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ScienceDirect

Journal of Environmental Sciences 2011, 23(7) 1199–1204

JOURNAL OF ENVIRONMENTAL SCIENCES
ISSN 1001-0742
CN 11-2629/X
www.jesc.ac.cn

Polybrominated diphenyl ether (PBDE) in blood from children (age 9–12) in Taizhou, China

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Received 30 September 2010; revised 24 November 2010; accepted 18 January 2011

Abstract

Polybrominated diphenyl ethers (PBDEs) as ubiquitous persistent organic pollutants have attracted much attention in recent years. Exposure to PBDEs could induce a high health risk for children. The aim of this study was to investigate the PBDEs exposure of children (9–12 years) from Taizhou, China. Fifty-eight blood samples were collected in one school in a mountainous area in Taizhou. The concentrations of $\sum_9$PBDEs (sum of BDE-28, -47, -99, -100, -153, -154, -183, -197 and -209) ranged from 2.66 to 33.9 ng/g lipid wet (lw) with a median of 7.22 ng/g lw. These concentrations were lower than those of children in USA, but close to European and Asian general population levels. The results showed that children in Taizhou countryside were at a low level of PBDEs exposure. The predominant congener was BDE-209, followed by BDE-28, -47, -197 and -153. High abundance of BDE-209 was consistent with the pollution background of PBDEs in China characterized by high brominated congeners as main pollutants.

Key words: polybrominated diphenyl ether; children; blood; exposure; Taizhou

DOI: 10.1016/S1001-0742(10)60553-0

Citation: Zhang X Y, Ruan X L, Yan M C, Zhao Y X, Wei W J, Qin Z F et al., 2011. Polybrominated diphenyl ether (PBDE) in blood from children (age 9–12) in Taizhou, China Journal of Environmental Sciences. 23(7): 1199–1204

Introduction

Polybrominated diphenyl ethers (PBDEs) are widely used in a variety of consumables, such as textiles, automotive parts, construction materials, printed circuit, television, computer housings and other electronic household equipments (Darnerud et al., 2001). Since PBDEs are not chemically bonded to the materials, which can easily escape from the materials and diffuse into the environment, they are broadly found in the environment and biotic samples including human tissues. On account of the potential human health risk of PBDEs, the manufacture and use of some PBDEs have been limited or banned. Technical production of penta-BDE and octa-BDE were withdrawn from the U.S. marketplace in 2004. And then in May of 2009 in Geneva, Switzerland, the commercial penta-BDE and octa-BDE have been added into the Annex A at the fourth session of the Stockholm Convention Conference of the Parties. Although it has been evidently indicated that Deca-BDE can enter into human body (Inoue et al., 2006; Bi et al., 2007; Jin et al., 2009), deca-BDE is still produced and applied in a large amount.

Animal experiments have shown that PBDEs have neurodevelopmental toxicity and thyroid disruption (Stoker et al., 2004) effects. In recent studies, PBDEs have been found in some human tissues, such as liver, breast, blood, serum, breast milk, fetal cord blood and placenta (Frederiksen et al., 2009). Humans are mainly exposed to PBDEs via food intake, air inhalation or ingestion of dust. PBDEs exposure of children has attracted much more attention because of the high susceptibility of children relative to adults. Exposure to PBDEs in children is primarily via the diet (including breast feeding), as well as dust or inhalation (Lorber, 2008). The data of exposure levels in Chinese children are extremely scarce, therefore it is necessary to investigate the exposure conditions of Chinese children.

Taizhou City of China is known as the location of some electronic waste (e-waste) recycling sites. Previous studies reported that residents from an e-wastes recycling site in Taizhou had high PBDEs levels (Leung et al., 2010; Zhao et al., 2010). In this study, blood samples of children

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were collected from a mountain area away from e-waste recycling sites in Taizhou, and investigated the levels and congener profiles of PBDEs.

1 Materials and methods

1.1 Sample collection

Fifty-eight blood samples (22 girls, 36 boys) were collected from an elemental school in a Taizhou mountain area, Zhejiang Province, China in Nov. 2008. Each child was required to complete a form regarding the age and sex. The ages ranged from 9 to 12 years old. Informed content was obtained from all the children.

