



Levels, sources and health risks of carbonyls and BTEX in the ambient air of Beijing, China

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Abstract

The atmospheric concentrations of carbonyls and BTEX (benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene) were measured simultaneously at a same sampling site in Beijing from September 2008 to August 2010. The average concentrations of the total measured carbonyls during autumn, winter, spring, and summer were 37.7, 31.3, 39.7, 50.5 $\mu\text{g}/\text{m}^3$, respectively, and maximal values for their diurnal variations usually happened at noontime. In contrast to carbonyls, the average concentrations of the total measured BTEX during the four seasons were 27.2, 31.9, 23.2, 19.1 $\mu\text{g}/\text{m}^3$, respectively, and minimal values for their diurnal variations always occurred in the early afternoon. The average concentration for carbonyls increased about 24% from September 2008–August 2009 to September 2009–August 2010, for BTEX, increased about 15%. Integrated life time cancer risks for three carcinogens (benzene, formaldehyde and acetaldehyde) in Beijing exceeded the value of $1\text{E}-06$, and the hazard quotient (HQ) of non-cancer risk of exposure to formaldehyde exceeded unity.

Key words: carbonyls; BTEX; sources; exposure and risk assessment; Beijing

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Introduction

Carbonyls and BTEX (benzene toluene ethylbenzene, *m,p*-xylene and *o*-xylene) in urban areas are one of the key problems for human living quality not only for their significant contribution in the formation of oxidants like ozone and peroxyacetyl nitrate (PAN) in the troposphere, but also for their adverse effects on human health (Haagen-Smit and Fox, 1956; Atkinson et al., 1997, 1999, 2000; Hoque et al., 2008). The most frequently adverse impacts of carbonyls on human health are eye and lung irritations (Carlier et al., 1986; WHO, 2000), while for BTEX they are mainly respiratory irritation and central nervous system damage (Stupfel, 1976; Crump, 1994; Durmusohlu et al., 2010). Formaldehyde, acetaldehyde and benzene are even suspected to be carcinogenic (Stupfel, 1976; Crump, 1994; Carlier et al., 1986; Báez et al., 2003; Dutta et al., 2009). It has been estimated by the World Health Organization (WHO, 1999) that a life exposure of $1 \mu\text{g}/\text{m}^3$ of benzene leads to about 6 cases of leukaemia per 1,000,000 inhabitants. The large emission of fossil combustion in urban areas, such as automobile exhaust, has been recognized to be their dominant primary source (Anderson et al., 1996; Báez et al., 2003; Wang et al., 2002). In addition, photochemical reactions involving volatile organic compounds (VOCs) constitute another important source for atmospheric carbonyls.

Risk assessment is one of the fastest evolving tools to evaluate the impact of the hazards on human health and to determine the level of treatment required to solve a specific environmental problem (Asante-Duah, 1993; LaGrega et al., 1994). Pollutants are usually classified as carcinogens and non-carcinogens for estimating their human health risks. To better evaluate the health risks, long-term measurement of pollutants are needed, however, up to now the measurements of atmospheric carbonyls and BTEX in Beijing only focused on short periods, mainly during the periods of high ozone levels episodes typically from July to September (Song et al., 2007; Duan et al., 2008; Liu et al., 2009a, 2009b; Xu et al., 2010). With respect to long-term monitoring, only our group (Pang et al., 2006) conducted a one-year field campaign for atmospheric carbonyls from November 2004 to October 2005. Because of the incredibly fast change experienced by Beijing City, such as the fast increase rate of private cars (about 10% per year) and the series of air clean actions adopted by Beijing Municipal Government (Liu et al., 2009a), the atmospheric BTEX and carbonyls are expected to vary accordingly. In this study, carbonyls and BTEX were simultaneously investigated for two consecutive years (2008–2010) in Beijing to further reveal their pollution levels, to recognize their main sources and to estimate their cancer and non-cancer risks.

