

1 Modelling the time-variant dietary  
2 exposure of PCBs in China over the  
3 period 1930 to 2100.

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**20 Abstract**

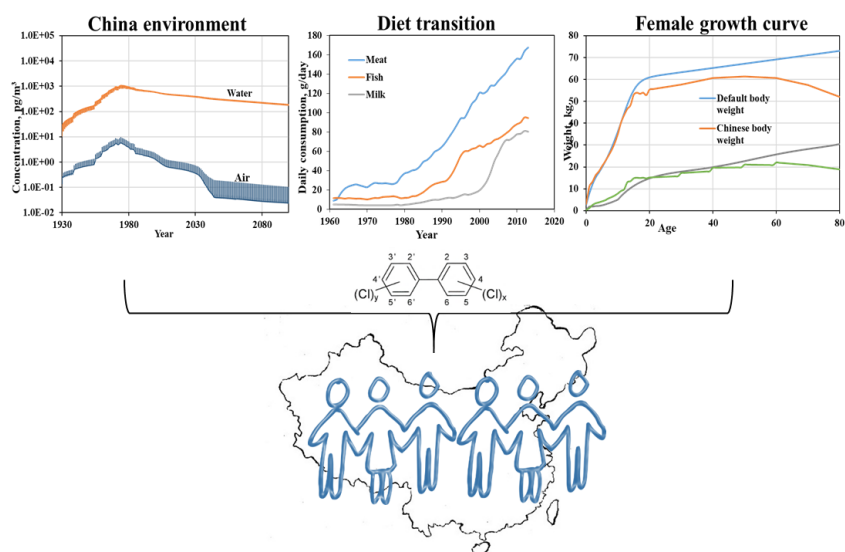
21 This study aimed for the first time to reconstruct historical exposure profiles for PCBs to the  
22 Chinese population, by examining the combined effect of changing temporal emissions and  
23 dietary transition. A long-term (1930-2100) dynamic simulation of human exposure using  
24 realistic emission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

**38 Keywords:**

39 Dietary exposure; polychlorinated biphenyls; human body burden; Chinese population;  
40 multimedia fate model

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## 42 Graphical abstract



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## 45 **1 Introduction**

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

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

64 However, developing a model to dynamically describe relationships between emissions and  
65 human exposure is challenging, given the numerous factors which affect source-exposure  
66 relationships of PCBs. Dietary exposure is an important source of PCBs, accounting for up to 90%  
67 of the total intake, especially for foods of animal-origin rich in lipids.<sup>8</sup> The combined effect of  
68 temporal trends in emissions and dietary preferences are suggested to have a significant effect on  
69 temporal trends in human body burdens.<sup>11</sup> A 6 to 13 fold decrease in PCB-153 body burden was  
70 reported from 1980-2020 due to dietary transition for an Arctic population (e.g., less reliant on  
71 traditional food items with high PCB concentrations such as seal meat<sup>11</sup>). In contrast, the intake

72 of food items with potentially high PCB concentrations have increased in China with 17, 3 and 8  
73 times higher consumption of meat, milk and fish from 1950 to 2013, respectively (FAOSTA:  
74 <http://faostat3.fao.org/home/E>).

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

89 The purposes of this study were therefore: 1) to reconstruct the historical exposure profile and  
90 predict future exposure trends under multiple scenarios for Chinese female cohorts using PCB-  
91 153 as a case study, which can be directly linked to mother-to-fetus transfer; 2) to assess the  
92 combined effect of dietary transition and emission trends on human exposure over the longitudinal  
93 and cross-sectional trends; 3) to explore the impact of different PCB emission sources on human  
94 body burden as predicted by the applied models.

