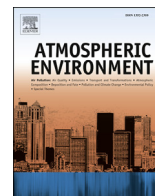




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Aldehydes in relation to air pollution sources: A case study around the Beijing Olympics



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HIGHLIGHTS

- The Beijing Olympic period was unique to study sources of ambient aldehyde.
- Three aldehydes were measured before, during, and after the Olympic period.
- Associations of the aldehydes with other pollutants were examined.
- Sources of the aldehydes were identified through principal component analysis.
- The source control for aldehydes require complex strategies.

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ABSTRACT

This study was carried out to characterize three aldehydes of health concern (formaldehyde, acetaldehyde, and acrolein) at a central Beijing site in the summer and early fall of 2008 (from June to October). Aldehydes in polluted atmospheres come from both primary and secondary sources, which limits the control strategies for these reactive compounds. Measurements were made before, during, and after the Beijing Olympics to examine whether the dramatic air pollution control measures implemented during the Olympics had an impact on concentrations of the three aldehydes and their underlying primary and secondary sources. Average concentrations of formaldehyde, acetaldehyde and acrolein were $29.3 \pm 15.1 \mu\text{g}/\text{m}^3$, $27.1 \pm 15.7 \mu\text{g}/\text{m}^3$ and $2.3 \pm 1.0 \mu\text{g}/\text{m}^3$, respectively, for the entire period of measurements, all being at the high end of concentration ranges measured in cities around the world in photochemical smog seasons. Formaldehyde and acrolein increased during the pollution control period compared to the pre-Olympic Games, followed the changing pattern of temperature, and were significantly correlated with ozone and with a secondary formation factor identified by principal component analysis (PCA). In contrast, acetaldehyde had a reduction in mean concentration during the Olympic air pollution control period compared to the pre-Olympic period and was significantly correlated with several pollutants emitted from local emission sources (e.g., NO_2 , CO, and $\text{PM}_{2.5}$). Acetaldehyde was also more strongly associated with primary emission sources including vegetative burning and oil combustion factors identified through the PCA. All three aldehydes were lower during the post-Olympic sampling period compared to the before and during Olympic periods, likely due to seasonal and regional effects. Our findings point to the complexity of source control strategies for secondary pollutants.

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1. Introduction

1.1. Unique opportunity to collect air pollution data

The Chinese government implemented a series of air pollution control measures to improve air quality during the 2008 Beijing Olympics and Paralympics. Control measures included the reduction of pollutant emissions from factories and industrial facilities and cutting by half the number of private cars on the road, according to an odd/even plate number rule. Additionally, all construction projects were suspended during the Olympic period (M. Wang et al., 2009). These control measures resulted in significant reductions in concentrations of primarily emitted pollutants (e.g., PM_{2.5}, SO₂, NO_x) (Huang et al., 2010; Li et al., 2010a, 2010b; M. Wang et al., 2009; W. Wang et al., 2009; X. Wang et al., 2009; X.S. Wang et al., 2009; Y. Wang et al., 2009; S.X. Wang et al., 2010; T. Wang et al., 2010; Wang and Xie, 2009; Xin et al., 2010; Zhou et al., 2010). However, it is less straightforward whether the same trend occurred for pollutants that had both primary and secondary sources, such as ozone and aldehydes. Our study utilizes a valuable data set before, during, and after the Olympic Games to verify whether, and at what extent, the change in emission source intensities resulted in the abatement of pollution.

Based on the intensity of the air pollution control measures (M. Wang et al., 2009), our study used three periods defined as follows: the pre-Olympic period (June 4th–July 19th) when some light controls were implemented, the during-Olympic period (July 20th–September 19th) when the full-scale control measures were implemented, and the post-Olympic period (September 20th–October 30th) when the control measures were relaxed. Extra control measures were also adopted during the Olympic (August 8th–August 24th) and the Paralympic periods (September 6th–September 17th), which included barring an additional 20% of government-owned cars from traveling on the road, suspending outdoor construction work, and temporarily closing some gas stations. Therefore, the during-Olympic period can be further divided into the sub-period 1 with full-scale control measures (July 20th–August 7th and August 24th–September 5th), and the sub-period 2 with the extra actions described above (August 8th–August 23rd and September 6th–September 17th).

