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Modeling the Time-Variant Dietary Exposure of PCBs in China over the Period 1930 to 2100

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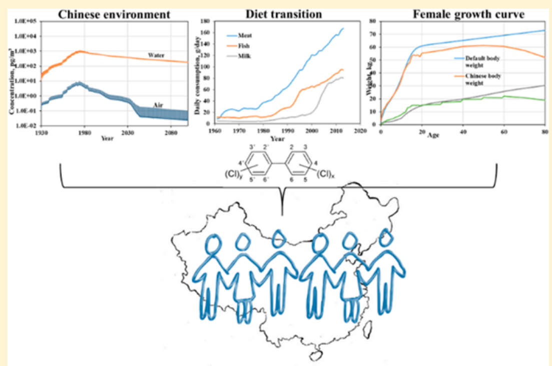
Supporting Information

ABSTRACT: This study aimed for the first time to reconstruct historical exposure profiles for PCBs to the Chinese population, by examining the combined effect of changing temporal emissions and dietary transition. A long-term (1930−2100) dynamic simulation of human exposure using realistic emission scenarios, including primary emissions, unintentional emissions, and emissions from e-waste, combined with dietary transition trends was conducted by a multimedia fate model (BETR-Global) linked to a bioaccumulation model (ACC-HUMAN). The model predicted an approximate 30-year delay of peak body burden for PCB-153 in a 30-year-old Chinese female, compared to their European counterpart. This was mainly attributed to a combination of change in diet and divergent emission patterns in China. A fish-based diet was predicted to result in up to 8 times higher body burden than a vegetable-based diet (2010−2100). During the production period, a worst-case scenario assuming only consumption of imported food from a region with more extensive production and usage of PCBs would result in up to 4 times higher body burden compared to consumption of only locally produced food. However, such differences gradually diminished after cessation of production. Therefore, emission reductions in China alone may not be sufficient to protect human health from PCB-like chemicals, particularly during the period of mass production. The results from this study illustrate that human exposure is also likely to be dictated by inflows of PCBs via the environment, waste, and food.



1. INTRODUCTION

Polychlorinated biphenyls (PCBs) are one of 12 legacy persistent organic pollutants (POPs) initially targeted by the Stockholm Convention,[1] because they are toxic and stable in the environment, undergo long-range atmospheric transport (LRAT), and bioaccumulate in the food chain, representing a potential threat to environmental and human health.[2] China started PCB production in 1965 and ceased production at the end of 1974.[3] During these years, the accumulated production amount reached approximately 10,000 tonnes, accounting for 0.8% of total global production. Although China is not a main PCBs producer and has banned them for decades, these chemicals are still of concern and are frequently detected in the environment and organisms.[4,5]

Biomonitoring is a potentially important tool to assess human exposure to PCBs from the ambient environment. In China, several biomonitoring studies have been conducted in heavily polluted regions, e.g., the e-waste recycling regions in the southern and eastern parts of China.[4,6−8] However, long-term cross-sectional (studies sampled at a single time point) and longitudinal (studies conducted on single individuals over a person's entire lifetime) biomonitoring studies in control areas are very rare.[9,10] As empirical human biomonitoring data are largely restricted to snapshots in time at contaminated hot spots, dynamic mechanistic models can offer complementary insights, helping to hypothesize key factors likely to affect past, contemporary, and future body burdens of the general Chinese population. Moreover, an integrated modeling strategy could inform future biomonitoring strategies as well as support interpretation of empirical data.

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However, developing a model to dynamically describe relationships between emissions and human exposure is challenging, given the numerous factors which affect source-exposure relationships of PCBs. Dietary exposure is an important source of PCBs, accounting for up to 90% of the total intake, especially for foods of animal-origin rich in lipids.[8] The combined effect of temporal trends in emissions and dietary preferences is suggested to have a significant effect on temporal trends in human body burdens.[11] A 6–13-fold decrease in PCB-153 body burden was reported from 1980 to 2020 due to dietary transition for an Arctic population (e.g., less reliant on traditional food items with high PCB concentrations such as seal meat[11]). In contrast, the intake of food items with potentially high PCB concentrations has increased in China with 17, 3, and 8 times higher consumption of meat, milk, and fish from 1950 to 2013, respectively (FAOSTA: http://faostat3.fao.org/home/E).