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1 Materials and methods

1.1 Sampling site

Air samples were collected on a rooftop (20 m above the ground level) in the Research Center for Eco-Environmental Sciences which lies in the north of Beijing City (39.8°N, 116.5°E) between the 4th and 5th ring roads. The detail information about the sampling site was described in our previous studies (Pang and Mu, 2006; Liu et al., 2009a).

1.2 Sampling and analysis

Carbonyls and BTEX were simultaneously sampled at least 4 days per month from September 2008 to August 2010, with carbonyls' samples collected at 2 hr interval and BTEX at 1 hr interval. Nearly all kinds of weather were selected each month to collected samples, excepting raining day: at least one clear day, one cloudy day, one haze day with average wind speed lower than 3 m/sec, and one windy day with hourly average windy speed higher than 8 m/sec.

The sampling process and analysis procedure of atmospheric carbonyls were mainly based on the US EPA TO-11A method (US EPA, 1999), and the details have been described in our previous publications (Pang and Mu, 2006, 2007). Briefly, ambient air was drawn through a 2,4-dinitrophenylhydrazine (DNPH)-coated silica gel cartridge (Waters, USA) by a mini-pump (NMP 830 KNDC, Germany) at a flow rate of 800 mL/min for 2 hr, and the corresponding DNPH hydrazones of carbonyls were separated by a Thermo ODS Hypersil reverse phase column (5 μ m, 250 mm \times 4.6 mm) and automatically analyzed by an photodiode array UV detector (at 360 nm) in an Alliance 2695 HPLC system (Waters, USA). The detected limit ($S/N = 3$) by this method was in the range of 80–200 ng/m³ for various carbonyls of 96 L sampling volume.

As for BTEX, they were sampled by drawing air through an absorption tube (15 cm long, 4 mm ID) filled with Tenax-TA (80–100 mesh, 100 mg) at a flow rate of 350 mL/min for 1 hr, and analyzed by a gas chromatography equipped with a Photo Ionization Detector (GC-PID, GC4400, East & West Analytical Instruments, Inc., China) after thermal desorption. The method detection limit (with a signal-to-noise ratio of 2) for the benzene, toluene,

ethylbenzene, *m,p*-xylene and *o*-xylene were 0.01, 0.02, 0.06, 0.07 and 0.07 μ g/m³, respectively. One blank sample was analyzed for each day, and found that the concentrations of BTEX in the blank tube were less than 3% of those in the ambient air samples. Distinct breakthrough for benzene was found when the absorption tube was kept under ambient air temperature, but greatly improved when the tube was kept in a bath of ice-salt bag. Therefore, all samples were collected under the bath of ice-salt bag, and the collection efficiencies for benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene were (81.5 \pm 5.6)%, (93.6 \pm 2.2)%, (95.8 \pm 0.8)%, (96.5 \pm 2.4)% and (91.5 \pm 4.6)% (the result of 10 reduplicate experiments with two absorption tubes in series), respectively. The recovery ratios for benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene were (95.7 \pm 3.1)%, (99.2 \pm 2.6)%, (98.6 \pm 2.4)%, (98.1 \pm 2.4)% and (96.2 \pm 2.2)% (the result of 22 reduplicate experiments), respectively. The details about preparation of the absorption tube, sampling and analyzing procedures have been described in our previous publication (Liu et al., 2009a).

Meteorological conditions (temperature, wind speed, atmospheric pressure and precipitation) during the investigated years were recorded by an automatic meteorological station (the Beijing Urban Ecosystem Research Station, Research Center for Eco-Environmental Sciences).