1.2 Chemicals

A standard solution of PBDE congeners (EO-5278 and BDE-197) (Cambridge Isotope Laboratories, USA) was prepared and applied for the quantification of our aimed congeners (BDE-28, -47, -99, -100, -153, -154, -183, -197 and -209). The surrogate standards used were BDE-71 and $^{13}C_{12}$BDE-209 (Cambridge Isotope Laboratories, USA). *n*-Hexane, methylene dichloride, acetone, and nonane were pesticide grade and purchased from Mallinckrodt Baker (USA). Anhydrous sodium sulfate, sulfuric acid, sodium hydroxide, and anhydrous ethanol were analytical grade (Beijing Chemical Factory, China).

1.3 Extraction and clean-up

The methods of PBDEs extraction and gravimetric lipid determination were developed based on the method of neutral compounds extraction from serum (Hovander et al., 2000). Briefly, surrogate standards (BDE-71, $^{13}C_{12}$-BDE-209) were added to the samples, and then hydrochloric acid and isopropanol were added in order following with shaking. The samples were subsequently extracted twice with hexane/methyl *t*-butyl ether (MTBE) (1:1, V/V). The combined organic phases were washed with a potassium chloride water solution, followed by evaporation to dryness for gravimetric determination of extracted lipid content. The concentrated extract was cleaned by passing through a 15-mm i.d. (diameter) column, which was packed, from the bottom to top, with 1 g activated silica gel, 8 g acid silica gel (40% concentrated sulfuric acid, W/W), 1 g activated silica gel, and 1 cm anhydrous sodium sulfate. The samples were followed by rotary evaporation to about 1–2 mL volume. Then under a gentle nitrogen stream the sample volumes were reduced to 0.1 mL. The final extract was transferred to GC vials. Throughout the extraction, cleanup and analysis procedure, the analytes were protected from light by wrapping the containers with aluminum foil or by using amber glassware.

1.4 Gas chromatography-mass spectrometry

The samples were analyzed on an Agilent 6890 series gas chromatograph coupled to an Agilent 5973 mass spectrometer (USA) using negative chemical ionization (NCI) in the selected ion monitoring mode. A DB-5 MS (15 m × 0.25 mm i.d., 0.25 μm film thickness) capillary column was used for the determination of PBDE congeners. Methane was used as a chemical ionization moderating gas and helium as the carrier gas at a flow rate of 1 mL/min. The ion source and interface temperatures were set to 150 and 300°C, respectively. The GC oven temperature program was carried out as follows: initial temperature 100°C held for 1 min, increased to 200°C at 10°C min, and then to 300°C at 20°C min, held for 20 min. One micro liter injection was made in the pulse splitters mode, with a purge time of 1 min (injector temperature 265°C. PBDEs (except BDE-209) were monitored with the m/z responses of 79/81 (bromide ions). BDE-209 and ^{13}C-BDE-209 used m/z responses of 486.6/484.6, 496.6/494.6, respectively.

1.5 QA/QC and data analysis

Sample recovery was evaluated using surrogate standards BDE-71 and ^{13}C-labled BDE-209, and the average recoveries were 64% and 69%, respectively. Analyte values were corrected for recovery. A procedural blank was run in parallel with every batch samples. The detection limits (LOD) were defined as a signal of three times the noise level. The LOD were 0.10–0.54 ng/g lipid weight (lw) for tri- to nona-BDEs and 3.47 ng/g lw for deca-BDE. The concentrations of those congeners below the LOD were regarded as 1/2 LOD.

