentration of formaldehyde in the sub-period 2 (36.48 $\mu g/m^3$) was lower than that in the sub-period 1 (37.73 $\mu g/m^3$), the difference (1.25 $\mu g/m^3$) was very small, making it harder to suggest that the air pollution control measures be associated with formaldehyde concentration reduction. Concentration of acrolein in the sub-period 2 (2.70 $\mu g/m^3$) was lower than that in the sub-period 1 (2.99 $\mu g/m^3$) as well, and the difference was 0.29 $\mu g/m^3$ (10%). Given that the standard deviation of mean acrolein concentration in the during-Olympic period was 0.78 $\mu g/m^3$, it is also difficult to suggest any direct association between the control measures and the acrolein concentration.

Average concentrations of aldehydes and the mean value of meteorological condition in three periods were plotted together pairwised in Fig. 3. We observed that formaldehyde and acrolein followed the changing pattern of temperature between periods. The 7% increase of temperature was accompanied by 4% and 20% increase in formaldehyde and in acrolein from the pre- to the during-Olympic period; and the 36% decrease in temperature was followed by 63% reduction in formaldehyde and 52% reduction in acrolein from the during- to the post-Olympic period. Acetaldehyde did not follow the trend of temperature from the pre- to the during-Olympic period; however, the post-Olympic period had both lowest acetaldehyde concentration and temperature.

Relative humidity may also contribute to the formation of formaldehyde and acrolein, because concentration of each of these aldehydes tracked the RH levels (see Fig. 3).

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In the post-Olympic period, RH decreased by 20% which was accompanied by large reductions in aldehydes concentrations.

The consistent trends among formaldehyde, acrolein and temperature suggest that the secondary source, e.g. photochemical reaction, be a dominant contributor to atmospheric formaldehyde and acrolein in Beijing. In contrast, an association between traffic reduction and acetaldehyde concentration reduction suggests that primary emission from motor vehicles mainly contributed to acetaldehyde in Beijing during the summer of 2008. The significant correlations among aldehydes and meteorological parameters, i.e., temperature and RH, further support the above suggestions. Relationships among aldehydes shown in Fig. 4 also suggest the presence of common sources for the three aldehydes.

As reported in Table 3, a significant correlation between formaldehyde and the secondary pollutant, i.e., daily average O_3 or daily maximum O_3, was observed; however, associations between formaldehyde and primary pollutants, i.e., CO and SO_2 were not significant. Surface ozone and formaldehyde were not reduced in the during-Olympic period even though O_3 is critical for the formation of ambient formaldehyde (Seinfeld and Pandis, 1998). Conversely, the primary air pollutants, i.e. SO_2, CO, and nitrogen oxide, were reduced in the during-Olympic period. Similar correlation patterns were found for acrolein. Significant correlation between formaldehyde and daily maximum photooxidant (O_3+NO_2) was observed, whereas ambient formaldehyde and daily average photooxidant was not significantly correlated (Fig. 5). It might be because aldehydes were measured for 24 h, while photochemical activities had strong diurnal variation; and it is more reasonable to use hourly concentration of photooxidant to examine the association. Volatile organic compounds (VOCs) also play an important role in the atmospheric chemistry of aldehydes. Unfortunately, we were unable to measure VOCs at this site. However, the Wang et al paper reported that non-methane hydrocarbons were reduced by 25% to 35% during the Olympic pollution control period from their pre-Olympic levels (Wang et al., 2010a). Li et al. also observed the lowest VOCs concentration during in the Olympic Games period compared to the pre-Olympic and

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the post-Olympic periods (Li et al., 2010). The lower concentration of acetaldehyde in the during-Olympic period might be related to the lower concentration of VOCs in the same period.

On the contrary, acetaldehyde was significantly associated with primary pollutants, i.e., CO, SO_2, and $PM_{2.5}$, but with none of daily average O_3, daily maximum O_3, and daily maximum photooxidant. It was also observed that other primary air pollutants, e.g. SO_2, CO, nitrogen oxide, and $PM_{2.5}$, were reduced in the during-Olympic period when extensive air pollution control measures were implemented. On the other hand, SO_2 and CO, with a longer lifetime in the atmosphere, may be transported over a longer distance. The stronger correlation of acetaldehyde with SO_2 and CO also suggests a possibility that acetaldehyde might come from longer-distance transport.

