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Ubiquity of bisphenol A in the atmosphere

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Capsule: This study gives first insight into the sources and global distributions of bisphenol A (BPA) in the atmosphere.

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Abstract

Bisphenol A (BPA) is a suspected endocrine disruptor in the environment. However, little is known about its distribution and transport in the atmosphere. Here, the concentrations of BPA in the atmospheric aerosols from urban, rural, marine, and the polar regions were measured using solvent extraction/derivatization and gas chromatography/mass spectrometry technique. The concentrations of BPA (1-17,400 pg m^{-3}) ranged over 4 orders of magnitude in the world with a declining trend from the continent (except for the Antarctica) to remote sites. A positive correlation was found between BPA and 1,3,5-triphenylbenzene, a tracer for plastic burning, in urban regions, indicating that the open burning of plastics in domestic waste should be a significant emission source of atmospheric BPA. Our results suggest that the ubiquity of BPA in the atmosphere may raise a requirement for the evaluation of health effects of BPA in order to control its emission sources, for example, from plastic burning.

Keywords: Bisphenol A (BPA); Organic aerosols; Plastic burning; 1,3,5-Triphenylbenzene; Size distribution

1. Introduction

Bisphenol A (BPA) is an intermediate in the production of epoxy resins and polycarbonate plastics. It was first synthesized in 1891 and has been widely used since 1950s in food cans and beverage containers including baby bottles (Kaiser, 2007; Vandenberg et al., 2007; Vogel, 2009). During the past several years, BPA has attracted much public attention (Cohen, 2003; Kaiser, 2007; Calafat et al., 2008; Chatterjee, 2008; Collins, 2008; Bucher, 2009; Service, 2009; Vogel, 2009) due to its potential association with adverse health effects such as prostate cancer, obesity, neurobehavioral and reproductive problems (Krishnan et al., 1993; Howdeshell et al., 1999; Hunt et al., 2003; Vandenberg et al., 2007; Lang et al., 2009; Vogel, 2009). Recently, Li et al. (2010) reported a sexual dysfunction in male Chinese factory workers that was linked to exposure to elevated levels of BPA. However, the controversy on

the safety and future of BPA still continues between policy-makers and environmental scientists (Kaiser, 2007; Vogel, 2009).

Many studies have been conducted on BPA in soil and aquatic environments (Staples et al., 1998; Fromme et al., 2002; Kang et al., 2007), as well as the widespread and continuous human exposure to BPA through food, drinking water, dental sealants, cell phones, and inhalation of indoor dusts (Wilson et al., 2001; Rudel et al., 2003; Vandenberg et al., 2007; Calafat et al., 2008; Geens et al., 2009). Studies also have reported that BPA can be found in human serum, urine, amniotic fluid, placental fluid, and umbilical cord blood (Vandenberg et al., 2007). The scientific understanding of BPA has expanded dramatically over the past decade (Vandenberg et al., 2007; Vogel, 2009). However, knowledge about BPA in the atmosphere (Matsumoto and Hanya, 1980; Wilson et al., 2001; Berkner et al., 2004; Matsumoto et al., 2005) to which human are exposed continually is extremely limited.

The atmosphere is a geochemical reservoir of various organic compounds, interacting with the oceans, land, and living organisms including human beings. One important environmental issue is the origin, transport and fate of organic pollutants in atmospheric aerosols and their health effects with responses to human exposure being both acute and chronic (Russell and Brunekreef, 2009). Although its volatility is lower than that of water, BPA can be released into the atmosphere via industrial production with a rate of some 100 t $year^{-1}$ (Staples et al., 1998). Sidhu *et al*. (2005) estimated the emission of BPA to be over ~75,000 kg $year^{-1}$ in the United States based on an uncontrolled domestic waste burning experiment. In addition, BPA could be emitted from the combustion of computer printed circuit boards in electronic waste (e-waste) (Owens Jr. et al., 2007) and from the spraying of paint (Peltonen and Pukkila, 1988).

The objective of this study is to investigate the abundance, spatial and temporal distributions of BPA in the atmospheric aerosols collected at urban, rural and marine sites, as well as the polar regions. The source of BPA will be discussed based on its correlations with some organic tracers. Furthermore, the size distributions of atmospheric BPA are also studied.

