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Pollution Status and Human Exposure of Decabromodiphenyl Ether (BDE-209) in China

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Ⓢ Supporting Information

ABSTRACT: Decabromodiphenyl ether (BDE-209/decaBDE) is a high-production-volume brominated flame retardant in China, where the decaBDE commercial mixture is manufactured in Laizhou Bay, Shandong Province, even after the prohibition of penta- and octaBDE mixtures. The demand for flame retardants produced in China has been increasing in recent years as China not only produces electronic devices but also has numerous electronic waste (e-waste) recycling regions, which receive e-wastes from both domestic and foreign sources. High concentrations of BDE-209 have been observed in biotic and abiotic media in each of the different areas, especially within the decaBDE manufacturers and e-waste recycling areas. BDE-209 has been viewed as toxic and bioaccumulative because it might debrominate to less brominated polybrominated diphenyl ethers (PBDEs) (lower molecular weight and hydrophobicity), which are more readily absorbed by organisms. The highest concentration of PBDEs in dust within urban areas reached 40 236 ng g^{-1} in the Pearl River Delta, and BDE-209 contributed the greatest proportion to the total PBDEs (95.1%). Moreover, the maximum hazard quotient was found for toddlers (0.703) for BDE-209, which was close to 1. This suggests that exposure to BDE-209 might lead to increased potential for adverse effects and organ harm (e.g., the lungs) through inhalation, dust ingestion, and dermal absorption, especially for the group of toddlers compared to others. In daily food and human tissues, the amount of BDE-209 was also extensively detected. However, the toxicity and adverse effect of BDE-209 to humans are still not clear; thus, further studies are required to better assess the toxicological effects and exposure scenarios, a more enhanced environmental policy for ecological risks regarding BDE-209 and its debrominated byproducts in China.

1. INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are used as brominated flame retardants (BFRs) in textile, plastic, and electronic industries and are organobromine compounds consisting of up to 10 bromine atoms. However, many PBDEs have multiple congeners, and typically, commercial PBDE products are a mixture of congeners rather than a single compound. Three of these mixtures, pentaBDE, octaBDE, and decaBDE, were 14, 6, and 80%, respectively, of the estimated 67 000 t PBDE produced annually worldwide.[1] The major constituents of pentaBDE are BDE-47, BDE-99, and BDE-100, with lesser contributions from BDE-153, BDE-154, and BDE-85. The major congener found in octaBDE is BDE-183 with minor contributions from BDE-203 and several octa- and nonaBDEs.[2] DecaBDE consists predominantly of the congener BDE-209 (≥97%), with low levels of other brominated diphenyl ether congeners such as nonaBDE (0.3–3%) and octaBDE (0–0.04%).[3] The physicochemical properties of BDE-209 are summarized in Table S1. The global demand for flame retardants has been rising over the past decade, especially considering the growth in consumption of electronics and

electrical products, which are continually being replaced with new and more innovative products. BDE-209 is widespread and one of the most prevalent PBDEs in the global environment.[3] Furthermore, it is estimated that Asia accounted for approximately half of the consumption of flame retardants in the world, and China is considered to have the highest demand for flame retardants in Asia.[4]

As an additive flame retardant, decaBDE is not chemically bound to the product or the material in which it is used. Therefore, it has the potential to "leak" into the surrounding environment, although low water solubility (<20−30 μg L^{-1}) and vapor pressure (<10^{-6} mmHg at 20 °C) of BDE-209 retard its release and limit mobility.[5] The distribution of BDE-209 in environmental matrices across China is shown in Figure 1. In spite of long environmental half-life and persistence of BDE-209 in air, soil, and sediments, there exists substantial evidence that BDE-209 can debrominate to lesser brominated PBDEs
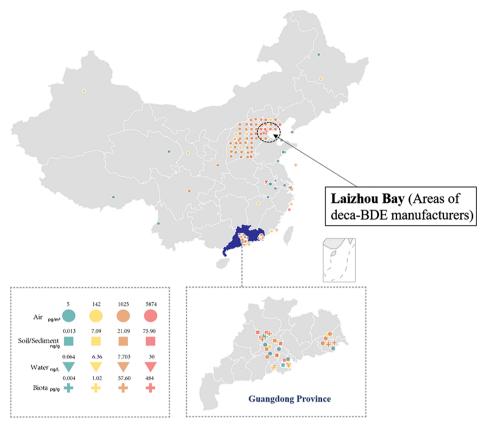
Figure 1. Distribution of BDE-209 pollution status in air, dust, soil/sediment, water, and biota (terrestrial, aquatic, and human) in China.[19,20,25–27,39,40,42–44,47,49,55,56,63,66,67,69,70,72,74,75,78–81,84,87,89,90,92–96,98–101,105,107,108,110–113,115,118,120,128,141–146]

that range from mono- to nonaBDEs in abiotic environment and biota. Mono- to nonaBDEs include listed persistent organic pollutants (POPs) such as tetra- to heptaBDEs and bromophenols,[3] which are more readily taken up by organisms because of their lower molecular weight. Additionally, some environmental conditions favor debromination, likely forming congeners with greater toxicity and bioaccumulation.[5] For example, when BDE-209 bounds with artificial and natural soil, sediment, and sand, photolytic debromination of BDE-209 can occur.[6–8] Ahn et al. observed that BDE-209 absorbed more readily to matrices consisting of clay minerals, metal oxides, and sediments that can debrominate to tri- to nona-PBDEs under exposure of 14-day sunlight irradiation.[8] Some studies have revealed the biotransformation of BDE-209. Rainbow trout (*Oncorhynchus mykiss*) and common carp (*Cyprinus carpio*) were fed cod spiked with BDE-209; BDE-209 was not detected in the fish, but hexa- to nonaBDE congeners were.[9] Metabolic debromination of BDE-209 occurs in the roots and shoots of aquatic plants from a wastewater receiving stream.[5] Deng et al. observed that a significant proportion of pentaBDE and octaBDE was debrominated from decaBDE in the roots of aquatic plants through simulated pot experiments.[10] The same phenomenon was observed in the soil–plant system under a greenhouse pot experiment.[11] DecaBDE was also proven to be labile, forming debromination congeners photolytically in natural matrices by using artificial sunlight.[7] These reports demonstrate that penta- and octaBDE might have a greater potential to bioaccumulate and cause toxicity.