The relationship between age and human body burden for POPs has been broadly discussed, but no consensus has been reached so far.[12] The influential factors mainly include exposure history, metabolic/depuration half-lives, sources, and exposure pathways. POPs' concentrations in the human body were frequently reported to be positively associated with age in human cross-sectional studies, due to long-term exposure and poor ability to metabolize these substances[13−15] where age and birth cohort effects are confounded. However, a decreasing trend in serum concentrations with age was also observed, which may be due to steady-state exposure levels being reached.[14] In addition, growth dilution may reduce concentrations for people aged younger than ∼16 years old.[16] Several studies reported no significant correlation between concentrations in humans and age of participants in an industrialized area,[17−19] while Sun et al. observed a positive relationship between age and concentrations of dioxin-like PCBs.[20] However, all studies were conducted after the ban on PCBs and were based on limited sample sizes. Therefore, there is a need to rebuild the exposure history for the Chinese population and systematically explore the age burden relationship under temporally variable emission scenarios and dietary shift patterns.

The purposes of this study were therefore 1) to reconstruct the historical exposure profile and predict future exposure trends under multiple scenarios for Chinese female cohorts using PCB-153 as a case study, which can be directly linked to mother-to-fetus transfer; 2) to assess the combined effect of dietary transition and emission trends on human exposure over the longitudinal and cross-sectional trends; and 3) to explore the impact of different PCB emission sources on human body burden as predicted by the applied models.

2. METHODS

2.1. Conceptual Approach. Assessing implications of emission trends and dietary transition on human exposure to organic contaminants requires an integrated approach combining a dynamic chemical fate model and bioaccumulation model. In this study, the overall approach was modified from the pioneering approach of Quinn et al.[11] with the following elements developed and synthesized: 1) emission rate estimations over time (1930–2100) worldwide and in China were developed; 2) environmental concentrations responding to the emission scenarios were predicted; 3) food web bioaccumulation covering the main pathways of chemical accumulation in the Chinese population (e.g., water-fish-human) was incorporated; 4) scenarios of different dietary patterns were explored; 5) scenarios defining trends of the dietary transition in the future and their possible implications for human exposure to PCBs were explored. Simulations were performed to calculate human body burdens (ng g^{-1} lipid) as a function of time (year), i.e., longitudinal body burden versus age trends.

2.2. Emission Scenarios. Several historical PCBs emission scenarios were explored to assess the individual and combined influence from three sources: (i) intentionally produced PCBs; (ii) e-waste imports; and (iii) unintentional formation. For the former two sources, global historical emission inventories (1930–2100) published by Breivik et al.[21,22] were used. While the "baseline scenario" estimates global PCB emission without considering transboundary movement of e-waste, the "worst-case scenario" additionally accounts for emissions associated with imported e-waste from OECD to non-OECD countries.[21] Emissions from unintentionally produced PCBs (UP-PCBs), which mainly originate from industrial thermal sources, have been identified as providing an important contribution to total PCB emissions in China in the near future.[23] Emissions from outside China from this source category are not considered, due to lacking a global emission inventory for UP-PCBs. The "default scenario" therefore was defined as total PCBs from intentional production, combined with e-waste imports and unintentional formation, where the individual influence of PCB emissions from imported e-waste and unintentional emissions was also evaluated. Each emission scenario was allocated to a 1° latitude × 1° longitude grid system based on a global population density database.[24]

2.3. Selected Models. *2.3.1. Fate Model.* To predict ambient environmental levels of selected PCB congeners in the global environment over time, the default scenario as defined in Section 2.2 was used as emission input to the multimedia fate model BETR-Global.[25,26] This model has previously been evaluated and successfully applied to PCBs.[21,25−27] The study region (covered grid cells assigned numbers of Grid 66, 69, 90, 91, 92, 93, 115, 116) is illustrated in Figure S1. The BETR-Global model has a spatial resolution of 15° latitude × 15° longitude, consisting of 288 grid cells. Each of these regions consists of up to seven bulk compartments, including ocean water, fresh water, upper air, lower air, soil, freshwater sediments, and vegetation. The detailed environmental parameters were sourced from a wide range of databases, and GIS was used to calculate the characterstics of each region.[28] The model regions are connected by advective transport via air, fresh water, and ocean water. PCBs emissions were allocated to the 288 grid cells. Only emission to lower air was considered. The initial model concentration was assumed to be zero. This model was run dynamically for the period from 1930 to 2100. Seven indicator PCBs (PCB-28, -52, -101, -118, -138, -153, -180) were selected for simulation, although PCB-153 was selected as an indicator PCB and mainly discussed here. Model input data characterizing the properties of individual PCB congeners was selected from the literature[29−32] and is summarized in Table S1.