2. Experimental section

2.1. Aerosol sampling

More than 260 atmospheric aerosol samples were collected from various cities in Japan, China, India and New Zealand, as well as remote sites including the Pacific, Indian and Atlantic Oceans and the Polar Regions. The marine aerosols were collected during a round-the-world cruise (R/V Hakuho, Nov. 1989 to Mar. 1990). Detailed sample information is given in Table 1. Total suspended particulate (TSP) samples and PM_{10} aerosol (particles having an aerodynamic diameter of <10 μm) samples were collected onto precombusted (450 °C for 6 h) quartz fiber filters (20 cm × 25 cm, Pallflex) using high volume air samplers at a flow rate of ca. 1200 L min^{-1}. $PM_{2.5}$ aerosol samples were collected on quartz fiber filters (ø80 mm) using a medium-volume air sampler. $PM_{0.7}$ aerosol samples were collected at Syowa Station, Antarctica using a high volume impactor sampler (Kimoto model CPS-105) on quartz fiber filters (20 × 25 cm, Pallflex). Three sets of size-resolved aerosol samples were collected at Sapporo, Japan using Andersen 8-stage impactor (Tokyo Dylec Company, Japan) at a flow rate of 120 L min^{-1}. This sampler allows the collection of ambient particles in nine size classes by eight impactor stages (cutoff aerodynamic diameters of 10, 6.4, 4.3, 3.0, 1.9, 1.0, 0.58, and 0.39 μm) and one back-up filter, collecting particles smaller than 0.39 μm. The sampling duration varied from one field campaign to another; ranging from 12 hours for the Chennai samples to one month for the Antarctic samples. All the filters were stored individually in a precombusted glass jar with a Teflon-lined screw cap at a dark freezer room (−20 °C) until the analysis.

2.2. Extraction and derivatization

For each sample, a filter aliquot (5-30 cm^2) was cut into pieces and extracted three times with dichloromethane/methanol (2:1, v/v) under ultrasonication for 10 min. The solvent extracts were filtered through quartz wool packed in a Pasteur pipette, concentrated by the use of a rotary evaporator, and then blown down to dryness with pure nitrogen gas. The extracts were reacted with 50 μL of N,O-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1%

trimethylsilyl chloride and 10 μL of pyridine at 70 °C for 3 h to derive trimethylsilyl derivatives of BPA. During this procedure, OH groups are derivatized to the corresponding trimethylsilyl (TMS) ethers. After the reaction, derivatives were diluted with 140 μL of *n*-hexane containing the internal standard (C_{13} *n*-alkane, 1.43 ng μL^{-1}).

2.3. Gas chromatography/mass spectrometry (GC/MS)

GC/MS has been successfully applied for the determination of BPA in environmental samples such as wastewaters (e.g., Matsumoto et al., 1977; 1982; Ballesteros et al., 2006), seawaters (Li et al., 2001), sediment cores (Kawahata et al., 2004; Peng et al., 2007), combustion products (Owens Jr. et al., 2007), and ambient aerosols (Matsumoto et al., 2005). In this study, the GC/MS analyses of the derivatized total extracts were performed on a Hewlett-Packard model 6890 GC coupled to a Hewlett-Packard model 5973 mass-selective detector (MSD). The GC separation was achieved on a DB-5MS fused silica capillary column (30 m × 0.25 mm i.d., 0.25 μm film thickness) with a GC oven temperature program: temperature hold at 50 °C for 2 min, increase from 50 to 120 °C at a rate of 15 °C min^{-1}, then further increase from 120 to 300 °C at a rate of 5 °C min^{-1} with a final isotherm hold at 300 °C for 16 min. Helium was used as the carrier gas at a flow rate of 1.0 mL min^{-1}. The sample was injected on a splitless mode with the injector temperature at 280 °C. The mass spectrometer was operated in the electron ionization (EI) mode at 70 eV and scanned from 50 to 650 Da. Mass spectral data were acquired and processed with the Chemstation software. Bisphenol A was identified by comparing mass spectra with those of library data and authentic standard. GC/MS response factor of BPA was determined using an authentic standard. Fragment ions of BPA at *m/z* 357 and 372 were monitored and used for quantification.

