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Variation of airborne quartz in air of Beijing during the Asia-Pacific Economic Cooperation Economic Leaders' Meeting

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ABSTRACT

Quartz particles are a toxic component of airborne particulate matter (PM). Quartz concentrations were analyzed by X-ray diffraction in eighty-seven airborne PM samples collected from three locations in Beijing before, during, and after the Asia-Pacific Economic Cooperation (APEC) Leaders' Meeting in 2014. The results showed that the mean concentrations of quartz in PM samples from the two urban sites were considerably higher than those from the rural site. The quartz concentrations in samples collected after the APEC meeting, when the pollution restriction lever was lifted, were higher than those in the samples collected before or during the APEC meeting. The quartz concentrations ranged from 0.97 to 13.2 $\mu\text{g}/\text{m}^3$, which were among the highest values amid those reported from other countries. The highest quartz concentration exceeded the Californian Office of Environmental Health Hazard Assessment reference exposure level and was close to the occupational threshold limit values for occupational settings. Moreover, a correlation analysis showed that quartz concentrations were positively correlated with concentrations of pollution parameters PM₁₀, PM_{2.5}, SO₂ and NO_x, but were negatively correlated with O₃ concentration. The results suggest that the airborne quartz particles may potentially pose health risks to the general population of Beijing.

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Introduction

Quartz is the most thermodynamically stable form of silica. It has been suggested in a number of studies that quartz is a lung carcinogen that poses considerable risks to human health (Hessel et al., 2000; Lynge et al., 1986). Studies in Sweden (Westerholm and Scand, 1980), Ontario in Canada (Finkelstein et al., 1982), and elsewhere (Begin et al., 1987;

Emerson and Davis, 1983) published in the 1980s showed that there is a probable relationship between exposure to crystalline silica and the occurrence of lung cancer. The International Agency for Research on Cancer (IARC) classified quartz as a Group 2A chemical (i.e., probably carcinogenic to humans) in 1987 (IARC, 1987). The agency later concluded, from epidemiological data and the results of animal model studies, that there was sufficient evidence for inhaled quartz to be

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classified as a carcinogen to humans exposed to certain occupational sources (IARC, 1996). The agency classified quartz as a Group I chemical (carcinogenic to humans) in 2012 (IARC, 2012).

A great deal of research into the carcinogenic properties of quartz has been performed, but there is still some disagreement about the mechanism through which quartz causes cancer. Borm et al. (2011) suggested that quartz could primarily be genotoxic directly by interacting with target cells or indirectly by causing inflammation (the inflammatory cell-derived oxidants being genotoxic). It has also been suggested in some publications that quartz particles can be phagocytized by cells and accumulate in the perinuclear regions of the cells. The presence of quartz within a cell could interfere with the segregation of the chromosomes when the cell undergoes mitosis, causing anaphase abnormalities to occur (Barrett et al., 1989).

Most systematic carcinogenic risk assessments for quartz have been focused on occupational exposure. For example, it has been suggested that the presence of highly reactive radicals on the surfaces of fractured silica that may be present in certain workplaces make the fractured silica highly cytotoxic (Castranova et al., 1996; Clouter et al., 2001). Dobias et al. (2006) suggested that quartz dust is genotoxic and is an important contributor to the genotoxicity of complex mixtures of fibrogenic respirable airborne particles in ambient air in certain workplaces. Westberg et al. (2013) measured cancer morbidity in Swedish iron foundry workers exposed to quartz. They found significant overall risks of the foundry workers developing lung cancer. However, it has been shown in *in vivo* and *in vitro* toxicological studies (Balduzzi et al., 2004; Porter et al., 2002) that environmental quartz particles may also have important inflammatory effects. A clear association between exposure to quartz and the development of active tuberculosis has been found not only among silicotic individuals but even in people exposed to quartz dust over long periods of time (Mason and Thompson, 2010; Solomon et al., 2000). The risks associated with exposure of the general population to quartz cannot therefore be ignored; but, until now, very few measurements of quartz concentrations in ambient air have been available.