2 Results and discussion

2.1 Average diurnal variation of carbonyls in each season during the two consecutive years

Eight carbonyls were identified in the ambient air, namely formaldehyde, acetaldehyde, acetone, propionaldehyde, butyraldehyde, benzaldehyde, valeraldehyde and hexaldehyde among which formaldehyde, acetaldehyde and acetone were by far the principal carbonyls and could be detected in every sample. The average diurnal variation of the three main carbonyls and the total eight carbonyl compounds in each season during the two consecutive years are presented together in Fig. 1. The ranges and arithmetic mean concentrations of the three principal aldehydes measured in this study and our previous study (Pang and Mu, 2006) at the same sampling site in each season are shown in Table 1. It is evident that the measured carbonyls

Table 1 Average concentrations (μ g/m³) of the three main carbonyls in each season during different investigated years

Season	Year	Number of samples	Formaldehyde		Acetaldehyde		Acetone		Reference
			Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	Range	
Autumn	2008	157	5.9 \pm 3.2	0.4–17.7	6.6 \pm 4.2	0.5–24.5	13.1 \pm 7.1	1.8–38.9	This work
	2009	88	8.3 \pm 5.8	0.5–28.7	9.8 \pm 6.8	1.5–34.3	22.1 \pm 13.1	4.6–50.3	This work
	2005	72	15.8 \pm 9.7	3.7–45.6	16.2 \pm 7.2	3.7–45.6	15.8 \pm 9.7	2.8–30.4	Pang and Mu, 2006
Winter	2008	116	4.8 \pm 4.5	0.1–20.6	6.0 \pm 5.7	0.4–22.7	11.6 \pm 6.8	0.4–38.9	This work
	2009	90	4.3 \pm 3.3	0.4–13.2	5.3 \pm 3.6	0.9–14.7	16.0 \pm 6.9	4.5–40.7	This work
	2004	86	4.2 \pm 2.8	1.1–12.1	7.6 \pm 5.3	1.4–30.1	7.8 \pm 4.6	1.8–25.3	Pang and Mu, 2006
Spring	2009	124	5.3 \pm 3.9	0.1–20.6	6.8 \pm 4.4	0.9–24.1	16.4 \pm 8.7	3.8–49.9	This work
	2010	84	6.6 \pm 3.7	0.7–16.2	6.9 \pm 3.5	0.4–15.3	28.9 \pm 8.7	8.8–44.0	This work
	2005	120	14.5 \pm 8.7	1.9–45.8	12.5 \pm 6.6	1.1–32.9	16.2 \pm 7.2	1.3–38.7	Pang and Mu, 2006
Summer	2009	116	8.8 \pm 4.7	0.9–23.9	9.5 \pm 5.1	1.0–26.0	19.3 \pm 7.5	7.3–40.2	This work
	2010	90	10.2 \pm 4.2	0.2–18.6	9.7 \pm 3.9	0.5–18.1	28.7 \pm 10.8	10.6–82.5	This work
	2005	110	19.5 \pm 8.9	2.6–52.9	14.1 \pm 7.4	1.8–35.7	20.7 \pm 9.9	10.6–53.2	Pang and Mu, 2006