In conclusion, in the summer of 2008 when Beijing hosted the Olympics, concentrations of formaldehyde, acetaldehyde and acrolein were found to be the highest among those previously reported in other cities around the globe. The air pollution control measures implemented during the Olympic and Paralympic Games appeared to be associated with a concentration reduction in acetaldehyde but not in formaldehyde and acrolein. Our results suggest that the secondary photochemical processes may have dominated the formation of formaldehyde and acrolein, whereas the reduction in primary emissions (mainly motor vehicles) may have contributed to the reduction in acetaldehyde concentration during the Olympic air pollution control period in Beijing.

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Table 1. Concentrations of aldehydes in pre-, during-, and post-Olympic periods (unit: µg/m^3)*.

	Pre-Olympics				During-Olympics				Post-Olympics				Whole period			
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Formaldehyde	35.70	9.04	15.83	60.12	37.25	11.18	17.06	68.60	13.88	13.89	1.02	57.50	29.34	15.12	1.02	68.60
Acetaldehyde	35.20	15.12	15.73	67.23	23.75	14.67	2.02	63.87	20.63	13.59	3.31	62.35	27.09	15.74	2.02	67.23
Acrolein	2.41	0.86	1.02	4.51	2.88	0.78	1.79	4.63	1.38	0.49	0.64	2.48	2.32	0.95	0.64	4.63

* Concentrations were field blank corrected; average concentration of aldehydes was calculated if the duplicated samples were collected.

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Table 2. Mean and standard deviation of air pollutants and meteorological parameters in three periods.

Air pollutants and meteorological condition	Pre-Olympics (6/2/2008–7/19/2008)		During-Olympics (7/20/2008–9/19/2008)		Post-Olympics (9/20/2008–10/30/2008)	
	Mean	SD	Mean	SD	Mean	SD
SO_2 (ppb)	7.82	4.00	4.65	2.61	5.92	3.51
NO (ppb)	3.66	2.09	2.29	1.92	21.81	17.43
NO_2 (ppb)	24.15	5.55	18.81	7.94	38.03	16.85
NO_x (ppb)	28.37	7.00	21.06	9.66	60.18	29.24
O_3 (daily average, ppb)	33.43	15.16	39.22	16.74	14.45	6.67
O_3 (daily maximum, ppb)	70.69	31.60	82.85	27.56	40.43	19.92
O_3+NO_2 (daily average, ppb)	57.45	13.70	57.42	15.23	52.43	18.47
O_3+NO_2 (daily maximum, ppb)	92.19	30.14	97.62	27.86	71.81	29.18
CO (ppm)	1.15	0.40	0.64	0.22	0.76	0.57
$PM_{2.5}$ (µg/m^3)	96.7	38.7	75.6	41.5	71.8	58.0
Temperature (°C)	25.33	3.24	27.11	2.86	17.23	3.58
Relative Humidity (%)	66.9	11.8	63.3	9.8	50.3	17.0
Wind Speed (m/s)	1.59	0.40	1.66	0.43	1.84	0.87

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Table 3. Spearman correlation coefficients among aldehydes, other air pollutants, and meteorological parameters.