Environmental pollution in China, especially air pollution in some Chinese mega-cities, has been increasing in severity in recent years (Sun et al., 2015; He et al., 2015). For example, several serious haze events occurred in Beijing in January 2013 (Zhang et al., 2014). It has been suggested in many publications that exposure to haze could cause a variety of respiratory diseases (Pavagadhi et al., 2013; See et al., 2006; Tie et al., 2009). Inhalation is the main pathway through which humans are exposed to quartz (US EPA, 1996); therefore, the relationship between quartz concentrations and the occurrence of haze should be investigated. Most previous literature reports on haze have been concerned with black carbon (Wang et al., 2015), heavy metals (Zhou et al., 2014) and aerosols (Sun et al., 2014). To the best of our knowledge, no studies of the exposure of humans to quartz in haze have been published to date.

In the work described here, we collected inhalable particulate fraction (PM₁₀) samples at three sites in Beijing between 2 November and 30 November 2014. The Asia-Pacific

Economic Cooperation (APEC) Economic Leaders' Meeting took place in Beijing entirely within the study period. In order to guarantee the air quality of the 2014 APEC meeting, Beijing and its neighboring regions, including Hebei, Tianjin, Shandong and Inner Mongolia, imposed temporary restrictions, included halting emission-heavy production and limiting cars on the streets based on their license plates (Chen et al., 2015). The aim of the study was to determine the quartz concentrations in the air and to assess how effectively the emission-control measures that were taken decreased atmospheric quartz concentrations during the APEC meeting. The potential risks posed to humans exposed to airborne quartz in Beijing were then discussed.

1. Materials and methods

1.1. Sample collection

Airborne PM₁₀ samples were collected using TH-150a air samplers (Wuhan Tian Hong Instruments Co., Ltd., Wuhan, China). The flow rate of the sampler is 100 L/min. Silver membrane filters (25-mm diameter, 0.45- μ m pore size) were available from Sterlitech Corp (Kent, WA, USA). In total, 87 air samples were collected at an interval of 24 hr in November 2014, which included about 1 week before and 2 weeks after the APEC meeting. The prevailing wind direction for most of the sampling period was northwest. Samples were collected at two urban sites, Haidian District (HD) and Dongcheng District (DC), and one rural site, Changping District (CP). HD is in northwest Beijing, DC is in southeast Beijing, and CP is in north Beijing. Meteorological data were obtained from the Beijing Urban Ecosystem Research Station (part of the Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences). Other meteorological data (including whether or not sampling periods had been affected by haze) were obtained from www.nmc.cn/publish/forecast/china.html.

1.2. Sample analysis

The samples were quantitatively analyzed using NIOSH method 7500 (NIOSH, 2003). The samples contained significant amounts of calcite, so quartz may have been lost through the formation of CaSiO₃. We therefore removed the calcite using the following procedure. A new filter was placed in a filtration apparatus, then the sample filter was removed from the sampling cassette, folded, and placed on top. A mixture of 25% (V/V) HCl and 2-propanol was added to the filter funnel and allowed to stand for 5 min, then a vacuum was applied and the acid and alcohol were slowly drawn through the filter. The filter was then washed with 10 mL distilled water three times before the vacuum was released. Both filters were then placed in a porcelain crucible, which was then loosely covered and baked at 800°C in a muffle furnace for 2 hr. A 10-mL aliquot of 2-propanol was added to the ash, then a glass rod was used to loosen the residue, and all of the residue was transferred to a beaker. The crucible was washed three times, and the washings were added to the beaker; then 2-propanol was added to the beaker. The beaker was covered with a