2.3.2. Bioaccumulation Model. Chemical bioaccumulation in food chains was modeled by a mechanistically based, nonsteady state bioaccumulation model (ACC-HUMAN),[33] which has been previously shown to provide reasonable results for PCB bioaccumulation in the human food chain.[33−35] It is subdivided into an agricultural and an aquatic food web. The considered uptake pathways of contaminants are diet and

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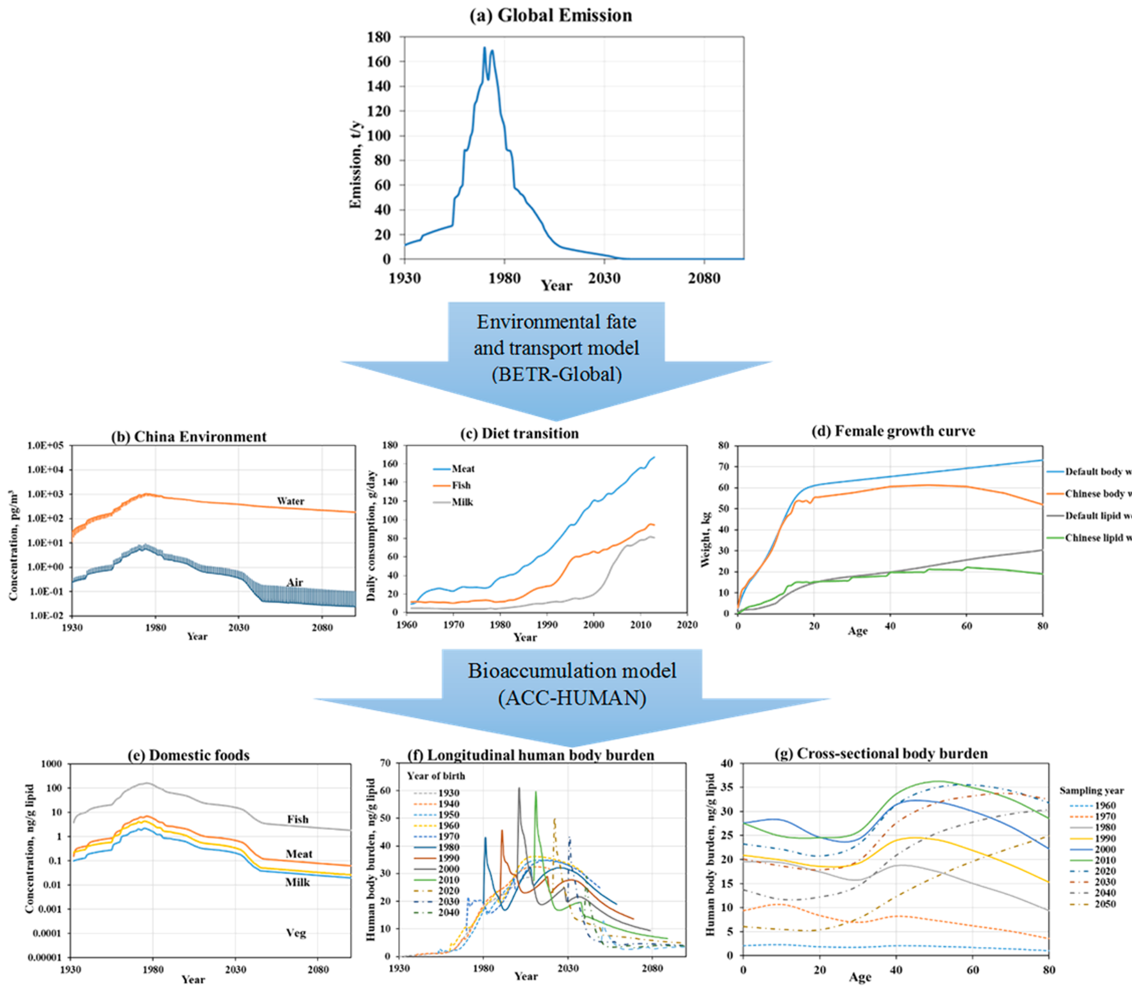

Figure 1. Schematic overview of the modeling approach employed to assess the combined effect of emission trends and dietary transition on human exposure to PCB-153 for the Chinese female population. The approach was modified after Quinn et al.[11] The global emission estimate of PCB-153 over the period 1930−2100 under a default scenario (a) was used as input to a global fate and transport model (BETR-Global) to predict average environmental concentrations in a target region (presented in Figure S1) (b). The estimated environmental concentrations in lower air and fresh water (b), dietary transition scenarios (c), and female growth curves (d) are used as inputs to the bioaccumulation model (ACC-HUMAN) to predict the concentration in respective food items (e) and the longitudinal human body burden for a 30-year-old Chinese female born in different years (f). The cross-sectional versus age dependence was modeled every ten years from 1930 to 2050 (g). The short dashed lines present the period with increasing emission (1930−1970), while the long−short dashed lines show modeling results after the ban of all intentional emissions defined in Section 2.2 (2020−2050).

inhalation, while the elimination pathways are metabolism, percutaneous excretion, digestive tract excretion, exhalation, childbirth, and breastfeeding.[33] Since PCBs mainly enter the body via diet, the inhalation pathway was not discussed here.