Because of widespread persistence and toxicity of PBDEs in the environment, as well as the potential hazard to humans, the manufacturing and utilization of PBDEs have been strictly regulated and have been partially prohibited for the last 12 years. In addition, the POPs Review Committee examined a proposal by Norway to list decabromodiphenyl ether under the Stockholm Convention on POPs and concluded that it met the screening criteria specified in Annex D.[3] Currently, BDE-209 is in the process of being added to Annex A. The phasing out of PBDE production has been primarily voluntary, whereas the phasing out of decaBDE was expected to be completed by 2012 in the United States.[12] The European Union decided to ban the use of mixtures of penta-, octa- (2004), and decaBDE (2008) in electronic devices.[13] In 2009, octaBDE and pentaBDE were added to the list of POPs, and at a later stage, the decaBDE formulation.[14] In China, commercial mixtures of pentaBDE were banned from production in 2004, and octaBDE was never produced.[4] In 2008, Albemarle Corporation (US) and Dead Sea Bromine Company (Israel) started to produce decaBDE in China. Chemtura Corporation (US) also produced decaBDE in China later. In 2016, several corporations in China produced decaBDE, including Everkem, ICL-IP Europe, Tosoh Corporation, Shouguang Weidong Chemical, Jinan Shangshan Chemical, Shouguang Jinyingtai Chemical, Yuancheng Technology, Shouguang Tianyi Chemical, Shandong Haiwang Chemical, and Jiangyin Suli Chemical.[15] However, production volumes currently remain unknown. Moreover, the demand for BFRs has reflected an increased trend, exhibiting rapid increases in production (8% annually)[16] while decaBDE accounted for approximately 45% of total BFRs produced in 2006 with 36 000 t.[17] The domestic manufacturing of decaBDE soared to approximately 41 500 t in 2005, which was substantially greater than the 26 000 t produced in 2000 but decreased to 20 500 t in 2011.[4] However, decaBDE continues to be in use without any
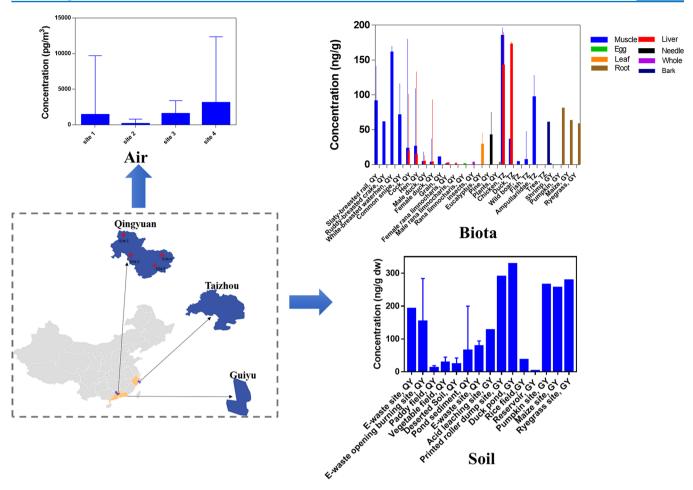
Figure 2. Concentrations of BDE-209 in air, soil, and biota in e-waste recycling/dismantling areas of QY, GY, and TZ.[25−27,39,40,42−44,47,56,63,66,67,147,148]

restrictive policy until now in China. The domestic decaBDE manufacturing facilities of China are primarily situated in Laizhou Bay, Shandong Province.[18] China is the second largest consumer in the world regarding the electronics market, and it was assumed that it might even replace America as the number one consumer in 2016. In 2005, consumption of the decaBDE commercial mixture in China amounted to approximately 30 000 t.[19] This figure was estimated to be 70 000 t by 2016, based on the increasing rate of approximately 8% annually in domestic demand for BFRs, which were primarily taken over by decaBDE.[20] Moreover, there are a considerable number of electronic waste (e-waste) recycling regions, especially in southern and southeastern China, where Guiyu (GY), Qingyuan (QY), and Taizhou (TZ) have become renowned worldwide as industrial e-waste dismantling regions.[21−23] In fact, many covert family-run workshops in those areas engage in disassembling electronic components by primitive methods such as acid washing and open burning, which has been banned by the Chinese government, thereby causing a substantial amount of PBDE emissions from heating printed wiring boards.[24] BDE-209 is a high-production-volume flame retardant added to appliances in China. Therefore, numerous investigations throughout various different environmental matrices in e-waste recycling areas in China have shown that BDE-209 was the dominant PBDE congener and major BFR pollutant in recent years.[25−28]

At present, China is the largest producer and supplier of decaBDE mixture.[29] BDE-209 continues to be the dominant congener in an ambient medium and still continues to be manufactured. The half-life of BDE-209 in human blood from Swedish works engaged in recycling process was estimated to be in the range of about a week, whereas the corresponding value for BDE-183 was estimated to be of 3 weeks,[30,31] which indicates that decaBDE is likely to undergo rapid metabolism. Therefore, the excessive number of electronic devices in China might pose the primary potential risk to the extensive number of people who work with electronic devices nowadays. Human exposure to BDE-209 has become a concern in the past decades, and there are several studies regarding human exposure to BDE-209 in China, where BDE-209 has been detected in breast milk, hair, and blood serum.[32−37] Taking into account the above-mentioned decaBDE, particularly in China, this article reviews the pollution status and distribution of decaBDE as well as the debromination and other metabolites in the biological and abiotic environment of China; the potential risk of human exposure to decaBDE is also discussed. On the basis of the abundance of data available, the most recent reports have been utilized to depict the impact of decaBDE contamination on the environment centered on its production or use.

2. REVIEW STRATEGY

We systematically utilized the electronic databases, for example, Web of Knowledge, ScienceDirect, ACS Publications, Google Scholar, and so forth from 2005 up until February 2017, using the following search terms: PBDEs, BDE-209, decaBDE, and decabromodiphenyl oxide, China. Moreover, broad search terms, such as fire retardant and BFRs, were also applied to ensure that there was no loss of any publications. Scientific journals, official reports, conference proceedings, and news reports that were discovered on the Internet were obtained. Our search was not confined to the English language but included Chinese as well. The references including the occurrence of BDE-209 in different types of areas in China, information about decaBDE/BFR production in China, human exposure of BDE-209, and degradation of BDE-209 were selected. We classified the areas where different pollution levels of BDE-209 in the biotic and abiotic environment of China were discovered, according to 126 identified references.

3. BDE-209 POLLUTION STATUS IN DIFFERENT REGIONS OF CHINA

3.1. e-Waste Recycling/Dismantling Areas. Environmental decaBDE is associated with the disposal of BDE-209-treated electrical and electronic equipment that are composed of products that have reached their end-of-life cycle. In China, it was estimated that the generation of e-waste could reach up to 3.66 million metric tons in 2010.[4] The decaBDE content within e-waste in China was estimated to be 1867 t.[38] Of that amount of e-waste flow, 12% accounted for deserted consumer electronics recovered for recycling, and only this small fraction of e-waste flow will be sorted and treated for reuse or recovery of base materials.[38] From the reports, so far, Guangdong Province appears to be the most contaminated area by e-waste, including the three regions of Guangzhou (GZ), GY, and QY. TZ in Zhejiang Province, where e-waste recycling activities have been prominent since the late 1970s, has also been reported on extensively. The levels of BDE-209 in the air, soil, and biota within these three areas are shown in Figure 2.

3.1.1. Qingyuan. QY county is located in the Guangdong Province, 50 km north of GZ city, where more than 1300 dismantling and recycling workshops exist.[26] About 700 000 t of e-waste are disposed off annually, within an area of 330 km^2, where it is processed primarily by primitive methods (open burning, acid processing, and mechanical shredding).[39] Moreover, abundances of total BDE-209 and nonaBDE among PBDEs were discovered in muscle samples of free-range chickens (78−82%) and ducks (70−81%) purchased from local farmlands in QY. Additionally, when comparing the level of those fed with grain and the soil where PBDE exposure to poultry could occur, the biodebromination of BDE-209 might occur in chickens and ducks.[26] However, the grain feed might not be a significant contributing factor to BDE-209 or other PBDE congeners because of the apparent difference in the concentrations of BDE-209 between chickens (2.7 ng g^{-1} lipid weight average) and ducks (0.35 ng g^{-1} lipid weight average), which have diverse living and feeding habits. Thus, it would appear that more polluted land-associated characteristics such as dust and insects might be attributable to the higher burden of BDE-209 in chickens.[26] Luo et al. investigated the relationship between the concentrations of BDE and five waterbird species near the e-waste recycling workshops in QY, which revealed that the Chinese pond heron (*Ardeola bacchus*)

and ruddy-breasted crake (*Porzana fusca*) were enriched with BDE-47, BDE-99, and BDE-100, whereas the white-breasted waterhen (*Amaurornis phoenicurus*) and common snipe (*Gallinago gallinago*) accumulated BDE-153, BDE-183, and BDE-154 and slaty-breasted rail (*Gallirallus striatus*) showed a significantly different congener profile that pronounced the accumulation of BDE-209, followed by BDE-153, and a higher nonaBDE than other species.[40] The considerable level of nonaBDE in these species could be due to the preferential uptake of nonaBDEs compared to that of decaBDEs because of the differences in bioavailability and partitioning or also because of the biotransformation from BDE-209. This was also discovered in the European starling (*Sturnus vulgaris*).[41] A higher concentration of BDE-153 might also be closely linked to different metabolic behaviors among different species, from BDE-209 or other high brominated PBDEs. Evidence suggests that direct exposure to BDE-209 or its metabolites in the food chain, such as the consumption of insects for birds and poultry, facilitates contact with BDE-209-contaminated soil, especially at e-waste dumping and recycling sites, and is also exposed to humans via diet. BDE-209 also showed a higher accumulation in the liver of frogs (*Rana limnocharis*) sampled from e-waste-contaminated areas in QY than tri- to heptaBDE congeners.[42] The high fat content of the liver might explain the deposition of BDE-209 because of its large molecular size and low hydrophobicity, restricting it from crossing membranes.[42] Moreover, maternal transfer of PBDEs has also been discovered in frogs whereas decaBDE transferred less than tri- to heptaBDE, which might be due to its relatively large molecular size.[42]