## 95 **2 Methods**

### 96 **2.1 Conceptual approach**

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

## 108 **2.2 Emission scenarios**

109 Several historical PCBs emission scenarios were explored to assess the individual and combined  
110 influence from three sources: (i) intentionally produced PCBs; (ii) e-waste imports; and (iii)  
111 unintentional formation. For the former two sources, global historical emission inventories (1930-  
112 2100) published by Breivik et al.<sup>21, 22</sup> were used. While the “baseline scenario” estimates global  
113 PCB emission without considering transboundary movement of e-waste, the “worst-case scenario”  
114 additionally accounts for emissions associated with imported e-waste from OECD to non-OECD  
115 countries.<sup>21</sup> Emissions from unintentionally-produced PCBs (UP-PCBs), which mainly originate  
116 from industrial thermal sources, have been identified as providing an important contribution to  
117 total PCB emissions in China in near future.<sup>23</sup> Emissions from outside China from this source  
118 category is not considered, due to lacking a global emission inventory for UP-PCBs. The “default  
119 scenario” therefore was defined as total PCBs from intentional production, combined with e-  
120 waste imports and unintentional formation, where the individual influence of PCB emissions from  
121 imported e-waste and unintentional emissions were also evaluated. Each emission scenario was  
122 allocated to a  $1^\circ$  latitude  $\times$   $1^\circ$  longitude grid system based on a global population density  
123 database.<sup>24</sup>

## 124 **2.3 Selected models**

### 125 **2.3.1 Fate model**

126 To predict ambient environmental levels of selected PCB congeners in the global environment  
127 over time, the default scenario as defined in Section 2.2 was used as emission input to the  
128 multimedia fate model BETR-Global.<sup>25, 26</sup> This model has previously been evaluated and  
129 successfully applied to PCBs.<sup>21, 25-27</sup> The study region (covered grid cells assigned numbers of  
130 Grid 66, 69, 90, 91, 92, 93, 115, 116) is illustrated in Figure S1. The BETR-Global model has a  
131 spatial resolution of 15° latitude × 15° longitude, consisting of 288 grid cells. Each of these regions  
132 consists of up to seven bulk compartments, including ocean water, fresh water, upper air, lower  
133 air, soil, freshwater sediments and vegetation. The detailed environmental parameters were  
134 sourced from a wide range of databases and GIS was used to calculate the characteristics of each  
135 region.<sup>28</sup> The model regions are connected by advective transport via air, fresh water and ocean  
136 water. PCBs emissions were allocated to the 288 grid cells. Only emission to lower air was  
137 considered. The initial model concentration was assumed to be zero. This model was run  
138 dynamically for the period from 1930 to 2100. Seven indicator PCBs (PCB-  
139 28,52,101,118,138,153,180) were selected for simulation, although PCB-153 was selected as an  
140 indicator PCB and mainly discussed here. Model input data characterising the properties of  
141 individual PCB congeners was selected from the literature<sup>29-32</sup> and is summarized in Table S1.

### 142 **2.3.2 Bioaccumulation model**

143 Chemical bioaccumulation in food chains was modelled by a mechanistically based, non-steady  
144 state bioaccumulation model (ACC-HUMAN),<sup>33</sup> which has been previously shown to provide  
145 reasonable results for PCB bioaccumulation in the human food chain.<sup>33-35</sup> It is subdivided into an  
146 agricultural and an aquatic food web. The considered uptake pathways of contaminants are diet  
147 and inhalation, while the elimination pathways are metabolism, percutaneous excretion, digestive  
148 tract excretion, exhalation, childbirth and breastfeeding.<sup>33</sup> Since PCBs mainly enter the body via  
149 diet, the inhalation pathway was not discussed here.

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

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

## 163 **2.4 Dietary information for the Chinese population**

### 164 **2.4.1 General diet pattern and transition**

165 Food supply data for domestic consumption from 1959 to 2013  
166 (<http://faostat3.fao.org/browse/FB/CL/>) was used as the default dietary pattern to represent  
167 dietary transition trends at a national level. This was calculated based on the food production plus  
168 imports minus exports. The domestic food supply of meat, milk and fish increased by around  
169 factors of 17, 3 and 8 (illustrated in Figure 1-c), on a national scale during the period 1959-2013.  
170 For the period from 1930 to 1959 without recorded diet information, the dietary pattern was  
171 assumed to be the same as 1959. This is a first approximation to gain a general overview of dietary  
172 transition in China. Potential uncertainties include regional supply variances between different  
173 sub-populations.