watch glass and placed in an ultrasonic bath for 3 min. The underside of the watch glass was washed into the beaker with 2-propanol. A fresh silver filter was placed in the filtration apparatus; then, with no vacuum applied, 5 mL of 2-propanol was added to the filter and the sample suspension was added. The beaker was rinsed onto the filter three times. Feathering of the sample outside the deposition area was minimized by allowing the suspension to settle for a few minutes before a vacuum was applied. The saturated filter was placed on a Teflon sheet, which was then warmed on a hot plate to dry the filter. The dry filter was then mounted in an X-ray diffraction sample holder for analysis using an X'pert Pro MPD instrument (PANalytical, Almelo, Netherlands). The instrument was operated using a CuK α anticathode, voltage of 40 kV, current of 40 mA, 2 θ scan range of 5–90°, step size of 0.02626°, and scan speed of 0.10941°/s.

1.3. Quality assurance and quality control

The NIST SRM1878b (Gaithersburg, MD, USA) certified standard was used to produce a calibration curve following the method described in the “calibration and quality control” section of NIOSH method 7500 (Estimated Limit of Detection (LOD): 0.005 mg per sample, range 0.02–2 mg per sample). A laboratory blank sample was analyzed with every 20 samples, and no quartz was detected in any of the blank samples.

2. Results and discussion

2.1. Variations in quartz concentrations before, during, and after the APEC meeting

The quartz concentrations ranged from 0.97 to 13.2 $\mu\text{g}/\text{m}^3$, and the mean was 5.53 $\mu\text{g}/\text{m}^3$. The highest quartz concentration during the whole campaign, 13.2 $\mu\text{g}/\text{m}^3$ at site HD, was found on 20 November, and the lowest quartz concentration, 0.97 $\mu\text{g}/\text{m}^3$ at site CP, was found on 6 November. The quartz concentration was relatively high compared with concentrations that have previously been found. For example, Davis et al. (1984) analyzed quartz in metropolitan areas of the USA and found that the quartz contribution to the total amount of dust varied by more than an order of magnitude, from 1% in Portland (OR) to 10.2% in Winnemucca (NV). The quartz concentration was generally between 1 and 3 $\mu\text{g}/\text{m}^3$ and was found to be unlikely to exceed an annual mean of 8 $\mu\text{g}/\text{m}^3$. Puledda et al. (1999) found quartz concentrations of 0.62–1.51 $\mu\text{g}/\text{m}^3$ (mean 1.05 $\mu\text{g}/\text{m}^3$) in an urban part of Rome (Italy). In another study, in 2007, quartz concentrations of 0.25–2.87 $\mu\text{g}/\text{m}^3$ (mean 1.31 $\mu\text{g}/\text{m}^3$) were found in Rome (De Berardis et al., 2007).

The quartz concentrations at sites HD, DC, and CP were 1.8–13.2 $\mu\text{g}/\text{m}^3$ (mean 6.0 $\mu\text{g}/\text{m}^3$), 1.0–12.1 $\mu\text{g}/\text{m}^3$ (mean 5.7 $\mu\text{g}/\text{m}^3$), and 0.97–8.54 $\mu\text{g}/\text{m}^3$ (mean 4.56 $\mu\text{g}/\text{m}^3$), respectively. The quartz concentrations were generally higher at sites HD and DC than at site CP. Urban–rural gradients in the airborne concentration of other pollutants, such as O $_3$, CO, and NO $_x$ (Barrero et al., 2015) and persistent organic pollutants (Li et al., 2010), have previously been found. The US

Environmental Protection Agency has categorized air pollutant sources as fuel consumption, industrial processes, solid waste (emissions from all process streams), transportation, and miscellaneous (fugitive emissions) (US EPA, 1992). Ambient quartz is emitted to the environment as a component of emitted particulate matter. These emissions can be classed as emissions through process streams, and they are strongly affected by human activities (Cowherd et al., 1988). Differences in population densities, traffic densities, the amounts of construction work taking place, and other factors relating to human activities may cause the quartz concentrations in rural and urban areas to be different.