	Formaldehyde	Acetaldehyde	Acrolein	SO_2	NO	NO_2	NO_x	[1]O_3	[2]O_3	[1]O_3+NO_2	[2]O_3+NO_2	CO	$PM_{2.5}$	T	RH
Formaldehyde	1														
Acetaldehyde	0.59**	1													
Acrolein	0.63**	0.43**	1												
SO_2	0.13	0.51**	0.005	1											
NO	−0.53**	−0.009	−0.53**	0.096	1										
NO_2	−0.31*	0.23*	−0.36*	−0.40**	0.80**	1									
NO_x	−0.41**	0.14	−0.47**	0.31**	0.89**	0.97**	1								
[1]O_3	0.41**	0.07	0.34*	0.21*	−0.73**	−0.57**	−0.65**	1							
[2]O_3	0.38**	0.04	0.33**	0.27**	−0.61**	−0.40**	−0.49**	0.92**	1						
[1]O_3+NO_2	0.060	0.14	0.017	0.58**	−0.19*	0.11	0.014	0.68**	0.77**	1					
[2]O_3+NO_2	0.26*	0.14	0.23	0.46**	−0.39**	−0.10	−0.21	0.79**	0.91**	0.91**	1				
CO	0.26*	0.46**	0.20	0.50**	0.17*	0.35**	0.29**	−0.01	0.08	0.30**	0.25**	1			
$PM_{2.5}$	0.39**	0.47**	0.24*	0.67**	−0.15	0.20*	0.09	0.31**	0.34**	0.56**	0.49**	0.65**	1		
Temperature	0.56**	0.10	0.59**	0.17*	−0.74**	−0.58**	−0.67**	0.79**	0.76**	0.43**	0.61**	0.08	0.33**	1	
RH	0.62**	0.30*	0.38*	−0.04	−0.17*	−0.02	−0.08	−0.17*	−0.17*	−0.28**	−0.18*	0.29**	0.32**	−0.05	1
Wind speed	−0.03	−0.03	0.08	−0.05	−0.34**	−0.44**	−0.41**	0.31**	0.17*	0	0.033	−0.27*	−0.07	0.18*	−0.31**

Significance of each coefficient is determined by the p-value, * indicates that the coefficient is significant at the significant level of 0.05; ** indicates that the coefficient is significant at the significant level of 0.01; [1] daily average (24-h) concentration; [2] 1 h maximum concentration within a day.

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Table 4. Concentrations of aldehydes in ambient air in different cities.

Location	Study season	Average aldehyde concentration (µg/m^3)			References
		Formaldehyde	Acetaldehyde	Acrolein	
Milan, Italy	Summer (August)	8.9	13.7	Non-available	Andreini et al., 2000
Rome, Italy	June–July 1994	22.77	18.27	1.75	Possanzini et al., 1996
Downtown Savannah, GA, USA	December 1995 through November 1996	2.0	2.3	Non-available	MacIntosh et al., 2000
Suburban area in Central New Jersey, USA	June–August 1992	15.37	4.75	Non-available	Zhang et al., 1994
Mexico city, Mexico	March–May 1993	43.5	33.8	Non-available	Baez et al., 1995
Rio de Janeiro, Brazil	May to November 2000	10.84	10.43	0.82	Grosjean et al., 2002
Guangzhou, China	June to September 2003	13.68	8.33	1.36	Feng et al., 2005
Guangzhou, China	August–September 2002	13.29	7.6	Non-available	Feng et al., 2004
Beijing, China	June to October 2008	29.34	27.09	2.32	Current study

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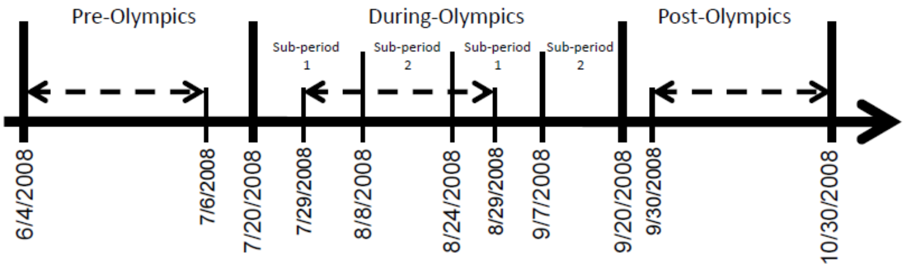

Fig. 1. Time scheme of the pre-, the during-, and the post-Olympic periods, two sub periods, and the periods when aldehydes measurement was conducted (indicated by dotted lines with double-headed arrows).