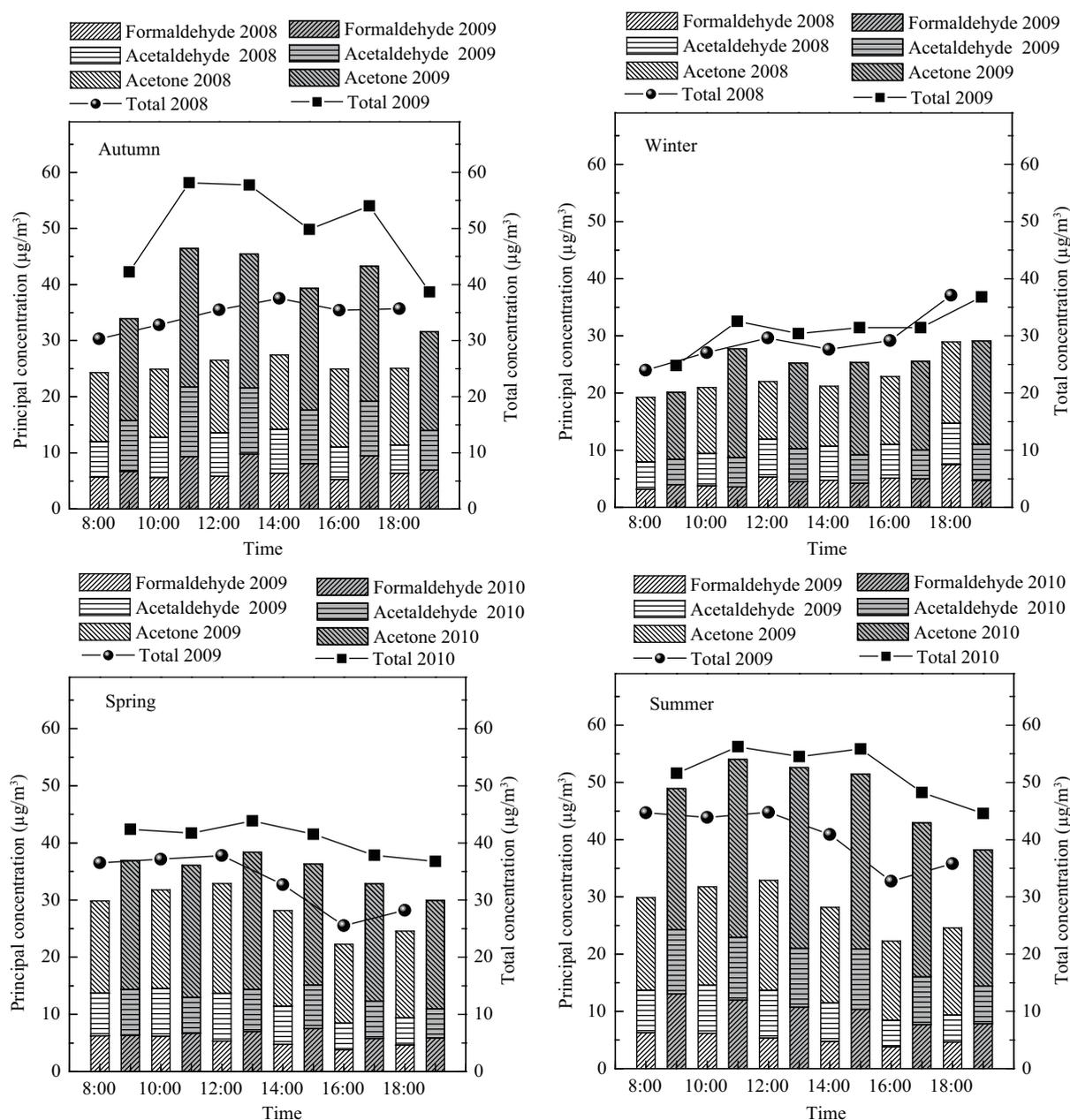


Fig. 1 Average diurnal variations of the three main and the total eight carbonyls in each season during the two consecutive years.

exhibited distinct seasonal and diurnal variations. The average concentrations of the total measured carbonyls during two consecutive years in autumn, winter, spring, and summer were 37.7, 31.3, 39.7, 50.5 $\mu\text{g}/\text{m}^3$, respectively. Maximal values for their diurnal variations usually happened around noontime in seasons other than winter. Both the seasonal and diurnal variations indicated that local photooxidation of VOCs made a great contribution to the observed carbonyls, e.g., the maximum contributions of local photooxidation to formaldehyde and acetaldehyde were calculated to be 70%–80% and 60%–70%, respectively during summer in Beijing according to our another study. Figure 1 also reveals that both the 2-hr average concentrations of the sum of the three principal carbonyls and the total eight carbonyls in each season during the second investigated year (Sep 2009–Aug 2010) were much higher than those of the first year (Sep 2008–Aug 2009).

The increment of the average total carbonyls concentration from the first year to the second year was probably ascribed to the increment of emissions of the primary carbonyls and their precursors, e.g., the number of cars increases by 10% every year in Beijing since 2006 (Liu et al., 2009).