The airborne quartz concentrations at the three sampling sites before, during, and after the APEC meeting are shown in

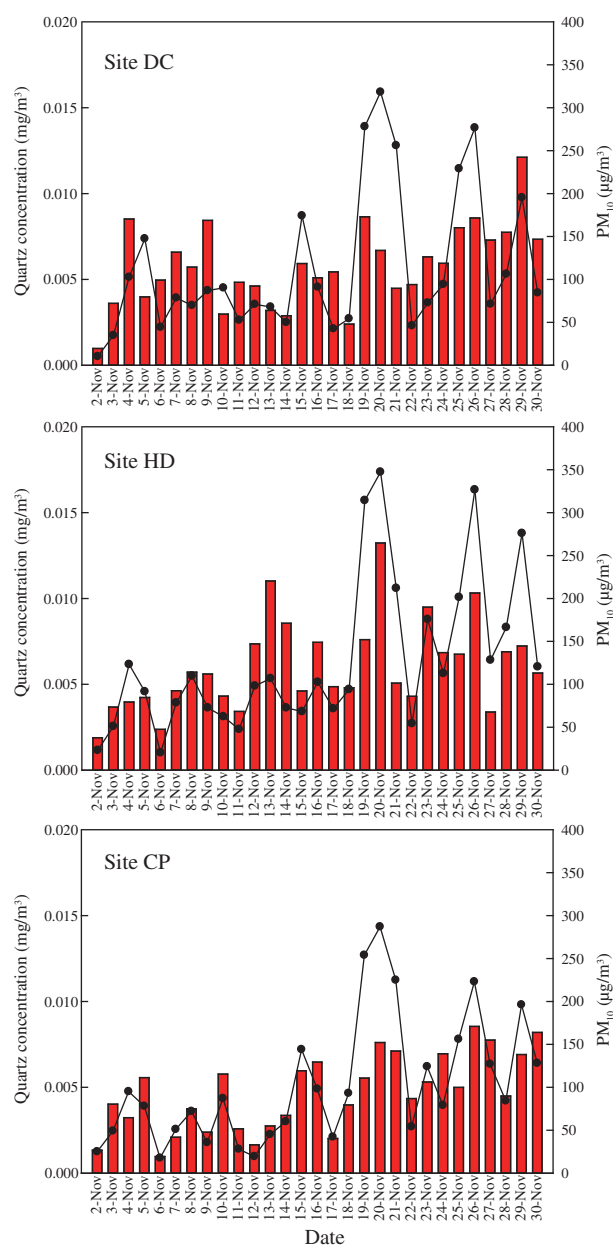


Fig. 1 – Quartz concentrations found in air samples from three sites in Beijing before, during, and after the APEC meeting in Beijing. APEC: Asia-Pacific Economic Cooperation.

Fig. 1. During the meeting, from 5 November to 11 November, the mean quartz concentration at site CP was $3.32 \mu\text{g}/\text{m}^3$, which was similar to the mean concentration before the meeting ($3.21 \mu\text{g}/\text{m}^3$). The mean concentration after the meeting was $5.47 \mu\text{g}/\text{m}^3$, which was almost twice the concentration before the meeting. The same trends were found at sites DC and HD. The mean concentrations before, during, and after the meeting were 4.91, 5.32, and $6.27 \mu\text{g}/\text{m}^3$, respectively, at site DC and 3.68, 4.3, and $6.2 \mu\text{g}/\text{m}^3$, respectively, at site HD. The mean concentration was 22% higher after the meeting than before the meeting at site DC and 56% higher after the meeting than before at site HD. The control measures were stopped once the meeting had ended, and the quartz concentrations increased rapidly. Similar results were found for persistent organic pollutant concentrations when the 2008 Beijing Olympic Games took place (Li et al., 2011). Our results suggest that the emission control measures that were taken quickly caused the concentrations of quartz and some other pollutants in the air to decrease.