From the air samples collected from QY by Tian et al., it was discovered that BDE-209 was the most dominant congener in almost all air samples, accounting for 47 ± 17 and 59 ± 16% of the total PBDE congeners in e-waste and rural areas, respectively.[43] Using equilibrium gas/particle partitioning theory, highly brominated BFRs, such as BDE-209, are mainly formed as particle-bound BFRs that could originate from the burning and shredding of e-wastes, resuspending oil and dust, and the formation of secondary aerosols in the air.[43] However, e-waste recycling in QY (low-latitude areas) is a significant source of less brominated PBDEs because of the emission being strongly temperature-dependent; thus, weak temperature dependence at the rural site implies that they are likely to undergo global transport.[43] In comparison to the relationship between concentrations of PBDEs in plant leaves and those in the gaseous phase, particle phase, and leaf surface particle, PBDEs in plants showed no clear correlation with particle-bound PBDEs in the atmosphere; however, there is an exception where BDE-209 in eucalyptus leaves exhibited a significant negative relationship ($r = -0.67$, $p < 0.05$).[44] Highly brominated PBDEs (nona- and decaBDE) reflected a lesser relationship within the same plant species, as compared to less brominated BDEs, which might result from the high uptake rates of highly brominated BDEs from leaf surface particles into leaves, influencing the concentration of BDE-209 within leaf surface particles.[44] The uptake mechanism is also connected to the relationship between the log K_{PA} (log plant/air partition coefficient) and log K_{OA} (log octanol/air partition coefficient) for BDE-209.[44] This has also been proven by increasing cuticular permeability with octanol/air partition coefficient (K_{OA}) of semivolatile organic compounds.[45] BDE-209 contributions in the plants of QY (60 and 50% average of the total PBDE burden in eucalyptus leaves and pine needles,

respectively) were comparable to those in spruce needles in Ottawa, Canada (66%), and tree bark in North America (60%).[46]

Wang et al. investigated the PBDEs in soils and plants in QY (Longyang and Shijiao, two administrative towns) and determined that BDE-209 was the predominant congener in all soil samples, accounting for 53.2−37.7% of the total PBDE contents, whereas only relatively low levels of BDE-209 were detected in woody plant roots [ranging from 0.3 ± 0.03 to 5.3 ± 0.6 ng g^{-1} dry weight (dw)], accounting for 19.7% of the total PBDEs, while lower brominated PBDEs (mono- through pentaBDE) account for 90.2% in the roots and 97.7% in the leaves on average.[47] This could result from preferential root uptake and translocation for lower brominated PBDEs and also metabolites from the debromination of higher brominated PBDEs in plants.[47] Moreover, the concentration of BDE-209 in the soil ranged from 28.8 ng g^{-1} (paddy field, 10−20 cm) to 468 ng g^{-1} (e-waste site, 0−10 cm) in QY, suggesting that the soil in QY has been polluted by BDE-209, via e-waste recycling activities.[48] Moreover, the concentration of BDE-209 was about 4-fold higher in the surface soil from a peanut field than that in the paddy field, which can be extrapolated by a different tillage or irrigation frequency that might contribute to the different concentrations and account for the transfer of PBDEs in the paddy and peanut fields in spite of the similar proximity (<1 km) to the e-waste.[48] The paddy field is more irrigated than the peanut field because rice is a semiaquatic plant requiring more watering during growth, which could cause increasing inputs of contaminant via irrigation water. However, this paper showed lower concentration of BDE-209 in the peanut field than that in the paddy field, indicating that the fate of BDE-209 in the humid soil of the paddy field might be different from that in the dry soil. Because of very low volatility of BDE-209, low concentrations of BDE-209 in the topsoil of paddy field might be mainly due to adsorption of particles linked with runoff and leaching. In the sediment collected from a pond in QY, BDE-209 was also the predominant congener with 65% of the total PBDEs.[28] Wang et al. detected soil and plant samples from intensive uncontrolled e-waste recycling sites near QY and agricultural fields in close proximity to it.[49] In this study, among the total PBDEs, BDE-209 accounted for 36−91% as the dominant congener in soil; this result was also consistent with the fact that the commercial decaBDE mixture is frequently added to plastic products used in high-tech and other electrical appliances in China and accounts for the key contaminant in this region as previously reported.[49,50] This pattern was also detected in vegetables, reflecting that BDE-209 was still the most predominant congener, followed by BDE-47, in all plant samples, and the highest concentration of BDE-209 was found in *Brassica alboglabra* L.[49] The tissue-to-soil ratio of vegetables ($C_{vegetable}/C_{soil}$) in these sites showed that the ratio of PBDEs without BDE-209 was much higher than the ratio of BDE-209, which means that lower brominated PBDEs were easier to be removed than the higher brominated PBDEs, and the ratios of roots or tubers were lower than the ratios of shoots in various vegetables. This suggests that PBDEs (including BDE-209) in shoots might generally be derived from the atmosphere rather than the soil. Another interesting finding is that *B. alboglabra* L. had the highest tissue-to-soil ratio of BDE-209, whereas *Lactuca sativa* L. had the highest tissue-to-soil ratio of other congeners without BDE-209. This indicates that the accumulation potential differs between the two species, which might pose diverse potential risks to the vegetables planted in this ambient area.[49]

3.1.2. Guiyu. GY is situated in Chaoyang district, Shantou city, GZ Province, Southeast China, with a total area of 52 km^2 and a population of 150 000. This town has been involved in e-waste dismantling/recycling (processes about 70% of the exported e-waste of world) for approximately 20 years.[22,25,51] According to reports on e-waste treatment in GY, approximately 30 000 to 40 000 people are involved in e-waste treatment, treating over 1 million tons of e-waste annually.[52] In the e-waste recycling workshops of GY, the total concentration of suspended particulates (TSP) in the air depicted a sustained growth throughout the working period, suggesting a continuous TSP emission from all workshops.[51] BDE-209 was the dominant congener at all sampling sites relevant to TSP in off-site and on-site e-waste dismantling workshops, which is likely to be particle-associated BDE-209 that can adsorb fine particles more readily when compared to other low brominated congeners. Despite the fact that BDE-209 is not influenced by high temperature as much as low brominated congeners during the process of incineration because of its high melting point (290−306 °C), these particles can easily volatilize.[51,53,54] Although BDE-209 is associated with large particles that might not be inhaled into human lungs, on-site e-waste dismantling workshops continue to be the area that reflects the highest exposure of all workshop employees according to the exposure assessment.[51] Deng et al. also investigated TSP and particles with aerodynamic diameters less than 2.5 µm (PM 2.5) at an e-waste recycling site in GY and measured concentrations of 124 and 62.1 µg m^{-3}, respectively.[22] The same result was reflected with higher concentrations of PBDE observed in the air because of the heating and open burning of e-waste when plastics containing PBDEs were heated, whereas BDE-209 was not investigated.[22] During both daytime and nighttime, the air samples were collected from an e-waste recycling site in GY and BDE-209 was found to be the predominant congener of all PBDEs (only in particle phase, associated chemicals were detected), whereas the concentration of BDE-209 did not show as high a concentration as in the Swedish recycling plant in which commercial decaBDE was the most abundant.[55] Moreover, when compared to the commercial decaBDE mixture, BDE-209 exhibited a lower concentration (9.4−23% in summer and 29.1−41.0% in winter).[55]