As shown in Table 1, the mean concentrations of formaldehyde and acetaldehyde during each season in this study were significantly lower than our previous investigation (Pang and Mu, 2006), whereas the variation trend of mean acetone concentrations during the investigated years was not as clear as those of formaldehyde and acetaldehyde, even contrary to their trend during winter. The exhaust of vehicles in the urban areas is usually considered as the dominant sources of various pollutants (Anderson et al., 1996; Báze et al., 2003; Dutta et al., 2009), especially for the megacity of Beijing that almost all factories with pollutants emission have been moved outside for purpose

of improving the air quality. The decrease of atmospheric formaldehyde and acetaldehyde during the two investigated years in comparison with the previous year seemed to be contrary to the sharp increment of the number of cars, e.g., the number of cars in 2009 and 2010 increased nearly twice of those in our previous investigation year of 2004 and 2005 (Liu, 2009a). Thus it was suspected that the more strict emission standard enacted for cars in Beijing, such as the vehicles with exhaust emissions failed to meet the European No.1 standard were all-day forbidden on the roads since 1 July, 2008 (BOCOG, 2005; Hao and Wang., 2005), was the main reason for the decrease of atmospheric formaldehyde and acetaldehyde. As for acetone, irregular

emissions, such as evaporation of solvents and paints etc. might be its dominant sources.

2.2 Average diurnal variation of BTEX in each season during the two consecutive years

The seasonal and diurnal variations of BTEX during the two consecutive years are shown in Fig. 2, and the ranges and arithmetic mean concentrations of BTEX are listed in Table 2. In contrast to the carbonyls, the order of the seasonal variation of the mean BTEX concentration were: winter ($31.9 \mu\text{g}/\text{m}^3$) > autumn ($27.2 \mu\text{g}/\text{m}^3$) > spring ($23.2 \mu\text{g}/\text{m}^3$) > summer ($19.1 \mu\text{g}/\text{m}^3$), the minimal values for their diurnal variations usually happened in the