2.2. Relationship between the quartz concentration and haze

The quartz concentrations at sites DC, HD, and CP during haze events, shown in Fig. 2, were $4.48\text{--}12.1 \mu\text{g}/\text{m}^3$ (mean $8.08 \mu\text{g}/\text{m}^3$), $5.07\text{--}13.2 \mu\text{g}/\text{m}^3$ (mean $8.37 \mu\text{g}/\text{m}^3$), and $5.54\text{--}8.54 \mu\text{g}/\text{m}^3$ (mean $6.78 \mu\text{g}/\text{m}^3$), respectively. The quartz concentrations at sites DC, HD, and CP when haze was not present were $1.0\text{--}8.52 \mu\text{g}/\text{m}^3$ (mean $5.25 \mu\text{g}/\text{m}^3$), $1.89\text{--}11.0 \mu\text{g}/\text{m}^3$ (mean $5.41 \mu\text{g}/\text{m}^3$), and $0.97\text{--}7.71 \mu\text{g}/\text{m}^3$ (mean $3.9 \mu\text{g}/\text{m}^3$), respectively. The quartz concentrations were significantly higher during haze events than when haze was not present.

Haze is mostly caused by the presence of an excess of particulate matter in the atmosphere. This particulate matter can be emitted from anthropogenic sources or produced through gas-to-particle conversion processes (Zhang et al., 2010; Zhuang et al., 2014). A critical particle size of $<10 \mu\text{m}$ is used in regulations because particles larger than $10 \mu\text{m}$

in diameter are predominantly deposited in the nasopharyngeal region and are rapidly cleared (US EPA, 1996). The PM_{10} concentrations in our samples remained in the range of $10.2\text{--}347 \mu\text{g}/\text{m}^3$ throughout the sampling period. The mean PM_{10} concentration during haze events was higher at site HD, at $279 \mu\text{g}/\text{m}^3$, than at the other sites (259 and $223 \mu\text{g}/\text{m}^3$ at sites DC and CP, respectively). The highest quartz concentrations that were found when haze was not present were 83.2 , 76.3 , and $71.5 \mu\text{g}/\text{m}^3$ at sites HD, DC, and CP, respectively. These data suggest that the PM_{10} concentration was related in some way to the quartz concentration. The quartz and PM_{10} concentrations were positively correlated ($R^2 = 0.59$, $p < 0.0001$), as can be seen in Fig. 3a. The presence of a relationship between the quartz and PM_{10} concentrations would allow exposure to quartz to be predicted when only information on exposure to PM_{10} is available. Fine particles ($\text{PM}_{2.5}$) have been found to be major contributors to PM_{10} during haze events, and $\text{PM}_{2.5}$ have been found to strongly accumulate air pollutants (Sun et al., 2006). The $\text{PM}_{2.5}$ and quartz concentrations were positively correlated ($R^2 = 0.35$, $p < 0.0001$), as shown in Fig. 3b. This relationship was consistent with the relationship between the quartz and PM_{10} concentrations.

High quartz concentrations were occasionally found when haze was not present during the period the control measures were implemented. For example, a quartz concentration of $11.0 \mu\text{g}/\text{m}^3$ was found at site HD on 13 November. We speculated that the meteorological conditions on days when this occurred caused the PM_{10} concentrations to be relatively high but no haze to form for specific (unidentified) reasons.

2.3. Relationship between the quartz concentration and other markers of air quality

The relationships between a number of air quality parameters ($\text{PM}_{2.5}$, SO_2 , O_3 and NO_x concentrations) and the quartz concentrations were investigated. Correlations were found