The surface soil exposed during e-waste recycling for an extended period of time and the residue after combustion of e-waste from GY were analyzed for PBDEs. The most dominant congener was BDE-209 that accounted for 35−82%, indicating that the commercial decaBDE was prevalent.[25] The concentration of BDE-209 was statistically higher in soil samples at the acid leaching, riverbank, and the printer roller dump site and was greater than that discovered at the rice field, reservoir, and duck pond.[25] The concentration of BDE-209 was approximately 3-fold to 5-fold higher than the total concentration from mono- to nonaBDE in the duck pond, rice field, riverbank, and reservoir, which suggested that the commercial decaBDE mixture is highly prevalent in GY and thus represents tremendous inputs into the environment.[25] However, it is uncertain whether decaBDE debrominated to low bromine BDE (i.e., BDE-47 and BDE-99). An analysis of Pearson's correlations revealed no significant correlations between BDE-209 and BDE-47 (probability levels of 0.05) and between BDE-209 and BDE-99 (probability levels of 0.01) at the printer roller dump site, rice field, duck pond, and the reservoir, suggesting
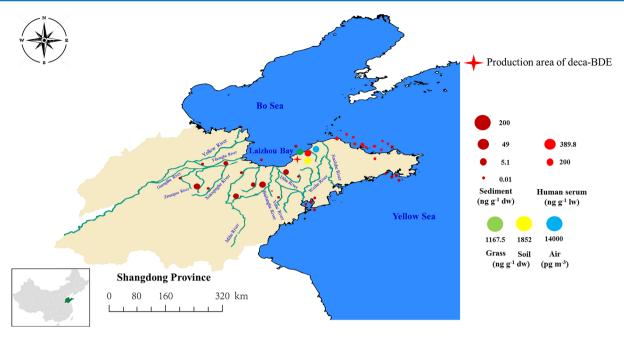
Figure 3. Concentration of BDE-209 in biota and abiotic media from the areas of decaBDE manufacturers in China.[70,72,74,75,77−80,108] The corporations in the production area (Bohai Chemical Industry Park, Yangkou town, Shouguang, Shandong Province; N 37°07′31.3″, E 119°51′2.88″), which manufacture commercial decaBDE mixture, include Shandong Brother Sci. & Tech. Co. Ltd (http://www.oceanchemical.com/), Shandong Tianyi Chemicals Co. Ltd (http://www.tychemical.com.cn/ in Chinese), and Shandong Futong Chemical Co. Ltd (http://en.futongchem.cn/).

that BDE-47 and BDE-99 did not come from the debromination of BDE-209. Huang et al. studied the dissipation of PBDEs and plant uptake in soil from e-waste recycling sites in GY, where higher proportions of low brominated PBDEs such as BDE-47, BDE-66, and BDE-99 in plant roots were found much more than those in the soil, whereas BDE-209 was found to have lower proportions in plant roots than those in the soil.[56] This illustrates that higher brominated PBDEs such as BDE-209 are not easily absorbed by plants from the soil. In comparison to lower and higher brominated PBDEs, there are higher eradication rates (16.1−34.8%) in lower brominated PBDEs (9.4−17.7%), irrespective of plant or soil species.[56] The contribution for dissipation of BDE-209 might be due to plant uptake, and losses might be caused by volatilization and degradation by the microorganisms in the soil. The low dissipation for BDE-209 is ascribed to higher partition coefficients, and BDE-209 is strongly bound to the soil (particularly soil organic matter), thereby becoming less degradable by the microorganisms in the soil. Moreover, the hydroxylated PBDEs (OH-PBDEs) were found in aerosol samples from a printed circuit board's recycling workshop in GY when waste printed circuit boards were heated with temperatures ranging from 250 to 300 °C, in which the most abundant congener was 6-OH-BDE-47 (0.32 ng m^{-3}).[57] However, if BDE-209 could decompose to low brominated PBDEs at high temperature, then OH-PBDEs (i.e., OH-BDE-47 and OH-BDE-99) would be formed by the oxidation reaction of PBDEs, which appears to be even more toxic than PBDEs themselves and reportedly disrupts thyroid hormone homeostasis, neurotoxicity, sex hormone steroidogenesis, and oxidative phosphorylation.[58−60]

3.1.3. Taizhou. TZ is a prefecture-level city in Zhejiang Province, Southeast China. e-Waste recycling activities have been conducted in several towns of TZ, where e-waste has been illegally dismantled and recycled, along with an annual

production exceeding 2.2 million tons, via primitive methods (i.e., open burning and acid picking) carried out in small-sized family workshops since the late 1970s.[27,61,62] Collected tree samples from the town of Luqiao, TZ, revealed that BDE-209 was the most abundant homologue, accounting for 56.9−87.2%, and the analytical results calculated by a least-square procedure showed that the commercial decaBDE mixtures were the main source of PBDEs in tree bark. The results from the same area in other environmental matrices also showed the same pattern, which was revealed from sediment and soil samples.[63,64] PBDEs can be released from plastics, which also contain BFRs in heating, because they are not chemically bound to products. Thus, open burning of e-waste might be a significant source of BDE-209, through which BDE-209 can be emitted into the ambient environment in this area. In another town (Fengjiang) of TZ, BDE-209 was detected in 27 snail samples with the highest concentration of 45 040 ng g^{-1} dw.[27] The same patterns were reflected in other reports on biological samples in e-waste recycling areas.[63,65,66] BDE-209 was distributed widely in snails, which indicates that high brominated congeners could be transported to the ambient environment and can be bioavailable in TZ as well. Fu et al. also speculated that the usage of decaBDE in electrical equipment was the primary source for BDE-209 in this region.[27] Moreover, Shang et al. found that major PBDE congeners could be utilized by earthworms from TZ, even if the molecular size of some congeners such as BDE-209 is larger than 9.5 Å, which was considered to be the maximum effective molecular cross section of organic chemicals to penetrate biological membranes.[61] These studies have all suggested the bioavailability of BDE-209 in terrestrial animals in e-waste recycling areas of TZ, which might cause potential harm to the ecosystem of this region.

Furthermore, 10 kinds of locally produced staple foods, for example, meat, fish, and eggs, were analyzed and estimated with regard to dietary intakes of PBDEs for inhabitants living in the

areas of TZ. The study found that BDE-209 was the most abundant congener in chicken, including samples of chicken livers and eggs, and BDE-47 was the most significant contributor in fish.[67] The median estimate of exposure of BDE-209 in children and adults was not beyond the reference dose (RfD) value calculated by the U.S. EPA (7000 ng kg^{-1} day^{-1}).[67,68] However, the principal component analysis (PCA) revealed that food samples collected from TZ, which is near e-waste recycling areas, are severely affected by pollution from the activities of e-waste recycling and disposal.[67] However, the metabolic process of BDE-209 is still not clear, but BDE-209 exposure to humans and other organisms still has a potential adverse effect on ecosystems in TZ.