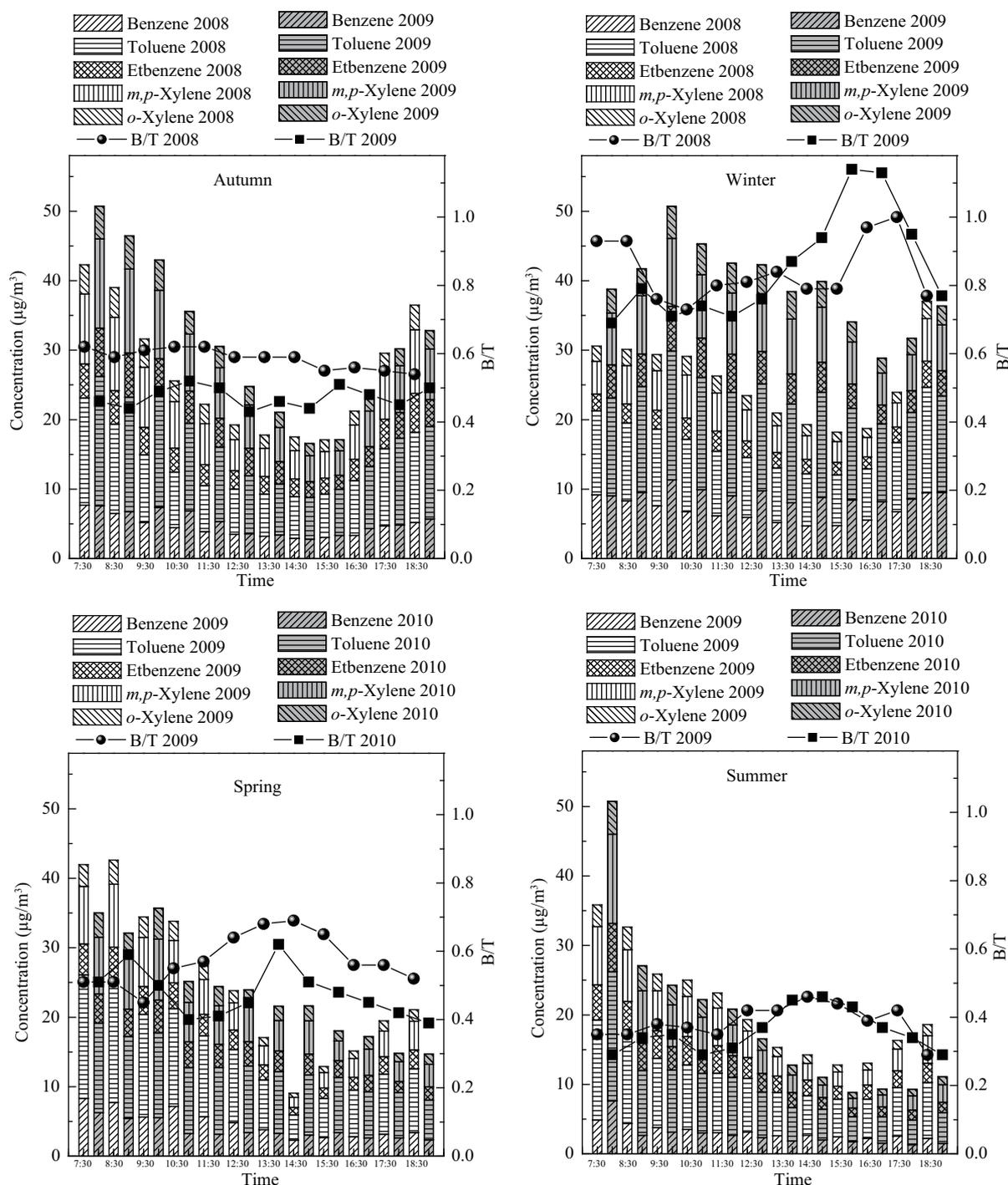


Fig. 2 Average diurnal variation of BTEX and the ratio of benzene/toluene (B/T) in each season during the two consecutive years.

Table 2 Average concentrations ($\mu\text{g}/\text{m}^3$) of BTEX in each season during different investigated years

Season	Year	Number of Samples	Benzene		Toluene		Etbzene		<i>m,p</i> -Xylene		<i>o</i> -Xylene	
			Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	Range	Mean \pm SD	Range
Autumn	2008	590	4.6 \pm 4.4	0.2–35.4	8.5 \pm 8.3	0.4–50.2	3.4 \pm 3.2	0.1–23.4	5.7 \pm 5.0	0.1–27.0	2.8 \pm 2.5	0.1–19.2
	2009	162	5.1 \pm 4.4	0.2–21.2	11.2 \pm 9.6	0.7–44.3	4.1 \pm 3.3	0.2–13.2	7.2 \pm 5.8	0.4–25.5	2.8 \pm 2.2	0.3–8.9
Winter	2008	190	6.9 \pm 6.7	0.9–24.1	10.4 \pm 10.6	0.8–41.0	2.5 \pm 2.5	0.2–11.9	4.9 \pm 4.4	0.4–21.9	2.2 \pm 2.0	0.2–9.9
	2009	140	9.2 \pm 7.6	0.8–31.2	14.5 \pm 13.2	1.2–58.7	4.4 \pm 4.5	0.3–18.2	7.5 \pm 7.1	0.6–34.1	3.5 \pm 3.2	0.3–14.8
Spring	2009	236	4.8 \pm 4.6	0.2–32.3	9.4 \pm 8.6	0.5–42.3	2.7 \pm 2.6	0.1–15.1	4.3 \pm 4.2	0.3–20.0	1.8 \pm 1.7	0.1–7.8
	2010	144	3.7 \pm 3.0	0.2–15.6	9.2 \pm 7.1	0.6–27.3	3.2 \pm 2.6	0.2–11.3	5.3 \pm 4.4	0.3–17.7	2.6 \pm 2.1	0.1–11.1
Summer	2009	218	3.2 \pm 2.3	0.3–11.9	8.6 \pm 5.5	0.8–26.7	3.3 \pm 2.1	0.4–10.2	4.6 \pm 3.4	0.5–15.9	1.9 \pm 1.3	0.3–7.4
	2010	178	2.1 \pm 1.5	0.2–8.0	5.9 \pm 3.6	1.2–17.9	2.3 \pm 1.3	0.5–10.0	3.4 \pm 2.3	0.4–9.9	1.7 \pm 1.1	0.4–9.9