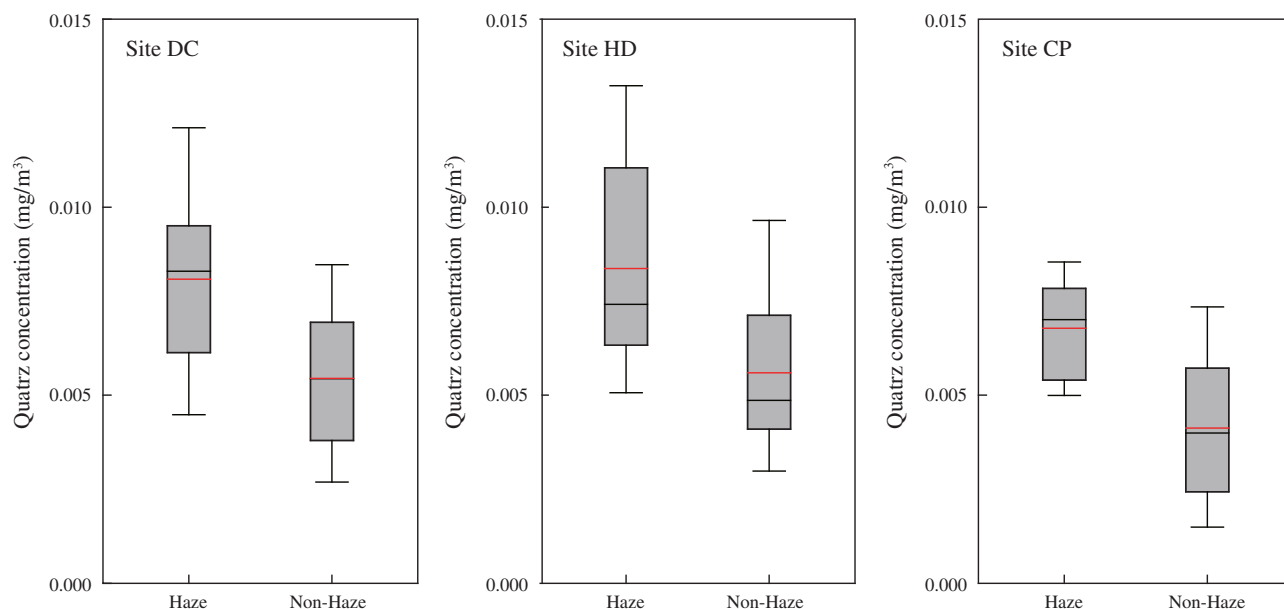


Fig. 2 – Quartz concentrations at three sites in Beijing during haze events and when haze was not present.

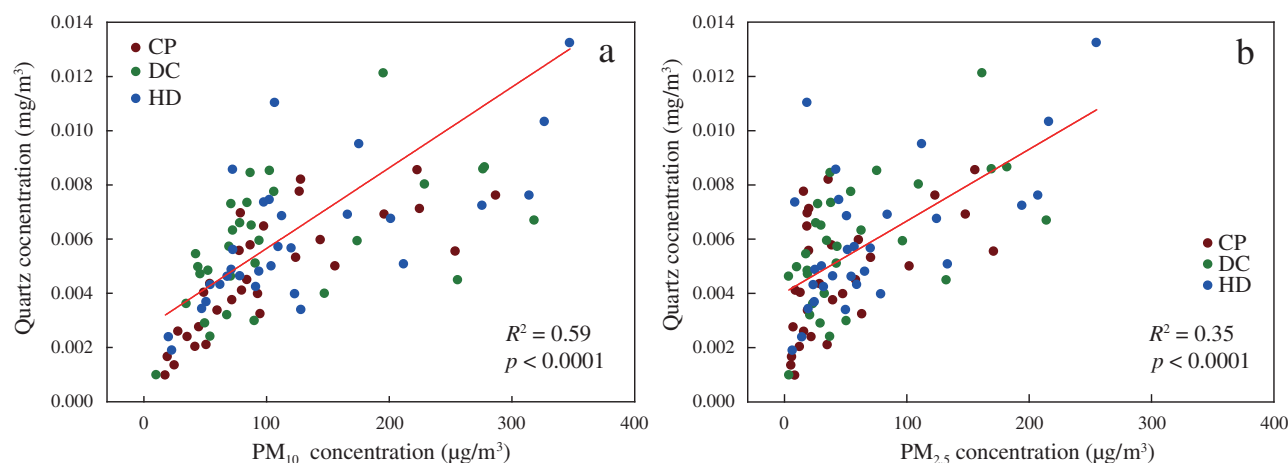


Fig. 3 – Relationships between the quartz and PM₁₀ (a) and PM_{2.5} (b) content at the three sites.