3.2. Areas of DecaBDE Manufacturers. Most of the commercial decaBDE mixtures in China are manufactured in the areas of Laizhou Bay, Shandong Province, Bohai Sea, and Northeast China. High abundance of BDE-209 in total PBDEs has been found in environmental media in this area (illustrated in Figure S1). Furthermore, the levels of BDE-209 (with a median concentration of 4.0 ng/sample) were observed around Laizhou Bay, Shandong Province, which contributed to 31% of BDE-209, thereby indicating that the manufacturing base is a significant source for total PBDEs in the atmosphere of North China. This was established through gridded field observations with passive air sampling.[69] Jun et al. detected the concentrations of PBDEs in soil and sediment near the manufacturers of decaBDE mixtures in the vicinity of Laizhou Bay and determined that BDE-209 had the highest concentrations (ranging from 58.2 to 7190.7 ng g^{-1} dw), accounting for 74.81−99.01% of the total PBDEs.[70] The concentration of BDE-209 in the production area and its adjacent area in Shandong Province exhibited a relatively high level and appeared to decrease gradually toward the surroundings (Figure 3). The coefficient of correlations between BDE-209 and the other BDE congeners (BDE-183, BDE-154, BDE-100, and BDE-99) was negative, whereas the positive relationship with BDE-99, BDE-153, BDE-154, and BDE-183 might be attributable to the debrominated degradation products of BDE-209.[70] Some studies also support the assumptions that highly brominated PBDEs in a solvent of water/methanol can be photodegradable to low brominated congeners under natural light,[6] and Gerecke et al. posited that BDE-209 could be anaerobically degraded to low brominated congeners in sewage sludge.[71] However, the studies for the degradation of BDE-209 still require further in-depth research. Moreover, BDE-209 is emitted into the air via the factories; however, the air−soil conversion rate of BDE-209 in Laizhou Bay is still not clear. Pan et al. reported that BDE-209 ranged from 0.01 to 53 ng g^{-1} dw with a mean value of 54 ng g^{-1} dw and from 0.66 to 12 ng g^{-1} with a mean value of 5.1 ng g^{-1} dw in riverine and marine sediments, respectively, which are located close to the manufacturing base of BFRs in Laizhou Bay.[72] This finding also suggested the strong influence of pollution discharges from local BDE-209 manufacturers. Pan et al. observed that the mean concentration of BDE-209 (7 ng g^{-1} dw) in 44 surface sediments collected from the Bohai Sea was 1 order of magnitude greater than those of the other congeners.[73] Laizhou Bay is the location surrounded by the Bohai Sea, which proves that the input generated by manufacturing activities is one of the major sources of BDE-209 in this area. This conclusion is also supported by another investigation, in which concentrations of BDE-209 generally decreased from the coastal areas toward the outer sea.[74] In the air samples from Laizhou Bay, the

total concentrations of PBDE (range of 75−240 ng m^{-3} and mean of 140 ng m^{-3}) were even much higher than those in ambient air from e-waste dismantling areas in southeastern China (0.900 ng m^{-3}).[75,76] BDE-209 was the dominant congener, accounting for 99.96% of the total congeners in the air, suggesting that it was caused by the extensive local production of BDE-209.[75] In 2016, Zhen et al. determined that Dalian Bay, which is located in the intersection of the Bohai Sea and Yellow Sea, exhibited a concentration (3.94−103 ng g^{-1} dw) of BDE-209 as was observed in surface sediments.[77] This is likely due to the presence of the electronics industry in Dalian city since the 1980s, and BDE-209 could also have been transported from the manufacturing bases in Laizhou Bay.

In the BDE-209 production area, the most abundant congener of PBDEs in human serum from operators (36 samples) was BDE-209 (ranging from 90.2 to 4181.0 ng g^{-1} and 69.8% on average), and it was also ascertained that the thyroid indexes for those operators are abnormal compared with normal ranges (specific data can be seen in Table S2), which raises concern of a health risk concerning the people in production areas or those working inside of the factories.[78] He et al. discovered that the increased BDE-209 in human serum in this region, between 2007 and 2011, might be due to the reason that penta- and octaBDE mixtures were banned from production during this period.[79] In biological samples, BDE-209 was still the predominant congener in plants and shellfish in these areas, which is consistent with the fact that the decaBDE mixture is the only PBDE product in Laizhou Bay at present.[80] By combining the high concentration of BDE-209 in sediments, those sediment-dwelling organisms might take up BDE-209 from sediments and amplify in food webs.

All above studies confirmed that BDE-209 was dominant in environmental media and organisms, which indicates that the production of commercial decaBDE products might be the primary source in Laizhou Bay including all Bohai seas. Whether BDE-209 belongs to POPs is still undecided; however, more concerns and regulations should be established toward these areas.

3.3. Wastewater Treatment Plant. BDE-209 exists in various consumer products, which could be emitted during usage and disposal and eventually transported into municipal wastewater treatment plants (WWTPs). The concentrations of BDE-209 and nonaBDE are found in three typical WWTPs in China, as depicted in Table S3. Manifestations of BDE-209 have been found in conventional WWTP in Shanghai, ranging from 27.9 to 49.1% in effluents and as high as 85.4% in dewatered sludge.[81] High fractions of nonBDE were also unexpectedly discovered from the samples of wastewater, accounting for 13.1− 30.4%, whereas only less than 10% was present in commercial PBDE products.[81] This discrepancy might be explained owing to the fact that the primary solar photodegradation products of BDE-209 appeared to be two nonaBDE congeners (BDE-206 and BDE-207) that further debrominate to less brominated PBDE congeners.[82] Gerecke et al. also confirmed that BDE-209 can debrominate in sewage sludge under anaerobic conditions.[71] Therefore, it is concluded that some of the nonaBDE might originate from BDE-209 and nonaBDE in WWTPs, which is a significant indicator for BDE-209. The source of BDE-209 in this WWTP was considered to be in-house dust because of the high concentrations of PBDEs observed in Shanghai (up to 3886.7 ng g^{-1}) and GZ (up to 9654 ng g^{-1}). These are two of the largest cities in China, and

BDE-209 exceeded 85%, which is comparable to the congener patterns in the influent.[83]

In Hefei city (population of 4.3 million), Anhui Province, East China, Wang et al. investigated WWTP in the city's oldest and largest wastewater facility of the surrounding Chaohu Lake Basin and determined that BDE-209 accounted for more than 90% of the total PBDEs in the wastewater, where the BDE-209 abundance is the highest as compared with those in Shanghai and Hong Kong (Figure S2).[84] The predominant congener of BDE-209 might be from the car dust because Hefei city boasts a great number of car manufacturing plants that are associated with a considerable amount of car accessories that contain BFRs, and BDE-209 was also found to be the dominant congener in the Nanfei River.[84] The car dust might be one of the most predominant sources of contaminant in WWTP in Hefei by air transport. Wang et al. discovered an interesting result in which the removal rate of BDE-209 is indistinctive (around 20%) during the treatment process, which might reflect the special characteristics of the wastewater or the treatment conditions.[84] However, a reasonable or distinct explanation is still unknown. Den and Tam ascertained that isolated microalgae (*Chlorella*, *Parachlorella*, *Nitzschia palea*, and *Mychonastes*) from WWTP exhibited a high removal rate of BDE-209 (82−90%) with a high tolerance (spiked up to 600 μg L^{-1} for 7 days).[85] In spite of BDE-209 being regarded as the lowest bioavailability of PBDEs, BDE-209 can be adsorbed by microorganisms (on microalgae cell surfaces) because of their high hydrophobicity, which leads to the high removal of BDE-209.[86] WWTPs in Hong Kong receive both domestic and industrial wastewaters; therefore, the congener profile in the influent indicated an enormous usage of penta- and decaBDE commercial mixtures because the predominant congeners were BDE-209, followed by BDE-47.[87] In WWTPs, the concentration of BDE-209 in the treated effluent was especially lower than that in the influent, and BDE-209 and BDE-47 in fluents switched to BDE-47 and BDE-99.[87] The same shift in the partitioning of PBDEs was also validated by Kim et al. because of a higher water partition coefficient (K_{ow}) of BDE-209 than that of BDE-47 and BDE-99; thus, there is a higher rate of BDE-209 as compared to that of the solid particles.[88]