2 Results and discussion

2.1 PBDEs levels in children's blood

A total of 58 blood samples from Taizhou countryside were analyzed. The range and median concentrations of 9 individual PBDE congeners (BDE-28, -47, -100, -99, -154, -153, -183, -197 and -209) as well as total PBDEs, are presented in Table 1. The median level of ∑PBDEs in all blood samples was 7.22 ng/g lw, ranging from 2.66 to 33.9 ng/g lw. Table 2 shows several studies concerning PBDEs levels in children. Compared with previous reports, PBDEs concentrations in the present study were lower than those of children in USA (59.6–418 ng/g lw) (Windham et al., 2010; Lunder et al., 2010; Fischer et al., 2006) and Mexico (29.5 ng/g lw) (Pérez-Maldonado et al., 2009), but close to that in European (5.00 ng/g lw) (Fängström et al., 2005). To date, the data of PBDEs levels in Chinese children is limited. Chen et al. (2010) have detected the PBDEs levels in 29 blood samples from 0–11 years old children in Dalian and found the median levels of main PBDE congeners of BDE-47 (6.00 ng/g lw), BDE-99 (5.27 ng/g lw) and BDE-153 (8.25 ng/g lw), which were higher than the levels of present study for Taizhou children. Recently, Shen et al. (2010) have investigated the PBDEs levels in children from three areas of Taizhou, and found that the PBDEs level in children of Luqiao, an e-waste recycling site, was obviously higher than those of children from non e-waste recycling areas. The result corresponds to this study. The PBDEs levels in Taizhou children in our study was comparable to PBDEs values reported for children from non e-waste recycling areas, and lower than those of children from the e-waste recycling site. Besides, the

Table 1 PBDEs concentrations, median concentration and range in blood samples of children in Taizhou* (unit: ng/g lw)