174 The default lipid content of human dietary items in ACC-HUMAN were reset to 5.2 % for fish  
175 and 3.2% for milk in Chinese food products.<sup>37</sup> Unlike Western populations, for which ACC-



176 HUMAN was originally developed, pork is the main meat type consumed in China.<sup>38</sup> Thus, the  
177 beef cattle component in ACC-HUMAN was re-parameterized. Chinese pigs are mainly fed on  
178 corn, but their diet may also include discarded food of animal origin, which would potentially  
179 underestimate the contaminant levels in pigs. However, this study was intended to be  
180 representative of generic trophic levels in China and acceptable modelling results are  
181 demonstrated in Section 3.1. Pork contains up to 30% lipid content, highest among varied meat  
182 types.<sup>37</sup> The worst-case scenario, assuming that the Chinese population only eats pork, was also  
183 assessed and modified in ACC-HUMAN model. The dietary transition excluded data for  
184 vegetables, since vegetable consumption has remained relatively stable at around 276 to 310 g  
185 day<sup>-1</sup> per person.<sup>39</sup> Considering the relatively low PCB concentrations in vegetables, it was  
186 assumed that the resulting variation would be minimal.

#### 187 **2.4.2 Regional differences**

188 A large variation in dietary patterns was observed in the Chinese population as recorded by the  
189 national Total Diet Study (TDS).<sup>40</sup> The year 2002 was used as a reference year to explore  
190 differences in human body burdens with different dietary patterns from TDS surveys and  
191 estimated environmental concentrations. All the surveyed locations from the Total Diet Study  
192 were assigned into each grid. The average environmental concentration of each grid was used to  
193 predict regional human body burden.

#### 194 **2.4.3 Scenarios for future trends**

195 In this study, identical dietary patterns were assumed for each cohort, although in reality  
196 individuals will have a wide range of dietary preferences. In order to test the influence of different  
197 dietary patterns on future exposure trends and to make recommendations on how to maximise the  
198 reduction in human body burdens through dietary transitions, future dietary exposure profiles  
199 were explored under multiple scenarios defined as: 1) Chinese population maintains current  
200 dietary patterns until the end of this simulation period (2100); 2) Chinese population follows the  
201 dietary pattern as their cohorts from European countries after 2013; 3) Chinese population follows  
202 the Chinese Dietary Guidelines suggested by the Chinese Nutrition Society<sup>41</sup> until 2100; 4)

203 Chinese population only eats vegetables; 5) Chinese population adheres to a meat-rich diet; 6)  
204 Chinese population keeps a fish-based diet. Specific values of each dietary scenario are presented  
205 in Table S2.

#### 206 **2.4.4 Food origin assumptions**

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

#### 226 **2.4.5 Human characteristics**

227 Dietary transitions were evaluated by comparing the lipid-normalized body burden of a 30-year  
228 old female over time under various dietary transition scenarios. By focusing on a single age group,  
229 the influence of longitudinal changes in the body burden of an individual will be eliminated.<sup>49</sup>

230 Chinese women were chosen as the target receptors for the simulations, as most studies did not  
231 observe significant gender difference in human body burdens.<sup>50</sup> Following the model defaults and  
232 until recently the reality in China, all women were assumed to be the first-born child to a 30-year-  
233 old mother and delivered one child at the age of 29. Each child was breastfed for six months as  
234 officially suggested.<sup>51</sup> Their whole-body lipid contents were re-parametrized based on Chinese  
235 population.<sup>52</sup>