Environmental concentrations of air and freshwater (outputs from the BETR-Global model) were used as inputs along with physical-chemical properties of a given PCB congener. Based on these inputs, the ACC-HUMAN model was used to calculate the time course of lipid-normalized PCB concentrations in the human body. All parameters suggested in the previous studies were adopted,[33,36] with the exception of dietary pattern transition and human characteristics (e.g., growth curve, lipid content, and body weight), which was modified for the Chinese population as illustrated in Figure 1(c) and (d). Different scenarios for dietary habits are defined in Section 2.4.

Cross-sectional data generated through biomonitoring studies are based on groups of different individuals sampled at the same time, whereas the longitudinal estimates derived from ACC-HUMAN model are for a single individual over a person's entire lifetime. Cross-sectional trends were determined from the model-derived longitudinal estimates of lipid-normalized concentrations for individual females born at successive 10-year intervals. This reduces the confounding effect of the birth cohort on the human body burden.

2.4. Dietary Information for the Chinese Population.

2.4.1. General Diet Pattern and Transition. Food supply data for domestic consumption from 1959 to 2013 (http://faostat3.fao.org/browse/FB/CL/) was used as the default dietary pattern to represent dietary transition trends at a national level. This was calculated based on the food production plus imports minus exports. The domestic food supply of meat, milk, and fish increased by around factors of 17, 3, and 8 (illustrated in Figure 1(c)), on a national scale during the period 1959−2013. For the period from 1930 to 1959 without recorded diet information, the dietary pattern was assumed to be the same as 1959. This is a first approximation to gain a general overview of

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dietary transition in China. Potential uncertainties include regional supply variances between different subpopulations.

The default lipid content of human dietary items in ACC-HUMAN was reset to 5.2% for fish and 3.2% for milk in Chinese food products.[37] Unlike Western populations, for which ACC-HUMAN was originally developed, pork is the main meat type consumed in China.[38] Thus, the beef cattle component in ACC-HUMAN was reparametrized. Chinese pigs are mainly fed on corn, but their diet may also include discarded food of animal origin, which would potentially underestimate the contaminant levels in pigs. However, this study was intended to be representative of generic trophic levels in China, and acceptable modeling results are demonstrated in Section 3.1. Pork contains up to 30% lipid content, highest among varied meat types.[37] The worst-case scenario, assuming that the Chinese population only eats pork, was also assessed and modified in the ACC-HUMAN model. The dietary transition excluded data for vegetables, since vegetable consumption has remained relatively stable at around 276 to 310 g day^{-1} per person.[39] Considering the relatively low PCB concentrations in vegetables, it was assumed that the resulting variation would be minimal.

2.4.2. Regional Differences. A large variation in dietary patterns was observed in the Chinese population as recorded by the national Total Diet Study.[40] The year 2002 was used as a reference year to explore differences in human body burdens with different dietary patterns from TDS surveys and estimated environmental concentrations. All the surveyed locations from the Total Diet Study were assigned into each grid. The average environmental concentration of each grid was used to predict regional human body burden.

2.4.3. Scenarios for Future Trends. In this study, identical dietary patterns were assumed for each cohort, although in reality individuals will have a wide range of dietary preferences. In order to test the influence of different dietary patterns on future exposure trends and to make recommendations on how to maximize the reduction in human body burdens through dietary transitions, future dietary exposure profiles were explored under multiple scenarios defined as 1) Chinese population maintains current dietary patterns until the end of this simulation period (2100); 2) Chinese population follows the dietary pattern as their cohorts from European countries after 2013; 3) Chinese population follows the Chinese Dietary Guidelines suggested by the Chinese Nutrition Society[41] until 2100; 4) Chinese population only eats vegetables; 5) Chinese population adheres to a meat-rich diet; and 6) Chinese population keeps a fish-based diet. Specific values of each dietary scenario are presented in Table S2.