2.4. Quality assurance/quality control (QA/QC)

All the experiments on extraction and derivatization, and GC/MS analysis were finished during 2008-2009. Although storage experiment of the samples was not conducted for BPA, we analyzed the 1991 Alert aerosol samples for dicarboxylic acids in 1993 (Kawamura et al.,

2010) and 2009 and found that the concentrations of major species such as oxalic acid showed no significant difference (<10%) (Kawamura and Tachibana, unpublished result, 2009). This suggests that no serious degradation occurred during the storage of filter samples for 16 years at −20°C. During each campaign, field blank filters were set in the air sampler, exposed to the air less than one minute without pumping, and recovered into the glass jar with a Telfon-lined screw cap. Field (n = 19) and laboratory (n = 4) blank samples were extracted and analyzed in the same way as ambient samples. Results showed no significant contamination. Target compound (BPA) was not detected in the laboratory blank filters, although it was detectable in some field blank filters (less than 1% of the average concentrations for ambient aerosol samples). The limit of detection (LOD) for BPA in the injected extracts was determined to be 0.3 pg μL^{-1} (S/N = 3). Recoveries for BPA in spiked blank samples (about 10 ng of authentic standard spiked onto precombusted quartz filters) were 96.3 ± 4.0% (n = 5), which were similar to those reported by Li et al. (2001). Recoveries for 1,3,5-triphenylbenzene, sucrose and trehalose were better than 80%. The data reported here were corrected for the field blanks but not for the recovery. Relative standard deviation of the concentrations of BPA based on duplicate analysis was <10%.

3. Results and discussion

3.1. Urban and rural sites

We detected BPA in atmospheric aerosols collected at different geographical locations in the world (Fig. 1). The concentration ranges of BPA are summarized in Table 1, together with previously reported data. The highest level of BPA was observed in PM_{10} aerosols from Chennai and Mumbai, India. In Chennai, the concentration range was 200-17,400 pg m^{-3} (average 4,550 pg m^{-3}). Interestingly, the temporal variation of BPA in the Chennai aerosols collected during both winter and summer was characterized by a clear diurnal pattern with nighttime maxima (Fig. 2). A detailed analysis of the organic molecular compositions of the Chennai aerosols (Fu et al., 2010) suggests that the open burning of municipal wastes

including plastics was very active in Chennai, especially during nighttime. Simoneit et al. (2005) reported that 1,3,5-triphenylbenzene can be used as specific tracer for open burning of plastics, especially when coupled with the presence of the antioxidant tris(2,4-di-*tert*-butyl-phenyl)phosphate (TBPP). In the Chennai aerosols, we detected TBPP in most of the samples with higher concentrations at nighttime than daytime. A good correlation between 1,3,5-triphenylbenzene and TBPP ($R^2 = 0.82$) has been reported elsewhere (Fu et al., 2010). The strong positive correlation between BPA and 1,3,5-triphenylbenzene ($R^2 = 0.85$, Fig. 3a) versus no correlations between BPA and levoglucosan (a tracer for biomass burning, $R^2 = 0.10$) or hopanes (biomarkers for fossil fuel combustion, $R^2 = 0.13$) for the Chennai aerosols suggest that the open burning of domestic plastic wastes could be a significant emission source of atmospheric BPA in this region. Similar positive correlations were found in aerosols from other urban areas in India, China, and New Zealand (Fig. 3).

In Beijing (BJ), China, concentrations of BPA in $PM_{2.5}$ aerosols (380-1,260 pg m^{-3}, average 630 pg m^{-3}) are 5-7 times lower than those in the Indian cities (Table 1 and Fig. 1). At Yufa, a suburban site near Beijing, the concentrations (230-860 pg m^{-3}, 500 pg m^{-3}) are similar to those in Chinese urban areas, but are much higher than those at the summit of Mt. Tai (100 pg m^{-3}) in Central East China, and a rural site (10 pg m^{-3}) in Bavaria, Germany (Berkner et al., 2004). In the Pearl River Delta region of South China, the level of BPA is higher in Guangzhou (GZ) than in Zhaoqing (ZQ) and Hong Kong (HK). This difference is reasonable because there are several large e-waste recycling sites (e.g., Qingyuan and Guiyu) (Ni and Zeng, 2009) that are closer to GZ than to ZQ and HK. The e-waste disposal and burning should be an important source of atmospheric BPA in this region. In Sapporo, Japan, the levels of BPA are similar to those reported in Osaka (Matsumoto et al., 2005). In Christchurch, New Zealand, they are much higher than in Auckland. The elevated levels of BPA in Christchurch may be partly explained by the city's geographical (a basin surrounded by hills) and meteorological (formation of inversion layer) conditions. In the United States, Wilson *et al*. (2001) reported that the ambient BPA concentrations (outdoor samples) in several child day care centers in central North Carolina ranged from <100 to 2,500 pg m^{-3},