3.4. Urban and Rural Areas. BDE-209 appeared to be the most abundant congener in urban areas and to have a low abundance in rural and remote areas where the economics are relatively undeveloped and where the utilization of products containing flame retardants was lower. However, reports showed a very low abundance no matter whether in urban or rural areas (Figure S3). The air (gaseous and particulate phase) in the urban areas of GZ exhibited 100 and 150 m above the ground in the daytime and during the night, between August and December 2010, whereas over 60% of the particulate PBDEs was found in particles collected by an aerodynamic diameter below 1.8 μm. Of the targeted gaseous and particulate PBDEs, BDE-209 was the predominant congener with a percentage of 72.2 ± 8.3 and 78.5 ± 6.9 among congeners by weight in the summer and winter, respectively, indicating that BDE-209 has the tendency to affiliate with the particles in the air, suggesting that BDE-209 has the potential of long-range transport.[89] The diurnal variability of meteorological factors has also proved to affect the dry deposition velocities of BDE-209 because the mean daily particle dry deposition fluxes of BDE-209 ranged from 88.6 ng m^{-2} day^{-1} in August to 122 ng m^{-2} day^{-1} in December.[89] Yang et al. utilized high-volume air samples to collect air samples during September 2008 to

August 2009 throughout China (11 urban, 1 suburban, and 3 background/rural) and determined that BDE-209 took up 64 ± 23% of the total 12 congeners, followed by BDE-47 (8 ± 8%) and BDE-99 (6 ± 5%) at all urban and suburban sites, whereas BDE-209 only accounted for 11 ± 2%.[90] This diversity is likely a result of BDE-209 tending to bind to particles so that less BDE-209 migrates with the air mass movement to the background/rural sites as compared with low brominated PBDEs.[90] The ratio of BDE-209 to the total PBDEs in both urban and background/rural sites of China was higher than that of the air in the United States, indicating a larger usage of commercial decaBDE mixture in China.[90] Tian et al. found that the atmospheric deposition profile in the urban areas of southern China was dominated by current-use halogenated flame retardant (BDE-209), whereas the profile at e-waste sites reflects that a massive amount of BDE-209 was used in the past. Moreover, the deposition source of BDE-209 within the rural sites was mainly attributable to the contribution of urban (45%) and e-waste (38%) sources, which was estimated by PCA with a multiple linear regression analysis.[39] On the basis of the ratio of vapor pressure and K_{OA} (15.3) of BDE-209 (<1), BDE-209 might undergo degradation during transportation, which was also supported by exceptionally high ratios of BDE-202 taken as a marker of environmental debromination of BDE-209.[39,91] This finding is consistent with other results stating that the photodegradation of BDE-209 might occur during long-range atmospheric transport (LRAT) on mineral aerosol, which is a significant fate process for BDE-209 in the environment.[8,39] The same pattern was also validated by Tian et al. in rural sites of southern China with 59 ± 16% BDE-209 of total PBDEs.[43] A significant concentration of BDE-209 was observed in the atmosphere (an arithmetic mean concentration of 478 pg m^{-3}) in urban areas within the city of GZ, southern China, accounting for 48 to 79% among the total PBDEs, which was similar to the profile in Japan but much higher than the abundance in North America and Europe.[92] This implies that different PBDE formulas were predominantly utilized in different regions. In other urban areas of Chaohu city, BDE-209 was detected with concentrations ranging from 3.9 to 262.3 pg m^{-3} (58.5 pg m^{-3} on average), whereas significant low concentrations of BDE-209 (ranging from 2.2 to 72 pg m^{-3}, 15.4 pg m^{-3} on average) were measured in rural areas (117°27′56″ E, 31°34′55″ N) and in the town of Zhongmiao, which is 40 km west of Chaohu city. Moreover, there were no significant differences for most of the congeners between the urban and rural areas in the gaseous (p = 0.0509−0.906) and particle phases (p = 0.077−0.802) except for BDE-209 (p = 0.002) in the particle phase, based on the results of an independent t-test analysis, pointing to different sources of BDE-209 in urban and rural areas.[93] Zhang et al. estimated the annual dry deposition of BDE-209 to be 1340 kg/year in southern China (within a combined area of GZ, Dongwan, and Shunde cities), and the annual wet deposition rates of BDE-209 were estimated to be 490 kg/year, which is an important mode to remove BDE-209 from the atmosphere in urban areas.[94] These reports mean that BDE-209 is a major potential source in both northern and southern China; the air mass back-trajectory analysis reflected that BDE-209 is widely distributed in urbanized and industrialized areas in tropical Asia.[95]

Because of large sorption capacity, soil is an important reservoir and sink for BDE-209 and further plays an important role in the distribution and the biogeochemical cycling of BDE-209. The concentrations of BDE-209 in the surface soil from

the Pearl River Delta, South China, ranged from 2.38 to 66.6 ng g^{-1} with a mean concentration of 13.8 ng g^{-1}, of which a range of 25.7−102 ng g^{-1} registered in the point-source contaminated soils.[19] The mass inventory of BDE-209 in soil was estimated to be 44.4 t, which was comparable to that in the sediments from the Pearl River Estuary. This suggests that surface runoff and soil erosion are the important modes to transport BDE-209 from soils to oceans in this region.[19,96] Wang et al. discovered the highest concentrations of BDE-209 (58 445 ng g^{-1} dw) in soil of the e-waste dismantling industrial park from 65 sampling sites investigated from diverse industrial characteristics in Zhejiang Province, South China, which are similar to the levels measured in floor dust (5560−80 600 ng g^{-1} dw) from the e-waste dismantling workshop from this area.[97] BDE-209 was reported to be the predominant congener in both soil and dust in Shanghai city, whereas the contribution of BDE-209 was much lower in dust as compared with that in soil.[98] This is perhaps due to extreme hydrophobicity of BDE-209, and it has been considered to possess low bioavailability and tendency to bind with soil and sediment tightly.

3.5. Lakes, Rivers, and Oceans. In abiotic media from lakes, rivers, and oceans in China, BDE-209 was the dominant congener, whereas the fish species in fresh and sea water revealed a very low abundance of BDE-209 (Figure S4). Air and water samples were collected from Lake Taihu, East China, in 2004 for analyzing PBDEs, and BDE-209 was the dominant congener in all assessed PBDE congeners, constituting 41 ± 21% on average of the total gaseous and particulate-bound PBDEs.[99] BDE-209 was measured in riverine runoff samples from major outlets within the Pearl River Delta with concentrations varying from 0.335 to 65.2 ng L^{-1}, which was the dominant congener with an annual input of 1960 kg/year.[100] Massive PBDEs imported to China in the form of e-waste were estimated to be 35 000 t/year, which is higher than the annual production of BFRs (about 10 000 t/year), and the annual riverine input of total PBDEs from the Pearl River Delta, indicating that the main inventory of BDE-209 might constitute accumulated imported e-wastes.[100]

BDE-209 was detected in river sediments from Shanghai with a median concentration of 45.7 ng g^{-1} dw as the most abundant BDE congener.[101] This result could be due to the increased usage of the commercial decaBDE mixture in Shanghai, which hosts several coal-fired power plants and municipal solid waste incinerators that might contribute to BDE-209 in these sediments.[101] Wang et al. investigated PBDEs in water and sediment samples from Liushi town, Zhejiang Province, South China, across different areas, in which water samples were dominated by BDE-209, BDE-47, and BDE-153 with BDE-209 accounting for roughly 50% of the total PBDEs, and the same profile was also found in most sediments (up to 58 445 ng g^{-1} dw),[97] which was similar to those in other areas in China[79,102] but much higher than those in other freshwater sediments such as the Spanish River, Vero (12 500 ng g^{-1} dw),[102] and the Lake Maggiore Basin in Italy and Switzerland (15 ng g^{-1} dw).[103] This might be the reason for the small dilution in the river systems in this area.[97] PBDEs were extensively investigated by Wang et al. in samples of sediments from several major river basins around China.[104] Among these PBDEs, BDE-209 was found in all samples from the Yangtze River Basin and Pearl River Basin with concentrations up to 61 500 ng g^{-1} dw in the Yangtze River Delta, whereas BDE-209 contributed to over 60% of the total PBDEs in the Yellow River Basin. This suggests that the commercial decaBDE mixture might have

been ubiquitously utilized in most cities of China.[104] The concentration of the most abundant BDE congener in surface sediments in the Beijiang River, Guangdong Province, South China (the concentration was up to 1558 ng g^{-1} dw) was much lower than that in the Yangtze River, lower than the Zhujiang River (up to 3580 ng g^{-1} dw) and the Dongjiang River (up to 7340 ng g^{-1} dw) in the same region.[105]