## 236 **3 Results and Discussion**

### 237 **3.1 Evaluation with observations**

238 The body burdens of women living in China were predicted using the BETR-Global and ACC-  
239 HUMAN models in sequence, as schematically presented in Fig 1. All results presented are based  
240 on predictions from central China (Grid 92) unless specified. In order to build confidence in the  
241 model, the predicted concentrations in dietary items and human body from the default emission  
242 scenario were compared with measurements from the literature (summarized in Table S3).  
243 Observations were mainly selected from the national Total Diet Survey (TDS), which represents  
244 a general diet pattern across China.<sup>6, 53</sup> The predicted concentrations in dietary items and human  
245 milk fit well with the estimations. The largest divergence occurred in fish, which was  
246 overestimated by up to a factor of 10. It is important to note that the national diet survey detected  
247 PCBs in cooked fish following a local recipe.<sup>53</sup> The cooking process, such as baking, broiling,  
248 frying and roasting, could result in PCBs loss,<sup>54</sup> which is not considered in the ACC-HUMAN  
249 model. Also, the surveyed dietary items were purchased in local groceries and aggregated as a  
250 pooled sample in the market-based study, large uncertainties exist in terms of their origin, trophic  
251 level and age class. When we look into other measurement studies,<sup>55-58</sup> Concentrations of PCB-  
252 153 in fish also presented wide geographical variation with more than two orders' difference as  
253 in Table S3, and our modelling results are within the reported range.

254 To our knowledge, there are no studies reporting both dietary profiles and PCB levels in a single  
255 population at more than one-time point in China so far. Therefore, it is difficult to evaluate  
256 rigorously these predicted trends with historical measurements. In China, two national surveys of  
257 POPs in human milk has been carried out in 2007<sup>59</sup> and 2011.<sup>60, 61</sup> A decline for PCB-153 and a  
258 increase was observed for dioxin-like PCBs from 2007 to 2011.<sup>60</sup> Also, an increasing trend of  
259 dioxin-like PCBs was observed in Shijiazhuang, a northern city of China, from 2002 to 2007.<sup>62</sup>  
260 The human body burden was predicted to decrease from 2010, which is not closely consistent  
261 with currently available measurements. However, it is difficult to confirm the specific trend due  
262 to the lack of continuous national monitoring and surveillance programs. But the predicted value  
263 of human body burden is in an acceptable range as presented in Table S3. In summary, the general  
264 trends of PCBs in biota, including human, fish, pig and vegetables, are consistent with limited  
265 monitoring data as discussed using the default scenario, which is used in the following discussions.