while in the majority of the air samples they were below the detection limit (100 pg m^{-3}). Our results show that the atmospheric levels of BPA in Indian megacities are roughly one order of magnitude higher than those in China, Japan, New Zealand, and the United States, suggesting that the human exposure of BPA in South Asia is more serious than other regions. It should be noted that BPA is present in both fine and coarse particles, a point to be discussed in Section 3.4. Thus, the atmospheric levels of BPA in $PM_{2.5}$ aerosols from Chinese megacities may be underestimated when compared with those in PM_{10} and TSP from Indian and other countries.

3.2. Marine sites

In the marine aerosols collected during a round-the-world cruise of R/V Hakuho, the highest level of BPA was observed off the coast of the Asian continent (Fig. 1). The general decrease of BPA from the Asian coastal region to the central North Pacific Ocean indicates that the Asian continent is a strong "emitter" of BPA, which can be transported long distances by the westerly winds. Such a point is further supported by the observation in the remote island Chichi-Jima (27°04'N, 142°13'E) in the western North Pacific, where the level of BPA is also higher than those in the central North Pacific. The results of 10-day air mass back trajectory analyses, together with terrestrial higher plant biomarkers, showed that the marine aerosols over the western North Pacific during winter/spring were transported from the Asian continent under the influence of westerly winds (Kawamura et al., 2003). At the Rishiri Island (45°04'N, 141°07'E), northern Japan, the concentration range of BPA (4-32 pg m^{-3}, average 15 pg m^{-3}) is in agreement with the observed concentrations on the coast off the western North and Central America. Higher levels of BPA were found in the Asian coastal regions than in the central North Pacific and central North Atlantic (Fig. 1). This pattern further suggests an atmospheric transport of BPA from continents to remote sites.

3.3. Polar regions

The polar atmosphere was once believed to be extremely clean. In 1950s, pilots flying over the North American Arctic observed a widespread Arctic haze (Law and Stohl, 2007). Arctic haze is a mixture of sulfate, ammonium, nitrate, black carbon, and particulate organic

matter including persistent organic pollutants (Law and Stohl, 2007; Fu et al., 2009). In the Arctic aerosols collected at Alert (82°30'N, 62°18'W) in 1991, the levels of BPA ranged from 1 to 11 pg m^{-3} with higher concentrations in the dark winter than in the early summer (Fig. 4). The observed temporal pattern is characterized by a winter maximum and summer minimum, which is similar to those of biomass burning tracers (e.g., levoglucosan) and other anthropogenic organics such as hopanes (biomarkers of petroleum) and polycyclic aromatic hydrocarbons (PAHs) (Fu et al., 2009). This pattern suggests that BPA can be transported from the mid-latitudes in Eurasia and North America to the Arctic, because the Arctic can act as a cold sink during winter to receive the aerosols and their precursors via long-range atmospheric transport (Law and Stohl, 2007). In addition, there was a slight decadal increase in the abundance of BPA from 1991 (average 5 pg m^{-3}) to 2000 (7 pg m^{-3}) at Alert.

In contrast to the Arctic, Antarctica is an isolated continent surrounded by the Southern Ocean, in which long-range atmospheric transport of anthropogenic pollutants is less significant. However, BPA was also detectable in submicron particles ($PM_{0.7}$) at Syowa Station in Antarctica, within the range of those in the Arctic aerosols. It may have been derived from the southern continents through long-range transport possibly via the large scale tropospheric meridional circulation (Bendle et al., 2007), and to a lesser extent from local emission sources such as the scientific stations in Antarctica.

The global scale decline of BPA from Asian continent to remote sites suggests that dry and wet depositions of particles may remove BPA from the atmosphere. Another possible scavenging process is the photodegradation of BPA during long-range atmospheric transport (Staples et al., 1998). The photooxidation products could be phenol, 4-isopropylphenol, and a semiquinone derivative of BPA (Howard, 1989). Further, atmospheric dilution of BPA during long-range transport could be responsible for the very low concentrations of BPA over the remote areas. It should be noted that because some samples from marine and polar regions were collected almost 20 years ago, the spatial distributions of BPA could be in part affected by the time spans of the sampling periods.