Sample number	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183	BDE-197	BDE-209	∑PBDEs
1	0.27	0.10	0.05	0.05	0.06	0.08	0.07	0.26	1.73	2.66
2	0.61	0.40	0.05	0.05	0.06	0.27	0.07	0.26	1.73	3.50
4	0.64	0.52	0.05	0.05	0.06	0.25	0.07	0.26	1.73	3.63
3	0.85	0.10	0.05	0.05	0.06	0.48	0.07	0.26	1.73	3.64
6	0.61	0.32	0.05	0.05	0.06	0.57	0.07	0.26	1.73	3.72
7	0.61	1.01	0.05	0.05	0.06	0.08	0.07	0.26	1.73	3.92
5	0.27	0.93	0.05	0.05	0.06	0.56	0.07	0.26	1.73	3.97
8	1.39	0.10	0.05	0.05	0.06	0.29	0.07	0.26	1.73	3.98
9	0.84	0.63	0.05	0.05	0.06	0.33	0.07	0.26	1.73	4.02
11	0.91	0.10	0.10	0.21	0.25	0.40	0.07	0.26	1.73	4.04
10	1.02	0.43	0.05	0.05	0.06	0.39	0.07	0.26	1.73	4.05
12	1.02	1.34	0.05	0.05	0.06	0.08	0.07	0.26	1.73	4.66
13	1.01	0.89	0.05	0.05	0.06	0.85	0.07	0.26	1.73	4.95
14	1.04	1.25	0.05	0.05	0.06	0.61	0.25	0.26	1.73	5.29
15	0.87	1.23	0.05	0.27	0.06	0.62	0.29	0.26	1.73	5.37
16	0.90	1.01	0.05	0.05	0.06	0.57	0.07	0.97	1.73	5.41
17	2.45	0.29	0.05	0.05	0.06	0.75	0.07	0.26	1.73	5.70
28	0.27	0.60	0.05	0.13	0.06	0.50	0.31	0.26	3.68	5.85
29	0.64	0.67	0.05	0.17	0.06	0.18	0.07	0.26	3.80	5.89
18	1.11	1.18	0.05	0.05	0.06	0.65	0.81	0.26	1.73	5.90
20	1.24	1.19	0.05	0.28	0.06	0.66	0.52	0.26	1.73	5.99
19	1.02	2.52	0.05	0.05	0.06	0.31	0.07	0.26	1.73	6.06
21	1.03	2.09	0.05	0.30	0.20	0.40	0.33	0.26	1.73	6.41
22	0.96	0.75	0.19	0.27	0.06	0.66	1.83	0.26	1.73	6.71
31	1.05	0.34	0.05	0.05	0.06	0.65	0.07	0.26	4.33	6.86
23	1.33	2.37	0.10	0.05	0.06	0.62	0.34	0.26	1.73	6.87
32	1.07	0.81	0.05	0.05	0.06	0.36	0.07	0.26	4.27	6.98
25	1.15	0.58	0.05	0.05	0.06	1.13	0.07	2.19	1.73	7.00
27	0.77	0.63	0.05	0.28	0.06	1.00	0.47	2.02	1.73	7.02
24	2.05	1.37	0.05	0.42	0.06	1.05	0.07	0.26	1.73	7.05
26	2.26	2.48	0.05	0.05	0.06	0.44	0.07	0.26	1.73	7.39
30	1.65	0.54	0.05	0.31	0.06	1.38	0.07	1.85	1.73	7.64
36	1.21	0.73	0.05	0.23	0.06	0.49	0.07	0.26	5.60	8.70
34	0.93	1.44	0.10	0.05	0.06	1.19	0.07	3.21	1.73	8.78
33	2.73	3.06	0.05	0.05	0.06	0.87	0.07	0.26	1.73	8.88
39	0.79	1.38	0.05	0.12	0.12	1.17	0.17	1.46	3.91	9.16
38	1.25	0.27	0.05	0.05	0.06	0.58	0.27	1.07	5.64	9.23
40	1.27	0.44	0.05	0.05	0.06	0.56	0.07	0.26	6.86	9.60
35	1.05	1.51	0.05	0.05	0.06	1.34	0.52	3.83	1.73	10.1
41	0.80	1.64	0.05	0.05	0.06	0.52	0.07	0.26	7.04	10.5
43	1.68	0.77	0.05	0.28	0.06	0.69	0.42	1.01	5.65	10.6
42	1.27	0.50	0.05	0.05	0.06	0.47	0.37	0.91	6.99	10.7
37	2.77	2.54	0.05	0.62	0.06	0.91	0.07	1.95	1.73	10.7
44	1.28	0.29	0.05	0.05	0.06	0.74	0.07	0.26	8.21	11.0
45	1.79	0.26	0.05	0.05	0.06	0.50	0.07	2.13	6.47	11.4
46	0.94	0.73	0.05	0.05	0.06	0.52	0.07	0.85	8.21	11.5
47	0.95	1.03	0.14	0.16	0.06	0.84	1.63	2.11	4.62	11.5
48	1.85	1.43	0.05	0.25	0.17	1.12	0.07	0.26	6.69	11.9
49	1.21	1.27	0.20	0.34	0.06	0.63	0.47	1.28	6.74	12.2
50	0.64	1.09	0.05	0.05	0.06	0.08	0.07	0.26	10.77	13.1
51	1.27	0.83	0.05	0.05	0.06	0.28	0.07	0.26	10.90	13.8
52	1.03	1.16	0.05	0.05	0.06	0.62	0.31	1.58	8.95	13.8
53	2.68	1.34	0.05	0.33	0.59	0.54	0.07	0.26	11.70	17.6
55	1.46	1.85	0.05	0.05	0.06	0.63	0.07	3.01	10.87	18.1
54	2.26	1.11	0.05	0.05	0.06	0.91	0.07	0.26	13.30	18.1
56	1.55	1.52	0.05	0.05	0.06	0.89	0.66	3.10	10.91	18.8
57	4.18	6.18	0.05	0.05	0.06	1.85	1.02	4.57	7.66	25.6
58	4.49	2.50	0.50	1.37	9.74	4.74	2.50	6.37	1.73	33.9
Median concentration	1.06	1.01	0.05	0.05	0.06	0.61	0.07	0.26	1.73	7.22
Range	0.27–4.49	0.10–6.18	0.05–0.50	0.05–1.37	0.06–9.74	0.08–4.74	0.07–2.50	0.26–6.37	1.73–13.3	2.66–33.9
LOD	0.54	0.19	0.10	0.10	0.11	0.17	0.14	0.52	3.47	

* Concentrations below the LOD were taken as 1/2 LOD.

Table 2 PBDEs concentrations in blood samples of children in the literature compared with our study (unit: ng/g lw)