### 266 **3.2 Body burden versus age trends**

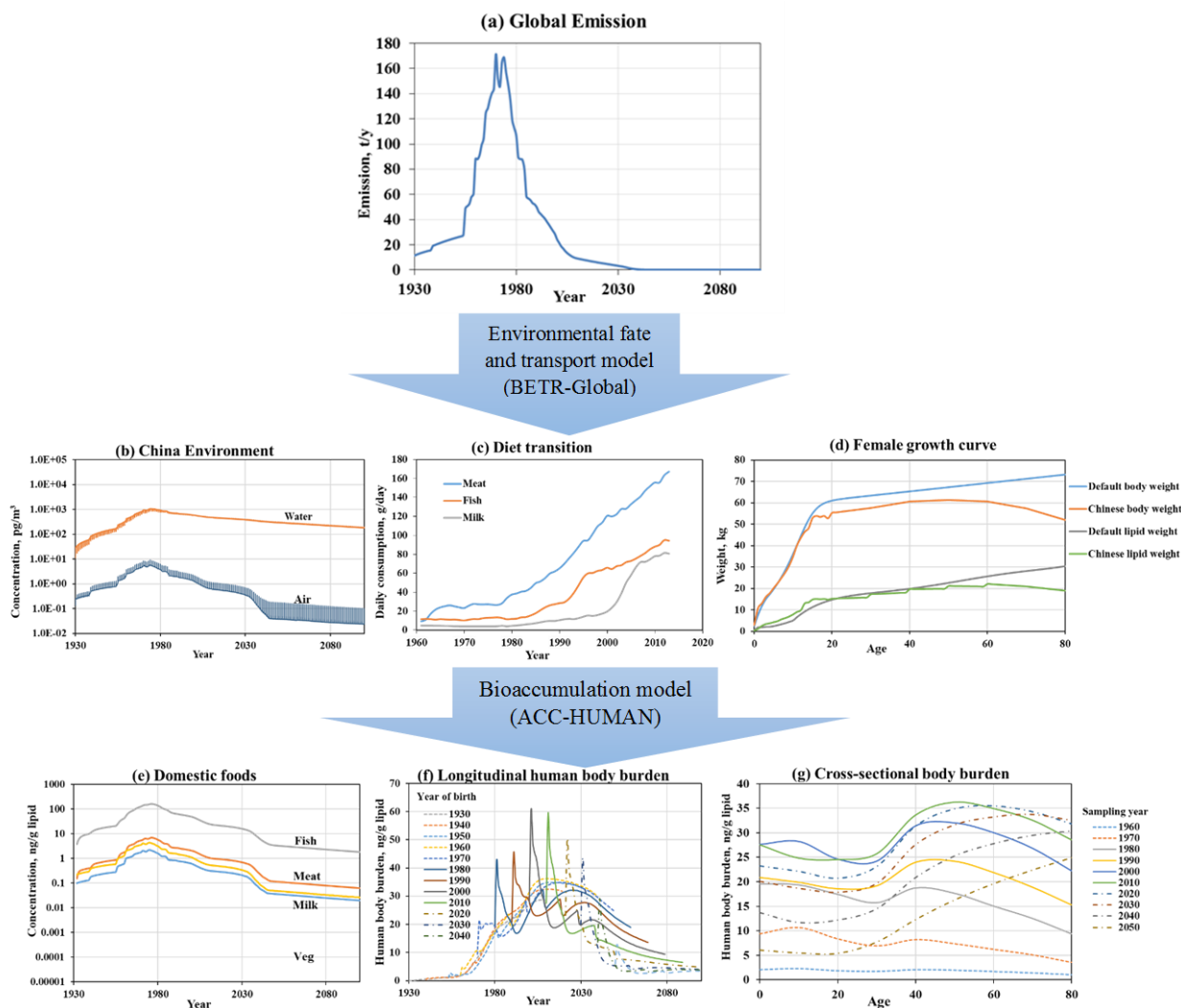
267 In order to understand the relationship between age and human body burden based on data  
268 modelled at different times, the cross-sectional and longitudinal body burden versus age trends  
269 of PCB-153 were calculated and sampled every 10 years from 1960 to 2050 for Chinese women  
270 as presented in Figure-1 (f) and (g). The relationships between age and human body burden in  
271 cross-sectional and longitudinal studies were strongly dependent on the sampling year. During  
272 the period with increasing emissions (1930-1970), the cross-sectional human body burden peaked  
273 at 10 years old, reflecting the increasing prenatal exposure and relatively low body lipid content  
274 at a younger age. For an individual born during this period, the body burden generally increased  
275 with age as illustrated in **Error! Reference source not found.**(g), which is attributed to rising  
276 exposure with increasing emissions. When emissions decreased (1980-2010), the age at which  
277 the maximum body burden occurred depends on the length of time after the emission peak. These  
278 predictions suggest that the peak age of human body burden occurs at increasingly older ages as  
279 time elapses after emissions ceased. For a single person born in this period, the predicted human

280 body burden was highest for a child at age one and reduced substantially due to growth dilution.  
281 This trend is consistent with other previous studies.<sup>12, 16</sup>

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

### 291 **3.3 Implications for long-term human exposure**

292 In a dynamic simulation, the predicted exposure of the physical and biotic environment will  
293 respond to changes in primary emissions. Since dietary intake is the main exposure pathway for  
294 humans exposed to PCBs, (spatially and temporally) variable chemical concentrations in food and  
295 individual differences in dietary patterns will lead to variable human body burdens.<sup>6</sup> In particular,  
296 under non-steady state emissions, human body burdens will depend on the age when the exposure  
297 began to reflect changes in the emission profile.<sup>12</sup>