early afternoon, and only the concentrations in winter and autumn during the second investigated year (Sep 2009–Aug 2010) were much higher (ca. 38%) than those of the first year (Sep 2008–Aug 2009). The evident difference between the carbonyls and BTEX was mainly ascribed to their different sources, that is, only primary sources contribute to atmospheric BTEX, while, as mentioned above, the photochemical conversions of VOCs are the dominant sources for atmospheric carbonyls, especially for the seasons other than winter. Besides sources' strength, the seasonal and diurnal variations of atmospheric BTEX in urban areas also strongly depend on meteorological conditions and photochemical activity. The frequent inverse temperature layer during winter season and stable meteorological condition in autumn that prevailed in Beijing, hinder the dilution of the pollutants, and thus led to higher concentrations of BTEX in the atmosphere. The frequent windy days in spring and strong photochemical activity in summer accounted for the low concentration of the observed BTEX. The relatively high concentration during rush hours in the morning and late afternoon implied that automobile exhaust was an important source for atmospheric BTEX in Beijing. The ratio of benzene to toluene (B/T) has been widely used as an indicator for distinguishing their various sources from vehicle emission. Investigating 27 roadside samples from 25 different cities, Barletta et al. (2005) concluded that an average B/T value of 0.6 (wt./wt.) ($\sigma = 0.2$) could be used to characterize vehicle emissions. Liu et al. (2005) also reported the B/T ratio was 0.61 from roadside in Beijing. The B/T values obtained in this study in autumn and spring were well within the reported range of vehicle emission, however, the B/T values in winter and summer were beyond the range. The extremely high B/T values in winter indicated the sources other than vehicle emission made great contribution to atmospheric benzene and toluene, e.g. central heating during winter in Beijing. The B/T ratio higher than unity has been reported for cities mainly dominated by coal burning (Barletta et al., 2005, 2008). Although most of central heating stoves that used coal as energy in Beijing have been replaced by the relatively clean energies of oil or natural gas since 1999, there are still a number of domestic stoves using coal for heating, and the emission from the domestic stoves might be responsible for the observed high B/T ratios in winter. The extremely low B/T in summers during the two investigated years was inconsistent with our previous study (Liu et al., 2010) which revealed that the B/T ratios during 2008 summer were around 0.6. Because strict control measures for the

“green Olympic Games” in 2008 were enacted during the special summer, vehicle emission might become the dominant source for atmospheric benzene and toluene. Thus, we suspected that the sources other than vehicle emission (such as the emissions from painting and gas stations etc. due to the relatively high ambient temperature) during the two investigated summer seasons might make great contribution to atmospheric benzene and toluene

2.3 Exposure and risk assessment

As mentioned in the Introduction section, atmospheric carbonyls and BTEX have adverse impact on human health, and formaldehyde, acetaldehyde and benzene are suspected to be carcinogenic (Báez et al., 2003; Dutta et al., 2009). In order to evaluate the threat to the Beijing inhabitants health caused by carbonyls and BTEX, the non-cancer hazard and integrated lifetime cancer risk (ILTCR) due to the exposure to formaldehyde, acetaldehyde and BTEX were estimated based on the data measured in this study.

The daily exposure (E) of an individual by intake process (only inhalation was considered) was calculated from Eq. (1):

$$E = C \times I_{ra} \times E_{da} / B_{wa} \quad (1)$$

Variable	Description	Value
E (mg/(kg·day))	Daily exposure	
C (mg/m^3)	Concentration of the pollutant	
I_{ra} (m^3/hr)	Inhalation rate*, adult	0.83*
E_{da} (hr/day)	Exposure duration, adult	24
B_{wa} (kg)	Body weigh, adult	65

* The value of inhalation rate (adult) is taken from US EPA (1997).