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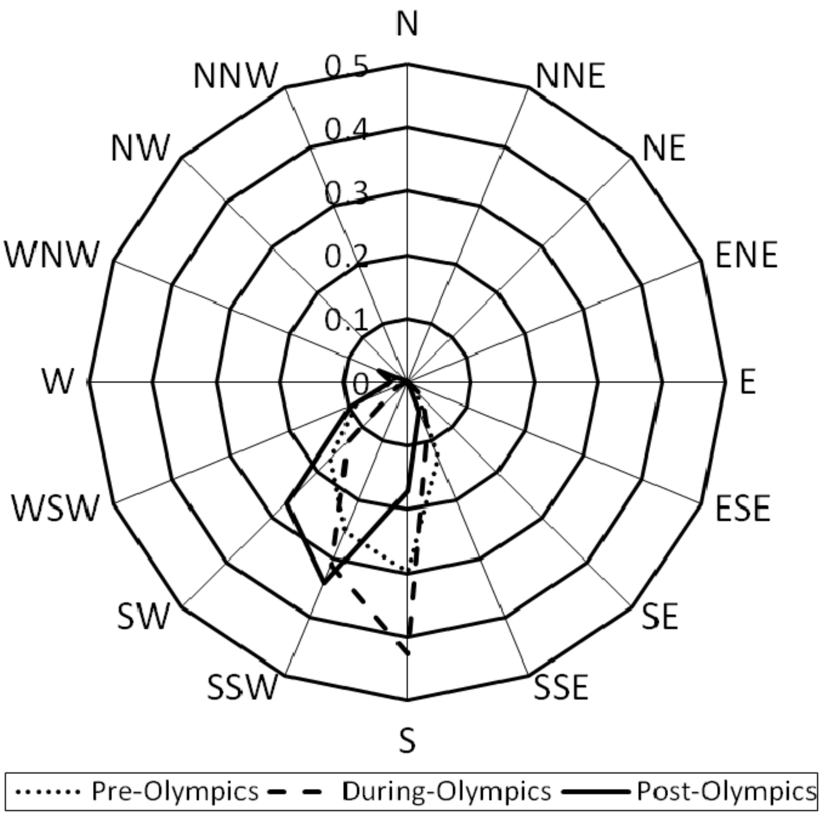

Fig. 2. Rose plot of wind direction in the three periods of Beijing Olympics.

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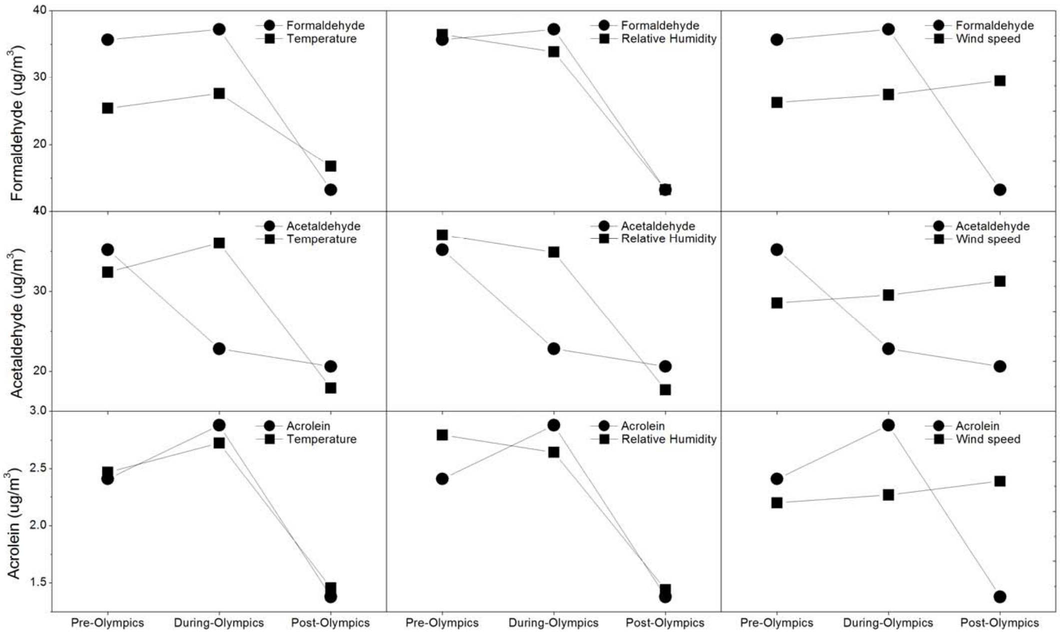

Fig. 3. Changes of ambient aldehydes and meteorological parameters, e.g. temperature, relative humidity, and wind speed, in the three sampling periods.