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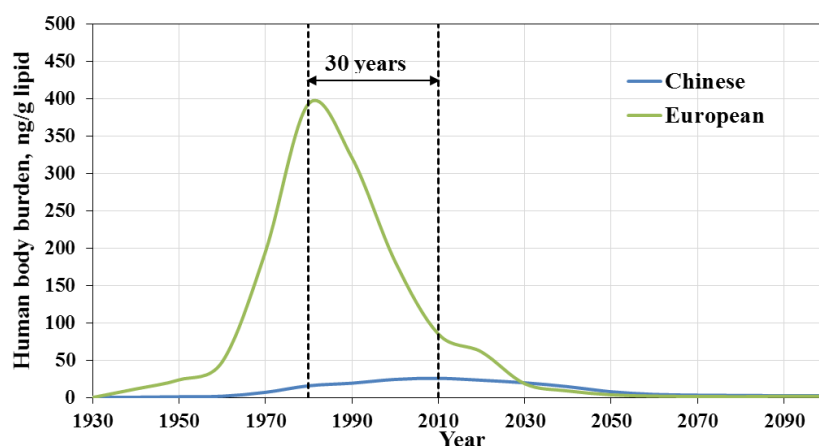
Figure 1. Schematic overview of the modelling approach employed to assess the combined effect of emission trends and dietary transition on human exposure to PCB-153 for Chinese female population. The approach was modified after Quinn et al.<sup>11</sup> The global emission estimate of PCB-153 over the 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 on different years (f). The cross-sectional versus age dependence was modelled 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 modelling results after the ban of all intentional emissions defined in Section 2.2 (2020-2050).

### 312 3.3.1 Historical exposure profile

313 Under the combined effect of changing emissions and cohort dietary transition, the body burden  
314 of the Chinese 30-year-old female cohort increased 75-fold over the last 70 years (1940-2010) for  
315 PCB-153, despite a 7-fold reduction in Chinese environmental concentrations driven by declining  
316 emission from 1975 to 2010. Dietary transition could result in an additional increase in human  
317 body burden of more than two orders of magnitude during the simulated time, when compared  
318 with the test scenario assuming a constant dietary pattern. In addition, the peak time of human  
319 body burden is predicted to have occurred in 2010 for a 30-year-old Chinese female cohort, while  
320 this took place in 1980 for a Western counterpart (Figure 2). The Western temporal trend of  
321 human body burden was assumed to be represented by a typical European female following  
322 European dietary preferences.<sup>33</sup> The combined effect of changing emission trends and dietary  
323 transition resulted in an approximately 30-year difference between the peak of human body  
324 burdens in the Chinese and European population. This time-lag is attributed to two main factors.  
325 One is the fast dietary transition from 1959-2010 with rapidly increasing consumption of animal-  
326 derived food (milk, meat and fish) in China. A change in PCB exposure was also observed for  
327 Arctic populations when replacing locally-sourced traditional food (with high concentrations of  
328 PCBs) with imported food.<sup>11</sup> In that case, a 50-fold reduction of PCB concentrations was observed  
329 over 40 years.<sup>11</sup> The other reason for the predicted time-lag is due to a less steep reduction in  
330 primary emissions within China compared to Europe as further discussed in section 3.4.2.

331 The European exposure profile closely followed the emission trends, peaking about 10 years after  
332 the emissions peak in 1970, which may be interpreted as the time-lag required for PCBs to move  
333 from the source into the human diet. This could be partly due to their relatively stable diet with  
334 only about a two-fold increase in animal-derived food from the 1960s to 1990s.<sup>63</sup> The cumulative  
335 human body burden of 175 ng g<sup>-1</sup> lipid in Chinese population was an order of magnitude lower  
336 than the Western body burden during the period from 1930 to 2100. However, the difference is  
337 mainly associated with historical exposure (1930-2010). During this period, the cumulative body  
338 burden accounts for >90% of the total body burden (during 1930-2100) for the Western

339 population while it only accounts for up to 54% for the Chinese population. From 2030, the  
 340 Chinese human body burden is predicted to exceed that of Europeans for the first time. Overall,  
 341 our model predictions indicated that Chinese body burdens are likely to remain relatively high for  
 342 decades to come, due to a combined effect of a slow decline in primary emissions and a dietary  
 343 transition towards increased intake of rich-lipid food.



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345 Figure 2. The human body burden (ng g<sup>-1</sup> lipid) of PCB-153 for a 30-year-old female cohort in  
 346 central China (Grid 92) and in Europe (Grid 61). Both populations were assumed to only eat  
 347 locally produced food.