2.4.4. Food Origin Assumptions. The food web bio-accumulation modeling was driven by ambient environmental levels calculated for study regions. Due to the increasing population, domestic food demand is still growing in China,[42] which leads to a limited ability to self-supply. Also, because of domestic food security issues,[43] Chinese residents tend to purchase imported food from developed countries, especially with regards to meat and milk.[42] For example, the import of liquid milk cumulatively rose by 800% in China from 2005 to 2013.[44] Under such circumstances, the potential influence of imported food on human body burden was preliminarily explored by comparing the body burdens in people only eating local food to an extreme scenario of a person exclusively eating imported food. It is difficult to track the detailed origin of all imported food.[45] Here, we tested two scenarios. One scenario is closer to reality, assuming people consuming imported food from several main importers, as identified by national survey data. The fish, meat/vegetables, and dairy products are mainly sourced from Russia (Grid 70),[46] United States of America (Grid 79),[47] and New Zealand (Grid 216).[48] The simulation period started from 2000 to 2100, since food trade is a recent phenomenon. Another is the worst-case scenario, assuming all imported food from a single overseas region with more extensive historical production and use of PCBs (Grid 61, mainly covering southern parts of Scandinavia, Germany, and UK). This region also captures the area for which the ACC-HUMAN model was originally developed, parametrized, and evaluated.[33] The stimulated period covered 1930−2100 for this scenario as an illustrative case study, to explore the impact of imported food on human body burden over the entire chemical life cycle (from production to cessation).

2.4.5. Human Characteristics. Dietary transitions were evaluated by comparing the lipid-normalized body burden of a 30-year old female over time under various dietary transition scenarios. By focusing on a single age group, the influence of longitudinal changes in the body burden of an individual will be eliminated.[49] Chinese women were chosen as the target receptors for the simulations, as most studies did not observe significant gender difference in human body burdens.[50] Following the model defaults and until recently the reality in China, all women were assumed to be the first-born child to a 30-year-old mother who delivered one child at the age of 29. Each child was breastfed for six months as officially suggested.[51] Their whole-body lipid contents were reparametrized based on the Chinese population.[52]

3. RESULTS AND DISCUSSION

3.1. Evaluation with Observations. The body burdens of women living in China were predicted using the BETR-Global and ACC-HUMAN models in sequence, as schematically presented in Figure 1. All results presented are based on predictions from central China (Grid 92) unless specified. In order to build confidence in the model, the predicted concentrations in dietary items and human body from the default emission scenario were compared with measurements from the literature (summarized in Table S3). Observations were mainly selected from the national Total Diet Survey, which represents a general diet pattern across China.[6,53] The predicted concentrations in dietary items and human milk fit well with the estimations. The largest divergence occurred in fish, which was overestimated by up to a factor of 10. It is important to note that the national diet survey detected PCBs in cooked fish following a local recipe.[53] The cooking process, such as baking, broiling, frying, and roasting, could result in PCBs loss,[54] which is not considered in the ACC-HUMAN model. Also, the surveyed dietary items were purchased in local groceries and aggregated as a pooled sample in the market-based study. Large uncertainties exist in terms of their origin, trophic level, and age class. When we look into other measurement studies,[55−58] concentrations of PCB-153 in fish also presented wide geographical variation with more than two orders' difference as in Table S3, and our modeling results are within the reported range.

To our knowledge, there are no studies reporting both dietary profiles and PCB levels in a single population at more than one-time point in China so far. Therefore, it is difficult to evaluate rigorously these predicted trends with historical measurements. In China, two national surveys of POPs in

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human milk have been carried out in 2007[59] and 2011.[60,61] A decline for PCB-153 and an increase for dioxin-like PCBs were observed from 2007 to 2011.[60] Also, an increasing trend of dioxin-like PCBs was observed in Shijiazhuang, a northern city of China, from 2002 to 2007.[62] The human body burden was predicted to decrease from 2010, which is not closely consistent with currently available measurements. However, it is difficult to confirm the specific trend due to the lack of continuous national monitoring and surveillance programs, but the predicted value of human body burden is in an acceptable range as presented in Table S3. In summary, the general trends of PCBs in biota, including human, fish, pig, and vegetables, are consistent with limited monitoring data as discussed using the default scenario, which is used in the following discussions.

3.2. Body Burden versus Age Trends. In order to understand the relationship between age and human body burden based on data modeled at different times, the cross-sectional and longitudinal body burden versus age trends of PCB-153 were calculated and sampled every 10 years from 1960 to 2050 for Chinese women as presented in Figure 1(f) and (g). The relationships between age and human body burden in cross-sectional and longitudinal studies were strongly dependent on the sampling year. During the period with increasing emissions (1930−1970), the cross-sectional human body burden peaked at 10 years old, reflecting the increasing prenatal exposure and relatively low body lipid content at a younger age. For an individual born during this period, the body burden generally increased with age as illustrated in Figure 1(g), which is attributed to rising exposure with increasing emissions. When emissions decreased (1980−2010), the age at which the maximum body burden occurred depends on the length of time after the emission peak. These predictions suggest that the peak age of human body burden occurs at increasingly older ages as time elapses after emissions ceased. For a single person born in this period, the predicted human body burden was highest for a child at age one and reduced substantially due to growth dilution. This trend is consistent with other previous studies.[12,16]

Due to the lack of historical empirical data, it is challenging to confirm the predictions of cross-sectional and longitudinal body burden versus age trends with measurements, particularly for findings before the ban of PCBs (1930−1970). Several cross-sectional studies conducted after the PCB ban have confirmed the significant roles of age, dietary habits, and geographical factors in determining human exposure in China.[6] However, most studies have limited sample sizes and narrow age bands and still did not reach a consistent agreement on the relationship between age and human body burden. For example, Sun et al.[62] and Wang et al.[4] reported that human tissues positively correlated with age, while Kunisue et al.[17] did not find any relationship between age and human body burden.