Country	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-197	BDE-209	∑PBDEs	Age	Period	Reference
Norway	0.20	2.00	0.37	0.66	0.86	0.39				4.48	4–14	1975–1999	Thomsen et al., 2002
Sweden		0.87	0.47	0.14	2.50				1.00	5.00	7	2002–2006	Fängström et al., 2005
USA	1.00 (0.30–2.20)	31.0 (11.0–65.0)	6.20 (1.80–15.0)	6.20 (2.10–14.0)	13.0 (3.40–32.0)		0.00 (0.00–1.70)	0.49 (0.13–2.00)	1.70 (0.90–19.0)	59.6	1–4	2006–2007	Lunder et al., 2010
USA	1.60	42.2	9.00	9.60	13.6	1.00				79.3	6–8	2005–2009	Windham et al., 2010
USA		94.0–137	28.0–34.0	39.0–57.0	64.0–75.0	3.00–7.00			9.00–143	239–418	5	2004	Fischer et al., 2006
Mexico		7.12 (ND–31.7)	6.00 (ND–26.0)	3.79 (ND–11.8)	4.46 (ND–9.10)	8.08 (ND–3.70)			< ND	29.5	6–13	2006–2009	Pérez-Maldonado et al., 2009
China (Dalian)	ND (ND–7.13)	6.00 (ND–19.4)	5.27 (ND–18.1)	2.35 (ND–11.9)	8.25 (ND–27.1)	ND (ND–14.8)	ND (ND–6.88)			31.6 (ND–188)	0–11	2006	Chen et al., 2009
China (Zhejiang)	0.22–1.10	0.66–5.51	0.344–17.1	0.15–4.42	< 0.01–15.1	< 0.01–5.37	0.32–2.88			4.54–65.2	5–11	2008	Shen et al., 2010
China (Taizhou)	1.06 (0.27–4.49)	1.01 (0.10–6.18)	0.05 (0.05–1.37)	0.05 (0.05–0.50)	0.61 (0.08–4.74)	0.06 (0.06–9.74)	0.07 (0.07–2.5)	0.26 (0.26–2.50)	1.73 (1.73–6.37)	7.22 (2.66–32.2)	9–12	2009	This study

PBDEs levels in Taizhou children in our study was similar to the PBDEs levels in general adult population in China reported in previous studies (Bi et al., 2006; Chao et al., 2007; Zhu et al., 2009) and comparable to the whole level of PBDEs in Asia population (Wang et al., 2007).

2.2 PBDE congener profile

BDE-28, -47 and -153 were found in almost all the samples, BDE-209 was found in about 56% samples, BDE-99, -183 and -197 were found in about 35% samples, while BDE-100 and BDE-154 were found in only 10% samples. BDE-209 was the most abundant PBDE congener in found samples, accounting for 46% to ∑PBDEs, followed by BDE-28, -47, -197 and -153, which were also the most frequently reported congeners in previous studies (Fig. 1). Figure 2 shows the concentrations of PBDE congeners as well as the contribution percentages of PBDE congeners in blood samples from Taizhou children.

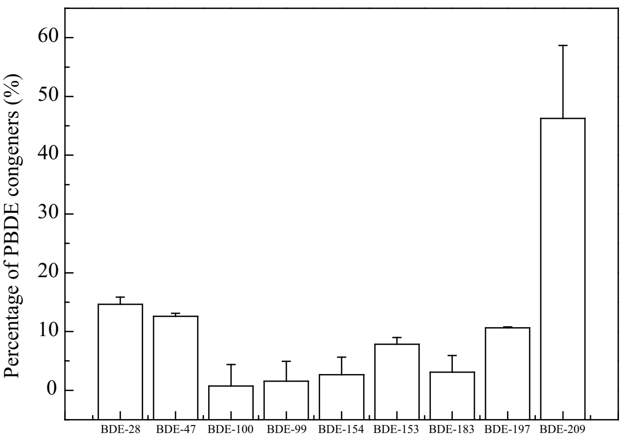

Fig. 1 Average percentages of PBDE congeners in blood samples from Taizhou children.

Although BDE-209 was found in only 56% samples, it was the most abundant congener in blood samples. In a previous study, BDE-209 was found dominant congener in human blood samples from an e-waste recycling site in Taizhou (Zhao et al., 2010). Moreover, some studies reported that BDE-209 was also the dominant congener in environmental and biotic samples in Taizhou (Liang et al., 2008; Qin et al., 2009; Han et al., 2009). These data demonstrated that BDE-209 can easily accumulate in human body and should not be neglected when investigating human exposure to PBDEs. Considering that BDE-209 is the dominant congener in Chinese population even Asian populations in previous reports, no detected BDE-209 in some samples was inferred possibly due to a high LOD of BDE-209 but not to the absence of BDE-209 in these samples. Therefore, we regarded the concentrations as 1/2 of LOD when the concentration of BDE-209 was below the LOD. Regrettably, some authors seemed to not note BDE-209 or cannot obtain good results due to a bad QA/QC (Shen et al., 2010; Chen et al., 2010).