### 348 3.3.2 Roles of changing emission trends

349 By running three scenarios (baseline, worst-case and default) from 1930-2100, the contributions  
 350 of imported e-waste and UP-PCBs from cement kilns, electronic arc furnace-produced steel and  
 351 iron sintering to the total human body burden have been estimated for  $\Sigma_7$ PCBs (Figure S5). Since  
 352 the imported e-waste contribution would be expected to vary spatially based on the physical  
 353 distance from the main e-waste recycling sites (mostly located in the southeast, Grid 116), the  
 354 northeast (Grid 66) was selected as a background region receiving <5% of the total emission of  
 355  $\Sigma_7$ PCBs from imported e-waste during 1930-2100. The southeast region (Grid 116) was chosen  
 356 to represent an e-waste polluted region, receiving more than 40% of the emissions of  $\Sigma_7$ PCBs  
 357 from imported e-waste (1930-2100). These two regions were compared in terms of the individual  
 358 contribution from the imported e-waste and unintentionally produced emissions.



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

### 373 **3.3.3 Regional differences in dietary exposure in 2002**

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

### 386 3.3.4 Impact of food origin

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

404 In the realistic scenario, which assumed that people started to eat imported food after the year  
405 2000, there is no significant difference between predicted human body burdens from eating local  
406 food and imported food. This is due to the low environmental concentrations both in China and  
407 the rest of the world after production bans were introduced. Unintentionally-produced PCBs have  
408 gradually taken a more important role in China,<sup>23</sup> thus human body burdens would be slightly  
409 higher for people eating locally sourced food up to 2030. But the unintentional emission of PCBs  
410 was only calculated domestically, which may cause potential underestimation for people eating  
411 imported food.

### 412 3.3.5 Impact of dietary pattern on future body burden

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

### 422 **3.4 Uncertainties and limitations**

423 While insight can be gained through the combined application of fate and bioaccumulation models,  
424 substantial uncertainties and data gaps remain. Reproductive behaviour was simplified to an  
425 initial approximation in this study for a Chinese female cohort giving birth to a child at age 29.  
426 This could be modified in future simulations with the consideration of recently announced two-  
427 child policy. The age when giving birth, the number of children and the type of milk (formula or  
428 breast milk) are important factors, that will affect the prenatal and postnatal exposure of a child,  
429 as well as the cumulative lifetime exposure of the adult.<sup>49</sup> Large uncertainty also exists in the  
430 intrinsic elimination parameters (i.e., changes in body weight) and ongoing exposure.<sup>66</sup> The  
431 confounding processes of on-going exposure, changes in body size/composition and other factors  
432 that would also influence human body burden over time, will make the intrinsic human  
433 elimination half-life of the Chinese population different from that of Western populations.  
434 Consequently, this study can only offer a general view of the exposure profile for the Chinese  
435 population.

436 The origin of food consumed in China is difficult to assess at the moment. In this study, it has  
437 been demonstrated that food from background sites has a minimal influence on the changes in  
438 human body burdens. The gradient between urban and rural regions as well as ‘hot spots’ was  
439 outside the scope of this modelling study. However, many studies have reported that PCB levels

440 in food from 'hot spots' can be elevated by several orders of magnitude, resulting in high body  
441 burdens in local residents, particularly in regions near e-waste cycling sites.<sup>67-72</sup>

### 442 **3.5 Future perspectives**

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

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

464 From a practical standpoint, it could be suggested that Chinese policy-makers go beyond only  
465 setting domestic emission goals. In order to maximise the reduction in human exposure to PCBs  
466 and other POPs, the best combination of diet pattern, food origin, cooking method, reproductive

467 strategy could be investigated. In addition, a large-scale national biobank network program, a  
468 repository that stores and manages biological samples, would be a valuable asset to facilitate data  
469 collection on human contaminant profiles.<sup>74</sup> For instance, cryogenic repositories for biological  
470 samples can be used in retrospective and prospective biomonitoring studies.<sup>75</sup>

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

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