From the coastal areas toward the outer sea, BDE-209 registered a decreasing concentration because of the influences of anthropogenic activities.[106] BDE-209 accounted for 60.6−97.9% (a mean value of 87.0%) of the total detected PBDEs in the Bohai Sea, which indicated that commercial decaBDE was widely utilized within the coastal cities of China.[106] This result was also validated by another report of BDE-209 in the sediment (75−99%) and mussels (61−99%) in the Bohai Sea.[107] The mean concentration of BDE-209 in coastal sediments (0.68 ng g^{-1} dw) collected from the Yellow Sea, North China, was lower than the level found in the Bohai Sea (2.29 ng g^{-1} dw), East China Sea, and Xiamen offshore area (14.94 ng g^{-1} dw); it was comparable to the level found in the Yangtze River Delta (0.41 ng g^{-1} dw).[107−110] These high levels of BDE-209 were found around BFR manufacturing areas, urbanized cities, and areas with numerous electrical factories.[108] In all mangrove sediments in Hong Kong, the concentrations of BDE-209 (ranged from 1.53 to 75.9 ng g^{-1} dw) were 1−2 orders of magnitude higher than those of other congeners in the same sediment, and the mangrove swamps are severely influenced by the pollution from the Pearl River Delta.[111] From the coastal East China Sea, extending about 1000 km from the Yangtze River Estuary to the south, the levels of BDE-209 were 0.3 to 44.6 ng g^{-1} dw, which were consistent with the current and historical usage of the decaBDE mixture in China. The compositions and distribution of BDE-209 suggest that BDE-209 in the coastal East China Sea could be primarily from coastal waste dismantling/recycling and Yangtze River input.[112] Wang et al. studied the depth profile of BDE-209 in a sediment core P01 from the Yangtze River and revealed that the input of BDE-209 increased gradually from 1930 to 2010.[113] The same profile, which was also found in the Pearl River Estuary, South China, illustrated that the concentration of BDE-209 in sediment cores remained until 1990 and exponentially grew afterward with a doubling time of 2.6 ± 0.5 to 6.4 ± 1.6 years, and the inventory of BDE-209 in sediments was 386.2 ng cm^{-2}.[96] Moreover, the flux of BDE-209 into the Pearl River Estuary was evaluated to be 29.7 ng cm^{-2} year^{-1} and increased annually with corresponding values in the annual industrial gross output of manufacturing electronics in this area. This suggests that the rapid growth of the electronics industry in the Pearl River Estuary since the early 1990s contributed to the sharp increase in BDE-209 fluxes during the past two decades.[96] This result in the Pearl River Estuary was also determined by Mai et al. who posited that the waste discharges from the cities adjacent to the Pearl River Estuary (such as GZ, Dongguan, and Shenzhen) were the major sources of BDE-209.[20] In addition, Chen et al. proffered that obvious elevated sediment concentrations of BDE-209 in the rural areas of the Pearl River Estuary were due to e-waste dismantling and that more lower brominated PBDEs might be from the biological or photolytic debromination of BDE-209 in the environment.[114] There is also an interesting observation that alternative BFR (decabromodiphenyl ethane) to PBDEs has exceeded the concentrations of BDE-209, which might also be a need for concern.[114] Although BDE-209 was considered to be input

mostly by LRAT in remote areas such as the Tibetan Plateau, Yuan et al. suggested that the small account of the local source in the Central Tibetan Plateau is based on the detection of BDE-209, which might be due to the sampling areas that are in close proximity with the two biggest cities in Tibet (Lhasa and Shigatse).[115]

4. HUMAN EXPOSURE

No direct studies of BDE-209 absorption in humans were reported. Human absorption was derived from the measurements of BDE-209 in human biological media after anthropogenic exposure. PBDEs, such as BDE-209, are capable of accumulating in tissues of organisms and are magnified via the food chain, and then they finally pose adverse effects on human health. BDE-209 in marine sediments represents an exposure path to fish, and the consumption of fish is a significant exposure pathway for humans.[113] Wang et al. estimated the potential cancer risk of human exposure at a 95% confidence level to BDE-209. The cancer risk was estimated based on the following equations

$$HI = \sum \frac{DI_i}{RfD_i}$$

$$Cancer\ risk = \sum DI_i \times CSF_i$$

where HI represents noncancer risk, DI_i represents the daily intake of congener i by consumption of fish $(mg\ kg^{-1}\ day^{-1})$, RFD_i is the reference dose of congener i $(mg\ kg^{-1}\ day^{-1})$, and CSF_i is the cancer slope factor of congener i $(mg\ day\ kg^{-1})$. For BDE-209, CSF_i is 0.0007 m day kg^{-1}[116] and RFD_i is 0.007 mg $kg^{-1}\ day^{-1}$.[117]

However, the concentrations of BDE-209 in fish and sediments were 3.09×10^{-7}, 1.67×10^{-7}, and 8.86×10^{-7} for children, teens, and adults, respectively, which are significantly lower than the level of the threshold (10^{-6}); the calculation of the hazard index for noncancer risk was also much less than 1 for the three groups, suggesting a no-cancer risk.[113] This result was also proven by Man et al. who employed the same method to calculate the cancer risk values of soils, based on the total concentration of BDE-209 from six sites of agricultural soils from Hong Kong, which were below 1 in a million people at 5th, 50th, and 95th centiles.[118] Because of the continuous use of the commercial decaBDE mixtures, it is expected that the levels of BDE-209 will still be found in human samples.[37]

4.1. Inhalation Exposure.
Inhalation or ingestion of indoor dust has been identified to be the main pathway of PBDE exposure for people, especially children.[119] Settled workplace dust collected from several places (e.g., shopping malls, hospitals, and electronic factories) and home dust in Hong Kong, Shenzhen, and GZ around the Pearl River Delta were measured for PBDEs with the highest levels (2122–40 236 ng g^{-1}), dust from homes ranged from 685 to 18 385 ng g^{-1}, and the predominant congener was BDE-209 in both workplace dust and home dust, which is the same level as the outdoor dust from Shanghai.[32,33] There were no significant correlations found between the concentrations of BDE-209 and the number of electronic appliances $(p > 0.05)$, and the decaBDE mixture is likely to undergo photolytic degradation owing to the ratio of BDE-209/(BDE-207 + BDE-206) of the workplace dust samples, which revealed a range from 12 to 30.[32] Wang et al. found the most significant contributor to

PBDEs in household dust sampled from GZ and Hong Kong $(PM_{2.5}$ and TSP) where $PM_{2.5}$ contained the highest BDE-209.[120] Wang et al. considered that dust ingestion was the predominant PBDE exposure pathway for toddlers, whereas no exposure route for BDE-209 exposure was provided.[120] The median hazard quotient of dust ingestion in China (samples collected from 23 provinces across China) for BDE-209 observed for infants, toddlers, children, teenagers, and adults ranged from 2.51×10^{-6} to 7.83×10^{-4}, which is far less than 1, which suggests a low risk to humans.[33] However, the maximum hazard quotient of BDE-209 was observed with the value of 0.73 for toddlers, which was close to 1.[33] In southern China, Ding et al. observed positive correlations between the indoor and outdoor dust samples for BDE-209, suggesting the exchange of BDE-209 between the two compartments.[121] Ni et al. observed that the ingestion of dust and soil constituted the major exposure routes for BDE-209, whereas contribution of other congeners (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, and BDE-183) could be ignored.[116] For instance, BDE-209 contributed to more than 90% of the total PBDEs' daily intake via soil ingestion or indoor dust exposure, and they also thought that indoor dust should not always be the most important exposure route for PBDEs.[116]