3.3. Implications for Long-Term Human Exposure. In a dynamic simulation, the predicted exposure of the physical and biotic environment will respond to changes in primary emissions. Since dietary intake is the main exposure pathway for humans exposed to PCBs (spatially and temporally), variable chemical concentrations in food and individual differences in dietary patterns will lead to variable human body burdens.[6] In particular, under nonsteady state emissions, human body burdens will depend on the age when the exposure began to reflect changes in the emission profile.[12]

3.3.1. Historical Exposure Profile. Under the combined effect of changing emissions and cohort dietary transition, the body burden of the Chinese 30-year-old female cohort increased 75-fold over the last 70 years (1940−2010) for PCB-153, despite a 7-fold reduction in Chinese environmental concentrations driven by declining emission from 1975 to 2010. Dietary transition could result in an additional increase in human body burden of more than 2 orders of magnitude during the simulated time, when compared with the test scenario assuming a constant dietary pattern. In addition, the peak time of human body burden is predicted to have occurred in 2010 for a 30-year-old Chinese female cohort, while this took place in 1980 for a Western counterpart (Figure 2). The

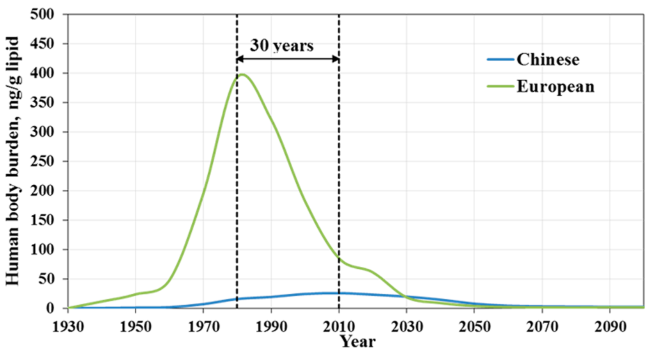

Figure 2. Human body burden (ng g^{-1} lipid) of PCB-153 for a 30-year-old female cohort in central China (Grid 92) and in Europe (Grid 61). Both populations were assumed to only eat locally produced food.

Western temporal trend of human body burden was assumed to be represented by a typical European female following European dietary preferences.[33] The combined effect of changing emission trends and dietary transition resulted in an approximately 30-year difference between the peak of human body burdens in the Chinese and European population. This time lag is attributed to two main factors. One is the fast dietary transition from 1959 to 2010 with rapidly increasing consumption of animal-derived food (milk, meat, and fish) in China. A change in PCB exposure was also observed for Arctic populations when replacing locally sourced traditional food (with high concentrations of PCBs) with imported food.[11] In that case, a 50-fold reduction of PCB concentrations was observed over 40 years.[11] The other reason for the predicted time lag is due to a less steep reduction in primary emissions within China compared to Europe as further discussed in Section 3.3.2.

The European exposure profile closely followed the emission trends, peaking about 10 years after the emissions peak in 1970, which may be interpreted as the time lag required for PCBs to move from the source into the human diet. This could be partly due to their relatively stable diet with only about a 2-fold increase in animal-derived food from the 1960s to 1990s.[63] The cumulative human body burden of 175 ng g^{-1} lipid in the Chinese population was an order of magnitude lower than the Western body burden during the period from 1930 to 2100. However, the difference is mainly associated with historical exposure (1930−2010). During this period, the cumulative body burden accounts for >90% of the total body burden (during 1930−2100) for the Western population, while it only accounts for up to 54% for the Chinese population. From 2030, the Chinese human body burden is predicted to exceed that of Europeans for the first time. Overall, our model predictions indicated that Chinese body burdens are likely to

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remain relatively high for decades to come, due to a combined effect of a slow decline in primary emissions and a dietary transition toward increased intake of rich-lipid food.