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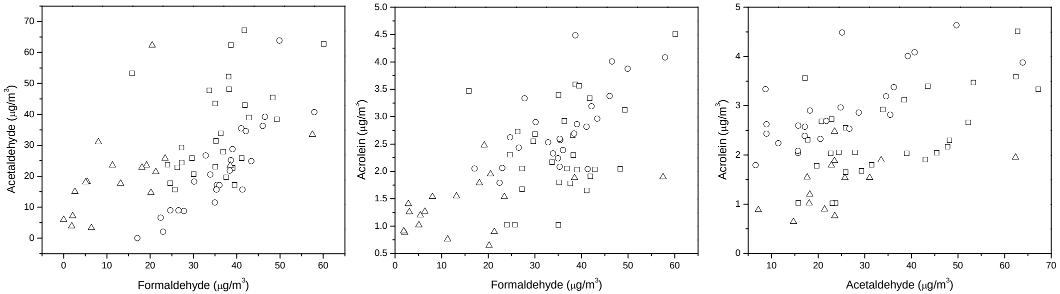

Fig. 4. Bi-variate plots showing relationships between aldehydes for the whole study period. Square symbols represent observations in the pre-Olympic period; round symbols represent observations in the during-Olympic period; and triangle symbols represent observations in the post-Olympic period.

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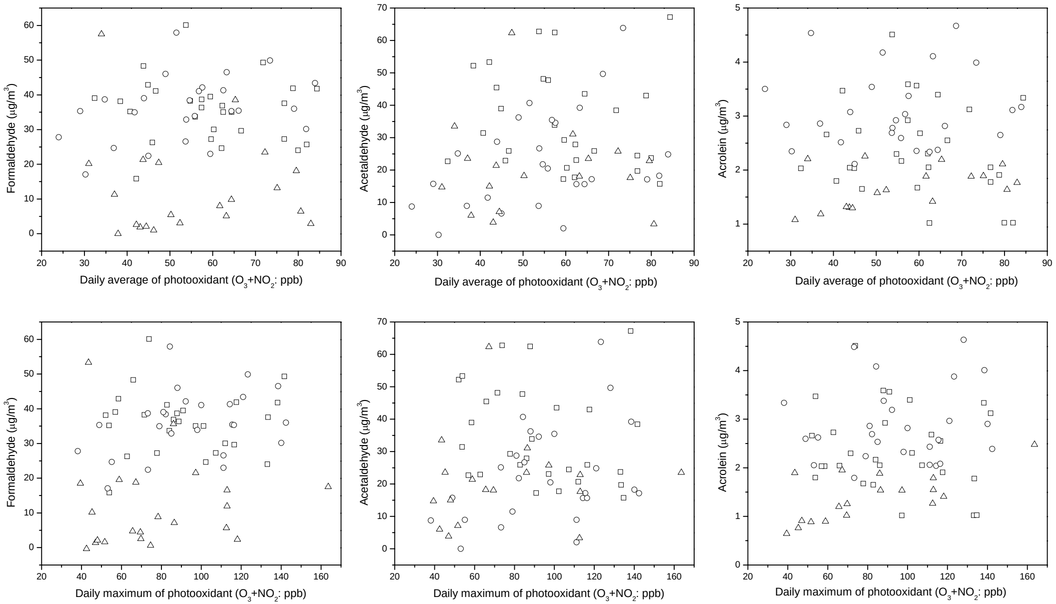

Fig. 5. Bi-variate scatter plots between each measured aldehyde and the daily average and the daily maximum photooxidant (O_3+NO_2) for the whole studying period. Square symbols represent observations in the pre-Olympic period; round symbols represent observations in the during-Olympic period; and triangle symbols represent observations in the post-Olympic period.

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