1.2. Ambient concentrations and sources of aldehydes

Aldehydes are reactive compounds that induce adverse health effects in humans and animals (Akbar-Khanzadeh and Mlynek, 1997; Benjebria et al., 1994; Cassee et al., 1996a, 1996b). Although a number of papers have been published assessing the air quality impact of emission controls during the Beijing Olympics (Huang et al., 2010; Li et al., 2010a; M. Wang et al., 2009; Y. Wang et al., 2009; Wang and Xie, 2009), only one paper dealt with formaldehyde and acetaldehyde measured at one quasi-suburban Beijing site, and none on acrolein. In fact, our overall knowledge about ambient acrolein exposure is extremely limited despite the high toxicity of this compound. Aldehydes can be directly emitted into the atmosphere from the incomplete combustion of biomass and fossil fuels (Schauer et al., 2001; Zhang and Smith, 1999), and formed in the atmosphere as a result of photochemical oxidation of reactive hydrocarbons (Altschuller, 1993; Possanzini et al., 2002). Important combustion sources of aldehydes include vehicles, power plants, and residential wood burning (Stahl, 1969; Lipari, 1984). Hence it is important to identify dominant sources in order to set up more effective control strategies. Compared to many other air pollutants (e.g., hydrocarbons, PM mass and certain species), the relative contributions of primary and secondary sources to aldehydes in metropolitan centers has been understudied (Altschuller,

1993; Chan and Yao, 2008; Feng et al., 2005).

To examine whether aldehyde concentrations were reduced during the air pollution control period, we measured formaldehyde, acetaldehyde, and acrolein for approximately one month during each period (pre-Olympic, during Olympic, or post-Olympic). In the during-Olympic period, aldehydes were measured in both sub-periods 1 and 2. Furthermore, in order to better understand the impact of the Beijing Olympic control measures, we also obtained data for numerous other air pollutants at the same monitoring site, and meteorological data (temperature, relative humidity, wind speed, and wind direction) from a nearby site, and analyzed their relationships to the aldehydes.

1.3. Influence of weather, meteorology, and regional sources on pollution in Beijing

Another important factor influencing the concentrations of ambient aldehydes could be meteorological conditions dominating the Beijing region during the summer months (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 summer is largely determined by the meteorology (Streets et al., 2007), in particular, temperature and solar radiation are key factors that control the photochemistry processes (Y. Wang et al., 2009). The influence of wind direction is associated with the origin of air masses transported from the surrounding areas of Beijing while wind speed controls the dispersion of air pollution. In summer months, Beijing typically experiences high temperatures (mean: 27 °C) and relative humidity (mean: 64%), both favoring the photochemical reactions. In the summer, Beijing also has few windy days, which is unfavorable for atmospheric dispersion of air pollutants.

In addition, neighboring regions impact Beijing air quality (W. Wang et al., 2009; X. Wang et al., 2009; T. Wang et al., 2010). PM concentrations, ozone, and sulfate have all been shown to have significant regional contributions. For PM, air masses transported from the south of Beijing have been shown to increase PM concentrations in the region while air masses from the northwest have been shown to decrease PM concentrations.

2. Methods

2.1. Air sample collection, storage, and analysis

Sample collection, storage and analysis were performed in conjunction with the Health Effects of Air pollution Reduction Trial (HEART) study (Zhang et al., 2013). The HEART study included a comprehensive characterization of air pollution before, during, and after the games. All the air samplers and monitors were collocated at a secured spot on the Peking University 1st Hospital campus that served as the clinical base for the health outcome measurements of the HEART study. The hospital was located in the center of Beijing, within the 2nd ring road, 3 km northwest of Tiananmen Square, surrounded by busy streets of local motor vehicle traffic, cyclists, and pedestrians.

2.2. Aldehyde measurement methods

A passive sampling technique was used to collect aldehydes on a 24-h integrated basis. A C18 cartridge (LC-18, 0.5 g/4.5 mL, Supelco, Inc. US) coated with dansylhydrazine (DNSH) was used to collect and derivatize the aldehydes. Samples and field controls were eluted with acetonitrile and aliquots of extracts were analyzed using an HPLC system with fluorescent detection. This method was

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