3.3.2. Roles of Changing Emission Trends. By running three scenarios (baseline, worst-case, and default) from 1930 to 2100, the contributions of imported e-waste and UP-PCBs from cement kilns, electronic arc furnace-produced steel, and iron sintering to the total human body burden have been estimated for $\sum_7$PCBs (Figure S5). Since the imported e-waste contribution would be expected to vary spatially based on the physical distance from the main e-waste recycling sites (mostly located in the southeast, Grid 116), the northeast (Grid 66) was selected as a background region receiving <5% of the total emission of $\sum_7$PCBs from imported e-waste during 1930–2100. The southeast region (Grid 116) was chosen to represent an e-waste polluted region, receiving more than 40% of the emissions of $\sum_7$PCBs from imported e-waste (1930–2100). These two regions were compared in terms of the individual contribution from the imported e-waste and unintentionally produced emissions.

During the period 1930 to 1990, contributions from imported e-waste and unintentional emissions were negligible. This is because China did not start to import e-waste until 1980, and sources of UP-PCBs were minimal.[64] In terms of the cumulative human body burden for $\sum_7$PCBs from 1930 to 2100, imported e-waste contributed >62% in Grid 116 but only ∼4% in Grid 66. The unintentional sources contributed <1% of $\sum_7$PCBs in both grids. Since year 2000, the contribution of imported e-waste to total human body burdens has become more important (46% in 2000 with an increasing trend over time) in Grid 116 peaking in 2040 when it is predicted to account for >90% of $\sum_7$PCBs. If the exposure from imported e-waste was excluded, the peak of human body burden in Grid 116 would occur in the year 2000 but instead peaks in 2020 with the inclusion of the e-waste import (Figure S5). Consequently, the ongoing importation of e-waste may result in up to a 20-year time lag of the peak human body burden in e-waste recycling areas. However, China has started to ban e-waste import since 2002 and apply stricter control regulations year by year.[65] Future emission scenarios and hence model results will be dictated by the efficiency of these control measures.

3.3.3. Regional Differences in Dietary Exposure in 2002. In the reference year of 2002, the percentage of fish and dairy products contributing to total dietary exposure varied widely, between 1 and 20% and 1 and 33%, respectively. In the western part of China (Grid 61 and 90), dairy accounts for a much higher proportion (33%) than in the other regions. In southeastern parts (Grid 93 and 116), large amounts of fish are consumed (up to 20%) (see Figure S2). As a combined result of environmental concentrations and dietary patterns, the highest human body burden of 29 ng g^{-1} lipid was predicted in 30-year-old females living in Grid 116, mainly covering Guangdong, Fujian, and Hunan provinces. The population living in Central China (Grid 92) had the lowest body burden, equivalent to only a third of that in Grid 116. However, this regional difference in human body burdens is relatively small compared to long-term trends. It should be noted that the spatial resolution of the BETR-Global model is relatively coarse (15° × 15°), and “hot spots” could not be recognized in this study. This may result in missing potentially high-risk regions.

3.3.4. Impact of Food Origin. In the worst-case simulation shown in Figure S3, the accumulative body burden for people only eating imported food was predicted to be four times higher (1930–2100) than for people consuming only locally sourced food. The largest difference occurred in 1980, when the Chinese population only eating imported food had an approximately 7-fold higher human body burden than people only eating local food. This can be attributed to China not starting to manufacture PCBs until 1965, resulting in a relatively low exposure of Chinese people eating locally sourced food. The peak burden occurred in 1990 for people completely relying on imported food, while it was predicted to have occurred in 2010 for people eating local food (Figure S3). Consequently, in the period of high production, populations with a high preference for imported food would receive higher PCB doses than people eating locally produced food. This is a specific finding and is not likely to be true for PCBs, as food was not largely imported until recently and even then was imported from regions with less historical production and use of PCBs, such as New Zealand. This illustrative case study was intended to highlight the potential impact of substance inflow via food importation over the whole chemical life cycle, especially for currently used chemicals with historical production. Under this situation, emission reductions in China alone may not be sufficient to protect human health. As a worst-case, it also provides an important range-finding function, which may be key for other potential POPs with ongoing mass production.

In the realistic scenario, which assumed that people started to eat imported food after the year 2000, there is no significant difference between predicted human body burdens from eating local food and imported food. This is due to the low environmental concentrations both in China and the rest of the world after production bans were introduced. Unintentionally produced PCBs have gradually taken a more important role in China,[23] thus human body burdens would be slightly higher for people eating locally sourced food from 2030; but the unintentional emission of PCBs was only calculated domestically, which may cause potential underestimation for people eating imported food.

3.3.5. Impact of Dietary Pattern on Future Body Burden. Predicted future trends of human body burden in a 30-year-old Chinese female living in Grid 92 who consumes locally produced food with different dietary scenarios from 2020 to 2100 were plotted in Figure S4. Only the vegetable-based diet was expected to rapidly reduce the human body burden, while the fish-based diet represented the highest exposure. The 2020 born cohort mainly eating fish would have around 8 times higher human body burden than those eating mainly vegetables. The elevated human body burden from eating fish reflects bioaccumulation along the aquatic food chain, which is approximately 2 orders of magnitude higher than that in the terrestrial food chain for the same region. The differences between other scenarios were relatively small, varying by less than a factor of 2.