3.4. Size distributions

The size distribution of organic compounds is a key factor in controlling their chemical and physical properties and potential health effects (Russell and Brunekreef, 2009). The fine fraction of atmospheric aerosols can penetrate deep into the human respiratory system and cause health problems (Cormier et al., 2006). For example, Pope *et al*. (2009) reported that an increase in the concentration of fine particulate matter ($PM_{2.5}$) by 10 μg m^{-3} causes a decrease in estimated mean life expectancy of 0.61 year in the United States. Recent toxicological and epidemiological studies have shown that there is still much inconsistency regarding which size fraction of aerosols can be taken up by the human body (Russell and Brunekreef, 2009). Nevertheless, little is known about the size distribution of BPA in the atmosphere.

In Fig. 5a, a bimodal size distribution is shown for BPA in the urban aerosols from Sapporo, Japan (July 1-4 and 12-14), with two maxima in fine (<1.9 μm) and coarse (>6.4 μm) modes. In contrast, we found a broad peak of 1,3,5-triphenylbenzene in fine modes (<1.9 μm) (Fig. 5b). The difference in size distribution patterns between BPA and 1,3,5-triphenylbenzene indicates that atmospheric BPA in the coarse mode may be derived from sources other than the burning of municipal wastes or plastic emission. During a period (July 4-7) of high BPA loading, the majority of BPA (62%) was found in the size of >1.9 μm. Interestingly, we found that the size distributions of two sugar compounds (sucrose and trehalose), the tracers for soil resuspension (Simoneit et al., 2004), showed major peaks in the coarse mode (Figs 5c and 5d). Moreover, the levels of these sugars were extremely high during July 4-7, which is in accordance with the high BPA level. Thus, soil resuspension could be a potential source for atmospheric BPA in the coarse fraction. Soils have been reported to contain a certain level of BPA (0.7-44.5 ng g^{-1}) (Sánchez-Brunete et al., 2009).

4. Conclusions

The detection of BPA in ambient aerosol samples from urban, rural, marine and the polar regions indicates that it is a ubiquitous component in the atmosphere. The open burning of plastics in domestic wastes was found to be a significant emission source of atmospheric BPA

in urban regions. The size distributions of BPA showed peaks in both fine and coarse fractions. Soil resuspension is suggested as a main source for BPA in the coarse fraction. In addition, the atmospheric level of BPA detected in this study is comparable to the well-known carcinogenic PAHs such as benzo[α]pyrene (BaP) in ambient air. This similarity raises an open question on whether or not such a low level of atmospheric BPA causes a negative health effect over the time of prolonged exposure. By analogy to its serious concerns in aquatic media and daily-used materials to which people are exposed, the ubiquity of BPA in the Earth's atmosphere suggests a potential environmental concern for the future. This potential is especially true in South Asia, where the atmospheric levels of BPA are very high.

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397 Table 1. Detailed information on the aerosol samples and bisphenol A (BPA) concentrations (pg m^{-3}).