The high level of BDE-197 in this study was probably derived from the debromination of BDE-209. Deca-BDE may break down into lower brominated diphenyl ethers. BDE-197 and other octa- and nona-brominated congeners have been reported at elevated levels in the serum of Chinese electronics dismantlers (Bi et al., 2007). BDE-153 was also observed as one of the dominant congener in human blood samples from other studies (Choi et al., 2003; Kumsue et al., 2007; Fernandez et al., 2007; Covaci et al., 2008). The high percentage of BDE-153 observed in the investigated blood samples might be a consequence of its long half-life. With a longer half-life than other congeners,

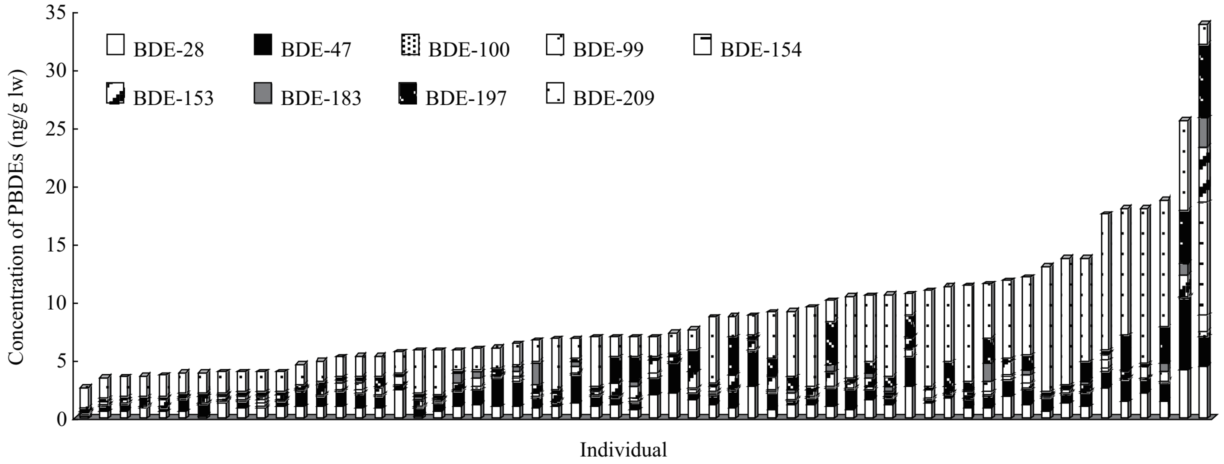

Fig. 2 Concentrations of PBDE congeners in blood samples from Taizhou children.

BDE-153 was more persistent and bioaccumulative in human bodies than other congeners (Chen et al., 2010). Fischer et al. (2006) have observed a decrease of BDE-209 accompanied with an increase of BDE-153, which was possibly another explanation for the high abundance of BDE-153.

BDE-47 and BDE-99, which are the most abundant congeners in American and European populations, contributed less to ∑PBDEs relative to high brominated congeners in Taizhou children. The finding could be explained by the pollution background of PBDEs in China, which is characterized by high brominated congeners as main pollutants (Wang et al., 2009; Chen et al., 2006; Duan et al., 2010).

3 Conclusions

In this study, we investigated the levels and congener profiles of PBDEs in blood samples from children in Taizhou countryside. The PBDEs levels in these children were lower than those in American children, and comparable to the values of European children and the general level of PBDEs in Asia population. The predominant congener in found samples was BDE-209, followed by BDE-28, -47, -197 and -153. The PBDE congener profile in Taizhou children was consistent with the pollution background characterized by high brominated congeners as main pollutants.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 20677074, 20437020) and the State Key laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-environmental Sciences, Chinese Academy of Sciences (No. KF2007-07, KF2008-11). We express our heartfelt gratitude to all blood donators. We also thank the doctors in Taizhou Hospital, Zhejiang Province, for their help in colleting all samples.

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