3.4. Uncertainties and Limitations. While insight can be gained through the combined application of fate and bioaccumulation models, substantial uncertainties and data gaps remain. Reproductive behavior was simplified to an initial approximation in this study for a Chinese female cohort giving birth to a child at age 29. This could be modified in future simulations with the consideration of the recently announced two-child policy. The age when giving birth, the number of

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children, and the type of milk (formula or breast milk) are important factors, that will affect the prenatal and postnatal exposure of a child, as well as the cumulative lifetime exposure of the adult.[49] Large uncertainty also exists in the intrinsic elimination parameters (i.e., changes in body weight) and ongoing exposure.[66] The confounding processes of ongoing exposure, changes in body size/composition, and other factors that would also influence human body burden over time will make the intrinsic human elimination half-life of the Chinese population different from that of Western populations. Consequently, this study can only offer a general view of the exposure profile for the Chinese population.

The origin of food consumed in China is difficult to assess at the moment. In this study, it has been demonstrated that food from background sites has a minimal influence on the changes in human body burdens. The gradient between urban and rural regions as well as "hot spots" was outside the scope of this modeling study. However, many studies have reported that PCB levels in food from "hot spots" can be elevated by several orders of magnitude, resulting in high body burdens in local residents, particularly in regions near e-waste cycling sites.[67−72]

3.5. Future Perspectives. This study has combined a complex array of factors which can determine human exposure to PCBs for the Chinese population. It highlighted the role of dietary pattern and two specific emission sources (intentional and unintentional emissions) on the long-term simulation of human exposure. Potential improvements to enhance future predictions of human body burdens could include the following: 1) more detailed information on diet (e.g., the geographical origin of consumed food) and its transition (continued dietary surveys) in target populations; 2) the reproductive behavior (age when giving birth, number of children) in the target population; and 3) applying increased spatially resolved fate/transport data to better distinguish local/remote food as well as gradients between urban and rural areas, particularly focusing on "hot spots". Food preparation and cooking processes may also affect pollutant concentrations in final ready-to-eat food items. Cooking processes have shown to cause losses of >50% of total PCBs via the loss of fat, particularly in high-lipid food items.[54,73] Therefore, identifying scenarios based on different cooking processes could be useful.

PCB-153 was used as an indicator congener here representing very persistent chemicals. Therefore, biotransformation did not play a key role in their fate and bioaccumulation along food chains. Similar simulations could be easily repeated for other well-documented persistent organic contaminants. However, even for such persistent organic contaminants, large variations were still observed for individual congeners with the age-cohort-effect, which has been demonstrated to be significantly influenced by the half-life of target compounds.[12] As a result, for chemicals which are more susceptible to biotransformation, metabolic potential in humans and other biota needs to be accurately parametrized in order to improve predictions.

From a practical standpoint, it could be suggested that Chinese policy-makers go beyond only setting domestic emission goals. In order to maximize the reduction in human exposure to PCBs and other POPs, the best combination of diet pattern, food origin, cooking method, and reproductive strategy could be investigated. In addition, a large-scale national biobank network program, a repository that stores and manages biological samples, would be a valuable asset to facilitate data collection on human contaminant profiles.[74] For instance, cryogenic repositories for biological samples can be used in retrospective and prospective biomonitoring studies.[75]

However, specifically from a global perspective, it is essential to highlight that PCBs do indeed travel around the globe via environmental flows (LRAT), via waste and food, and all these flows are connected and affect exposure trends and patterns, in addition to any human exposure caused by domestic emissions affecting concentrations in both abiotic and biotic environment. Emission reductions in China alone may not be sufficient, but global emission reductions are needed to reduce exposure to the Chinese population and elsewhere. Taken together, the results from this study illustrate that future human exposure is also likely to be dictated by inflows of PCBs via the environment, via waste and food. This, in turn, tracking of food sources alone may not be sufficient. International measures to track and control the movement of PCBs via waste and the environment into China could also play an important role in the reduction of exposure.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b01228.

> Model input data characterizing the properties of individual PCB congeners, Table S1; scenarios of different diet patterns to predict future trends, Table S2; comparison of observed PCB-153 concentrations in different dietary items and the human body, Table S3; defined study region of China together with the BETR-Global grids, Figure S1; regional human body burden for a 30-year-old Chinese female, Figure S2; comparison of human body burden for a series of 30-year-old females, Figure S3; predicted future trends of human body burden in a 30-year-old Chinese female, Figure S4; individual contribution of imported e-waste to the total human burden, Figure S5 (PDF)

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Notes

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