Aerosol Sample				Concentration		Reference
Location	Sampling Date	Type	Number	Range	Average	
Urban Site						
Chennai, India	Winter and summer, 2007	PM_{10}	49	200-17,400	4,550	This study
Mumbai, India	Winter and summer, 2008	PM_{10}	24	100-9,820	2,480	This study
Beijing, China	August, 2007	$PM_{2.5}$	10	380-1,260	630	This study
Yufa, China	August 2007	$PM_{2.5}$	10	230-860	500	This study
Guangzhou, China	Winter and summer, 2007	$PM_{2.5}$	15	70-2,340	480	This study
Zhaoqing, China	Winter and summer, 2007	$PM_{2.5}$	15	20-1,980	400	This study
Hong Kong, China	Winter and summer, 2007	$PM_{2.5}$	30	30-690	240	This study
Osaka, Japan	Oct. 2000-Mar. 2001	TSP	36	10-1,920	510	(Matsumoto et al., 2005)
Sapporo, Japan	Summer, 2008/2009	TSP	7	70-930	340	This study
Auckland, New Zealand	Jun.-Jul., 2004	PM_{10}	19	4-1,340	170	This study
Christchurch, New Zealand	Jun.-Jul., 2004	PM_{10}	18	95-1,480	520	This study
North Carolina, U.S.	Spring, 1997	PM_{10}	3	<100-2,500	880	(Wilson et al., 2001)
Rural Site						
Mt. Tai, Central East China	June, 2006	TSP	5	30-240	100	This study
NE-Bavaria, Germany	May-Nov., 2001	TSP	7	5-15	10	(Berkner et al., 2004)
Marine Region						
North Pacific Ocean	Nov., 1989	TSP	4	1-2	2	This study
California Coast	Nov.-Dec., 1989	TSP	3	8-16	11	This study
North Atlantic Ocean	Dec., 1989	TSP	2	4-6	5	This study
Indian Ocean	Feb., 1990	TSP	1	-	6	This study
South China Sea	Feb., 1990	TSP	1	-	6	This study
East China Sea	Feb.-Mar., 1990	TSP	2	7-27	17	This study
Chichi-Jima Is., Western North Pacific	Jun.-Oct., 1990	TSP	9	2-23	8	This study
Rishiri Is., North Japan Sea	September, 2003	TSP	17	4-32	15	This study
The Polar Region						
Alert, Canadian High Arctic	Feb.-Jun., 1991	TSP	16	1-11	5	This study
Alert, Canadian High Arctic	Feb.-Apr., 2000	TSP	9	2-17	7	This study
Syowa Station, Antarctica	Feb.-Dec., 1991	$PM_{0.7}$	2	1-11	6	This study

398

Figure captions

Fig. 1. Spatial distributions of the mean concentrations of bisphenol A (BPA) in atmospheric aerosols collected at different locations in the world. Urban and rural sites are presented in red, while marine and remote sites including the polar regions are in white, including a rural site at Bavaria with extremely low level of BPA that has been reported by Berkner *et al*. (2004). The BPA levels of outdoor air samples in several child day care centers in both urban and rural areas in North Carolina (NC) were reported by Wilson *et al*. (2001). The green triangle shows the location of Mt. Tai (1534 m, a.s.l.) in Central East China. The green stars show the sampling sites of Rishiri Island off the coast of Hokkaido, Japan, and Chichi-Jima Island in the western North Pacific.

Fig. 2. Temporal variation of bisphenol A (BPA) detected in the urban aerosols from Chennai, South India. The shaded circles represent the night-time samples.

Fig. 3. Positive correlations between the concentrations of 1,3,5-triphenylbenzene, a tracer for plastic burning (Simoneit et al., 2005), and BPA in urban aerosols from (a) Chennai, India, (b) Mumbai, India, (c) Beijing, China, (d) Guangzhou, China, (e) Zhaoqing, China, (f) Hong Kong, China, (g) Auckland, and (h) Christchurch, New Zealand.

Fig. 4. Temporal variation of bisphenol A detected at Alert in the Canadian High Arctic in 1991.

Fig. 5. Size distributions of organic compounds detected in urban aerosols from Sapporo, Japan in the summer of 2008. (a) bisphenol A, (b) 1,3,5-triphenylbenzene, (c) sucrose, and (d) trehalose.

425 **Fig. 1** (Fu and Kawamura)