between the concentrations of some of the species, and the correlation coefficients and probabilities that the relationships were significant are shown in Table 1. Positive correlations were found between the PM_{2.5}, SO₂, NO_x and quartz concentrations, and a negative correlation was found between the O₃ and quartz concentrations. O₃ is usually a secondary pollutant, formed through a series of photochemical reactions between NO_x and volatile organic compounds in the atmosphere. The PM_{2.5}, O₃, SO₂, and NO_x in air are primarily emitted in vehicle exhausts, in emissions from construction and industrial sites, and through the combustion of fuel (Wang et al., 2007; Yang et al., 2011). Our results indicate that airborne quartz in Beijing may have similar sources to PM_{2.5}, O₃, SO₂, and NO_x.

2.4. Potential health effects of airborne quartz in Beijing

Quartz particles have been found in the lung parenchyma (Paoletti et al., 1991) and bronchoalveolar lavage fluids (Falchi et al., 1996) of non-occupationally exposed subjects even though only low airborne quartz concentrations are usually found in rural and urban areas. Quartz is therefore one of the most biopersistent components of airborne particulate (Brauer et al., 2001; Churg and Brauer, 2000).

Threshold limit values of 25 and 50 µg/m³ have been set for occupational settings by the American Conference of Governmental Industrial Hygienists (ACGIH, 2006) and the US National Institute for Occupational Safety and Health (NIOSH, 2003), respectively. To the best of our knowledge, the

only reference level that is available for quartz in urban air was proposed by the Californian Office of Environmental Health Hazard Assessment, and a chronic reference exposure level of 3 µg/m³ was adopted in California in 2005 (OEHHA, 2005). The highest quartz concentration that we found was four times higher than this reference exposure level and was close to the threshold limit values, indicating that potential risks posed by airborne quartz to the general population of Beijing cannot be ignored.

3. Conclusions

The quartz concentrations in the air in Beijing were relatively high compared with concentrations that have been found in other countries. The concentrations were higher at the urban sites than at the rural site, and we concluded that human activities are the primary sources of airborne quartz in Beijing. Lower quartz concentrations were found before and during the APEC meeting than after the meeting, suggesting that the air pollution control measures that were taken effectively decreased the quartz concentrations in air in Beijing. Significant correlations were found between the quartz concentrations before, during, and after the meeting and the PM₁₀ ($R^2 = 0.59$, $p < 0.0001$) and PM_{2.5} ($R^2 = 0.35$, $p < 0.0001$) concentrations. Positive correlations were found between the quartz concentrations and the NO_x and SO₂ concentrations, and a negative correlation was found between the quartz and O₃ concentrations, indicating that the airborne quartz had similar sources to NO_x and SO₂. The highest quartz concentration exceeded the Californian Office of Environmental Health Hazard Assessment reference exposure level and was close to the occupational threshold limit values for occupational settings developed by the American Conference of Governmental Industrial Hygienists and the US National Institute for Occupational Safety and Health. We concluded that airborne quartz potentially poses risks to the general population of Beijing and that the carcinogenic risks posed to humans through exposure to airborne quartz should be investigated further.

Table 1 – Correlation coefficients between the quartz concentrations and the concentrations of other pollutants used as markers of air quality.

Correlation	PM ₁₀	PM _{2.5}	SO ₂	NO _x	O ₃
Site DC	.518**	.579**	.344	.373*	-.320
Site HD	.690**	.677**	.400*	.405*	-.316
Site CP	.768**	.493**	.419*	.474**	-.433*

* Significant at $p = 0.05$.

** significant at $p = 0.01$.

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