The ILTCR was calculated from Eq. (2):

$$\text{ILTCR} = E \times \text{SF} \quad (2)$$

where, SF (mg/(kg·day)) is a slope factor of inhalation unit risk for toxics when the exposure-carcinogenic effect is considered as linear, taking from EPA (<http://www.epa.gov/iris>).

Non-cancer risk was expressed as hazard quotient (HQ), which is defined as the ratio of yearly average daily received concentration (LEC) and the reference concentration (RfC).

$$\text{HQ} = \text{LEC}/\text{RfC} \quad (3)$$

If $HQ > 1$, it indicates long term exposure may result in adverse health effects. The RfC for each pollutant was taken from EPA (<http://rais.ornl.gov>) and California Office of Environmental Health Hazard Assessment (<http://www.arb.ca.gov/toxics/healthval/acute.pdf>).

Based on the above equations, the average daily exposure, individual HQ and ILTCR were estimated and listed in Table 3. Among the carcinogens, the ILTCR integrated lifetime cancer risk of formaldehyde was found to be the highest, followed by benzene and then acetaldehyde, and all of them exceeded the value of $1E-06$, indicating significant cancer risk (Dutta et al., 2009). The individual HQs of formaldehyde exceeded unity, indicating chronic health effect. It should be mentioned that the above estimation only considered the outdoor pollution, whereas, due to the delicate decoration, the indoor carbonyls (especially for formaldehyde) and BTEX are more serious than outdoors (Wang et al., 2007) and urban residents spent their more time inside, and hence the estimations can only represent the lower limits. In addition, it is known that the carbonyls and BTEX were the dominant precursors for atmospheric photo-oxidants which will further induce their indirect health impact on residents as well as ecosystems. Therefore, further control measures are urgently needed for mitigating the emissions of these poisonous compounds in Beijing. Because vehicle emission is an important source for BTEX and carbonyls (including the indirect source), and the number of cars in Beijing has exceeded 4.9 million, to effectively mitigate vehicle emission, besides more strict emission standard enacting, the current traffic status of prevailing traffic congestion with high pollutant's emission is urgently needed to be improved.

Table 3 Estimate of individual pollutant exposure, associated non-cancer hazard and cancer risk during the two investigated years

Pollutant	Yearly average concentration (mg/m ³)	Daily average exposure (mg/(kg·day))	HQ	ILTCR
Formaldehyde	6.61E-03	2.03E-03	3.30E+00	9.11E-05
Acetaldehyde	7.50E-03	2.30E-03	8.34E-01	1.84E-05
Benzene	4.72E-03	1.45E-03	1.57E-01	4.19E-05
Toluene	9.58E-03	2.93E-03	2.39E-02	
Ethbenzene	3.29E-03	1.01E-03	3.29E-03	
<i>m,p</i> -Xylene	5.64E-03	1.73E-03	8.06E-03	
<i>o</i> -Xylene	2.74E-03	7.58E-04	3.53E-03	

HQ: hazard quotient; ILTCR: integrated lifetime cancer risk.

3 Conclusions

Distinct diurnal and seasonal variations of atmospheric BTEX and carbonyls at a sampling site in Beijing were observed, implying vehicular exhaust was an important source for BTEX and photochemical conversion of hydrocarbons (especially for the seasons other than winter) was dominant source for carbonyls. Additionally, domestic stoves used coal as energy made significant contribution to BTEX during winter, and the evaporation of painting and solvents etc. also made evident contribution to BTEX

in summer. The obvious yearly increase of the carbonyls and BTEX (especially during autumn and winter) indicated their sources' strength was still increasing, which might be ascribed to the increase of the number of cars. The assessments of health risk based on the data investigated in the two consecutive years revealed that the exposures to the atmospheric benzene, formaldehyde and acetaldehyde exceeded the value ($1E-06$) of the integrated lifetime cancer risk, and the HQ of non-cancer risk of exposure to formaldehyde exceeded unity, indicating significant health risk. Therefore, effective control measures are urgently needed to improve the current pollution status of Beijing.

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