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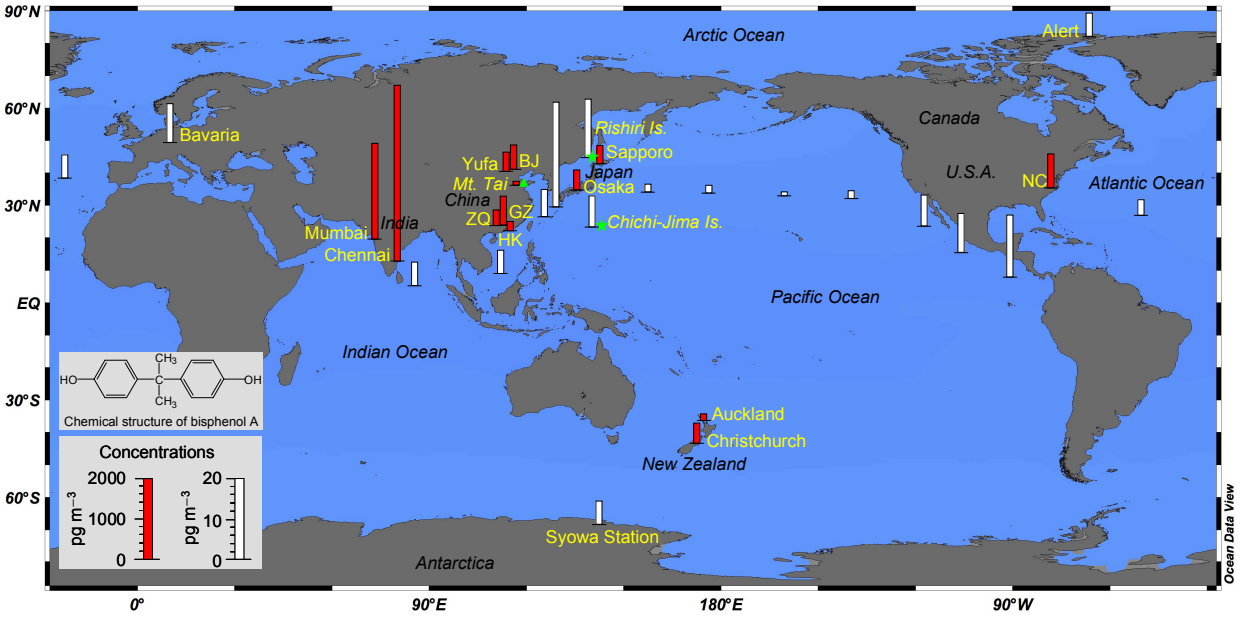

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429 **Fig. 2** (Fu and Kawamura)

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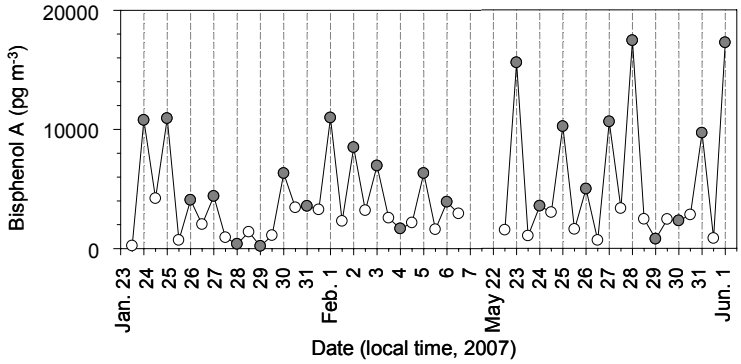

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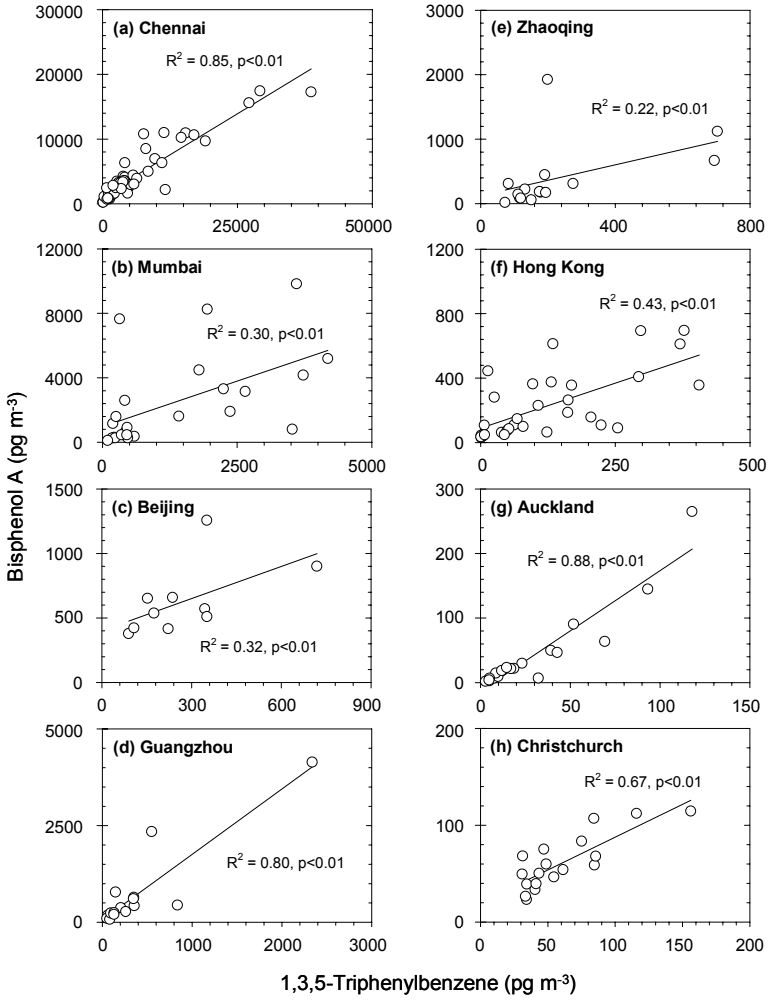
Fig. 3 (Fu and Kawamura)