4.2. Dietary Exposure.
BDE-209 was not always considered to be bioavailable because of its high molecular weight; therefore, the premise arose that it is not harmful to organisms and humans.[122] However, the occurrence of BDE-209 was discovered in several organisms, that is, roaches, gray seals, sharks, and so forth.[122−124] In southern China, the area with the heaviest PBDE contamination, the presence of BDE-209 in several seafood products (shrimp, crab, and shellfish) was found in 44.4% of the total samples, which suggested that BDE-209 appears to be more bioavailable than previously thought.[65] In this study, BDE-209 levels ranged from nd (not detected) to 3.21 ng g^{-1} dw, which were lower than those from South Korea (0.23−14.8 ng g^{-1} dw), Hong Kong (0−14.3 ng g^{-1} dw), and the Netherlands (4.9 ng g^{-1} dw).[63,65,125−127] Wu et al. (2008) found PBDE congener profiles in rice frogs (*Rana limnocharis*), in which BDE-209 only constituted 2−14%, with BDE-99 and BDE-153 being the largest contributors, which were different from those in organisms from aquatic (predominance of BDE-47) and terrestrial sources (predominance of BDE-209).[128] This is attributed to the feeding habits of frog, which combine aquatic and terrestrial circumstances. A relatively lower BDE-154 (5−9%) was also observed.[128] Wu et al. suggested that BDE-154 might be from the biotransformation of BDE-209, which is also consistent with other reports that BDE-154 is a metabolic product of BDE-209 in fish.[128−130] In July 2016, Su et al. detected PBDEs from selected food items, including seven aquatic foods (Mandarin fish, catfish, Chinese snakehead, Black bass, crucian carp, and white Amur bream), purchased from different markets in Nanjing city, South China, and BDE-209 was not detected in fish or other food.[131] This observation could be due to the fact that lesser brominated BDEs are more easily accumulated than high brominated congeners, which also have higher molecular weights. In addition, BDEs with higher molecular weights, such as BDE-209, are more easily biotransformed than those with lower molecular weights. Those biotransformed lower brominated congeners might form MeO-PBDEs and OH-PBDEs that are more likely to cause adverse effects, because analogues of PBDEs exhibited dioxin-like potency such as 6-OH-BDE-47 and 6-MeO-BDE-47, which have been quantified

in various environment media and marine organisms.[132] Ni et al. also estimated that the cancer risk for BDE-209 exposure in adults ranged from 5.8×10^{-23} to 5.5×10^{-21} (far less than 1) and that hazard quotients for BDE-209 were 1.2×10^{-5}, which was based on the daily intake from food consumption in Shenzhen, China, which suggested that the low deleterious risk regarding BDE-209 and BDE-209 exposure via food is very low for adults.[116]

Duck eggs, as a common dietary component, were collected from TZ city, which has many e-waste recycling sites in eastern China and in the majority of samples (duck yolk), and BDE-209 was the most abundant congener (62%), with a mean concentration of 500 ng g^{-1} lipid weight (lw), and it was also detected in feed samples (0.19–5.12 ng g^{-1} dw) from the printed circuit board's recycling location.[133] Exposure estimates for duck eggs did not exceed the reference values by the U.S. EPA, though BDE-209 was not estimated.[133]

4.3. Burdens of the Human Body. BDE-209 was suspected to be low in human serum because of the short half-life of 15 days or being less bioaccumulative because it exhibits the largest molecular size and the highest octanol–water partition coefficient of the total PBDEs.[134,135] However, Wang et al. found that BDE-209 was the most abundant congener in all serum samples by weight, which accounted for 40–68% of the total PBDEs. These were collected from Zhejiang Province, South China, through blood samples from 194 Chinese injection workers from electrical appliance factories and 205 catering workers, leather factory workers, infertile men, and umbilical cords. The injection workers apparently showed higher concentrations of BDE-209 than others without obvious PBDE exposure.[36] This finding was consistent with another report regarding the Chinese resident population, indicating that the population in China are continuously exposed to high levels of BDE-209 in blood serums because of its short half-time (fast turnover rate).[136] However, another study depicts a different pattern, where BDE-209 was detected at relatively low concentrations in all samples taken from umbilical cord blood from South China. Some levels of BDE-197 detected were due to the debromination of BDE-209, as was also found in human bodies and laboratory studies.[35,37,137,138]

Moreover, breast milk has been taken for a biomarker to evaluate the extent of human exposure to PBDEs in the studies of numerous countries, which is of significant concern, as contamination is passed on to the next generation.[34,35] BDE-209 was reported in breast milk from the United States and Spain, which varied from 0.1 to 48%.[139,140] In China, BDE-209 was not the dominant BDE congener in breast milk, and few studies reported the detection of BDE-209 in breast milk.[17] However, a Chinese study showed that BDE-28, BDE-153, and BDE-183 were predominant components in human breast milk from mothers because of the high exposure rate in the decaBDE production areas, suggesting that high decaBDE exposure might lead to increasing levels of low brominated BDEs in the human body.[136] Therefore, more research regarding BDE-209 congeners in human breast milk should be conducted in further studies.

5. FUTURE RECOMMENDATIONS

On a regional scale, the levels of BDE-209 in the environment of China are relatively high as the predominant congener among other congeners. Although more safe nonhalogen flame retardant has appeared in the market, BDE-209 will be still mainly used based on its much cheaper price. Because the proposal for BDE-209 listed to POPs was submitted to POPs Review Committee, Stockholm Convention in 2013, now BDE-209 is under the review for Annex A.[3] BDE-209 still needs continuous attention in the next few decades, especially because pentaBDE was banned in 2004 and octaBDE was banned from China and was never produced in China. However, China continued to accept e-wastes from other countries, and thus, more and more e-waste containing BDE-209 will continue to be generated. Therefore, BDE-209 in several e-waste recycling areas still needs to be continually monitored, especially for the operators and residents living in the region. Eco-friendly approaches such as feedstock recycling, mechanical recycling, and bromine recovery should also be utilized for e-wastes so as to reduce the emission of BDE-209. In addition, BDE-209 emission occurs in the life of products impregnated with this flame retardant or at the ultimate disposal stage. The emissions from product life cycles vary over the life cycle of products, which cannot be accurately monitored. Thus, regulatory measures regarding the management of BDE-209, such as monitoring and production control, are urgently made in China.

The high occurrence of BDE-209 in several different recycling areas has caused a potential exposure risk for organisms and humans. High concentrations of BDE-209 emitted from the manufacturing areas to the surrounding environments continue to grow at an alarming rate. However, commercial decaBDE mixtures are not presently controlled by any regulations. There is an urgent need for further consideration and studies regarding regulations, policies, and standards with respect to the usage and production of decaBDE mixtures. Although a considerable amount of information is available on PBDEs, it is somewhat difficult to draw a clear conclusion regarding BDE-209 exposure in humans because BDE-209 was not detected in many studies. However, the consumers of fish and duck eggs were heavily exposed to BDE-209. Studies on breast milk and blood from individuals residing or working in e-waste recycling areas (QY, GY, and TZ) and Laizhou Bay revealed a high level of BDE-209 when compared to other areas. Home dust is also a primary inhalation exposure of BDE-209 to adults. The current exposure estimates of BDE-209 from different areas in China are below the RFDs of the U.S. EPA, and the cancer risk is below 1. However, the high levels of BDE-209 in workers in e-waste sites and the decaBDE mixtures sites should encourage more research to be conducted, with an overall and accurate risk assessment of BDE-209 for laws and regulations of use and production for decaBDE mixture in China.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.7b00559.

Detailed information for physical properties and chemical identity of BDE-209, thyroid hormone concentrations in human serum, concentrations of BDE-209 in WWTPs, and abundances of BDE-209 in several media (Figures S1–S4) (PDF)

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