436 **Fig. 4** (Fu and Kawamura)

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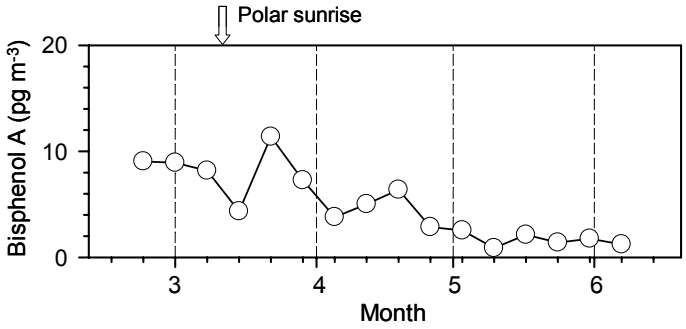

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440 **Fig. 5** (Fu and Kawamura)

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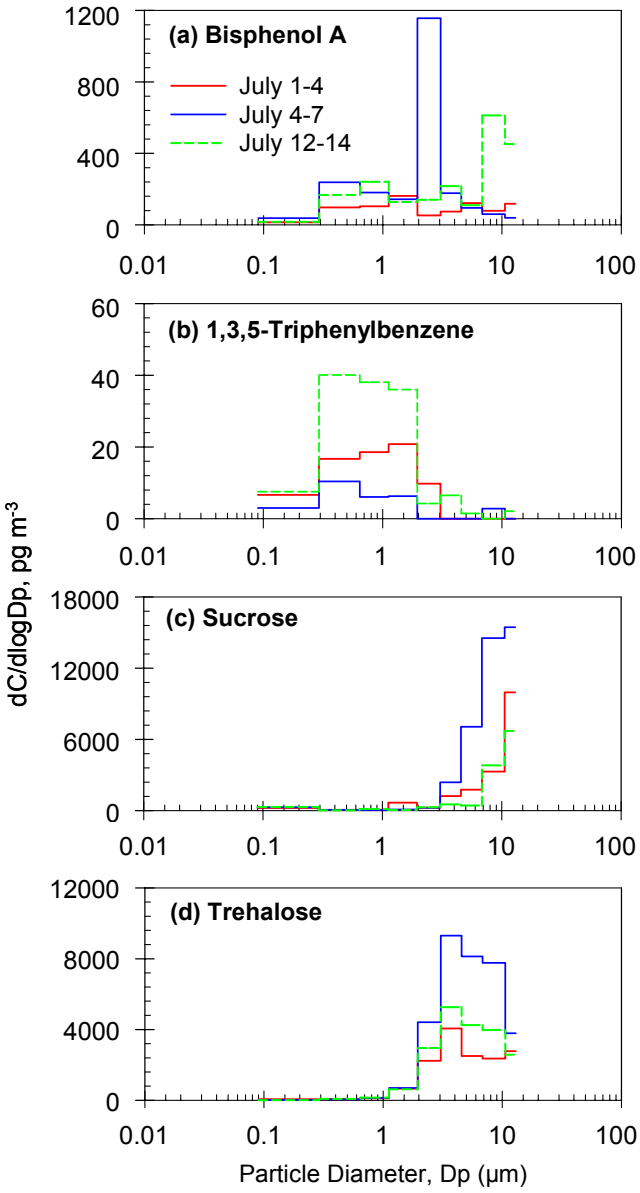

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