Who in the world is most exposed to polychlorinated biphenyls? Using models to identify highly exposed populations

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Abstract

Human subpopulations experience different exposure to persistent organic pollutants (POPs) because of differences in the structure of their food webs and the extent of environmental contamination. Here we quantify the time-variant exposure of different human populations around the world to one representative POP, namely the polychlorinated biphenyl (PCB) congener 153, based on a dynamic simulation of both global environmental fate (using the model BETR-Global) and human food chain bioaccumulation (using the model ACC-HUMAN). The approach identifies subpopulations whose diets include a carnivorous mammal as experiencing the world’s highest PCB-153 exposure, i.e. the very large biomagnification potential of their food web more than makes up for the remoteness of their living environment. However, for subpopulations that do not eat warm-blooded carnivores, the proximity to sources of PCBs is more important than food web structure and environmental conditions for differentiating their exposure to PCBs.

1. Introduction

Chemical risk assessment and regulation for human health aims to protect the most vulnerable members of the population. Most commonly, these include pregnant women and women of child bearing age, infants, and children because of the potential susceptibility of the developing foetus and growing child to chemicals. The most vulnerable sometimes also comprise the elderly and those with compromised health, because they may often be less well equipped to cope with the effects of chemicals. For substances that due to their persistence and long-range transport potential have the ability to reach people in many parts of the world, it is of interest not only to recognize the most vulnerable within a population, but also to identify the most vulnerable populations globally. While there may on occasion be genetic differences between different human populations affecting the toxicokinetics (i.e. absorption, distribution, metabolism, and excretion) of chemicals, population differences in chemical vulnerability are more likely dominated by differences in exposure. Studying vulnerability thus comprises the identification of those populations worldwide who are most likely to suffer high exposure to a substance because of where they live and what they eat.

Different human populations differ in their exposure susceptibility to chemicals because they differ in terms of their living environment, lifestyle and dietary habits [1]. The latter is particularly important for bioaccumulating substances. We previously applied a modelling approach to quantify and compare the relative exposure susceptibility of a select number of human populations to persistent organic pollutants (POPs) with a wide range of partitioning properties [1]. Combined environmental fate and human food chain bioaccumulation simulations were performed for human populations residing in, and eating exclusively
food regionally produced in, temperate Europe, the Arctic, a tropical island, a tropical forest, a cold steppe, and a warm steppe. A unit emission rate of the chemical to the environment was assumed in each case and relative exposure susceptibility was evaluated by comparing concentrations in humans living in the different environments. We noted that the exposure susceptibility to persistent chemicals of those whose diet includes food derived from warm-blooded carnivores (e.g. marine mammals) can exceed that of a temperate European reference population by orders of magnitude.

Chemicals are emitted into different environments at widely different rates, and the exposure of a population will depend on the rate of emission to that population’s living environment. For populations living in regions with minor local emissions, exposure may further depend on the extent to which chemicals are transported from elsewhere, particularly for chemicals that are not rapidly degraded in the environment. When exposure to the chemicals is primarily via food, then the proximity of food producing regions to emissions can be expected to affect exposure. MacLeod et al [2] illustrated these relationships in a modelling study in which they ran hypothetical scenarios in which the same quantity of a chemical was emitted in different regions of North America and the resulting human exposure of the total North American population was calculated. For benzene, a rapidly degraded chemical for which inhalation is the primary vector to humans, exposure of the total population was greatest when the chemical was emitted in a region with a large local population. For tetrachloromethane, a more persistent volatile chemical, exposure was greatest when it was emitted in a region that was upwind of a region with a large local population. For benzo[a]pyrene and TCDD, two chemicals for which food is the primary vector to humans, exposure was greatest when the chemical was emitted in a region with a major food producing region.

To fully account for differences in the exposure susceptibility to bioaccumulating substances between populations globally, it is thus necessary to complement information on food web properties and dietary habits with a consideration of spatially explicit emissions and global long-range transport processes. Work to date has generally focused on one of these dimensions and utilized hypothetical scenarios. With the development of more powerful modelling tools and spatially explicit emissions estimates, it is now possible to do realistic simulations that combine global scale emissions and fate with local/regional food web properties.

Here we quantify and compare the exposure susceptibility of different human populations to one substance by combining the human food chain calculations of Undeman et al [1] with a global fate and transport calculation. We locate the human subpopulations studied by Undeman et al [1] within the global environment, the spatial and temporal contamination of which is simulated using historical global emissions and the BETR-Global model (figure 1). These sub-populations have distinct and extreme, yet plausible diets (i.e. they are not representative of the average population in the region to which they are assigned), in accordance with our goal of identifying the most highly exposed subpopulations globally. This approach is applied to polychlorinated biphenyl (PCB) congener 153, an important POP with a well-established global emission history, whose global fate and transport has previously been simulated successfully [3].

While our treatment of emissions, environmental fate and bioaccumulation is realistic, our treatment of food sourcing remains hypothetical, as we were unable to find global scale data on the sourcing of feed and food for specific populations. Local food sourcing is not an unreasonable assumption for many populations globally, especially historically, but global trade in food leads to populations with diets that are increasingly sourced elsewhere [4, 5]. We therefore present simulations with different food sourcing assumptions. First we compare populations that eat exclusively locally produced food, but vary in terms of both their dietary habits and the distance of the living environment from PCB sources. In a second step we compare Swedes who eat fish harvested either internationally or locally from the Baltic Sea. Conceptually, this comparison involves a difference in the distance of the food-sourcing regions from emissions. We relate this to a study comparing Inuit who eat traditional food sourced locally with those who eat food imported from the South. Conceptually, this not only corresponds to a change in the distance of the food-sourcing regions from emissions, but also to a change in the food chain/dietary habits. We use these comparisons to identify the populations that are most highly exposed to PCB-153 and to explore how dietary habits and food web structure, proximity to sources, environmental properties, food sourcing and chemical properties influence this exposure.

2. Results

Contamination of the physical environment. Concentrations of PCB-153 calculated for surface air and seawater of the BETR-Global cells hosting the populations (identified in figure 1) are displayed for the time period 1930–2010 in figure 2. For a more quantitative comparison, table 1 lists cumulative emissions and average contamination in air and sea water for the time period 1930–2010, as well as their value relative to that in cell 62 hosting the Southern Swedish population. The contamination of air and water is strongly related to proximity to sources. The cell representing NE Europe had the highest calculated concentrations, because of its close proximity to major PCB use areas in Europe. Air contamination in the Sahel is a factor of three lower, but is still quite substantial, because of
proximity to European and Nigerian PCB emissions. Concentrations in air in the Brazilian Amazon, Eastern Indian Ocean and Central Asia are a factor of 10–20 times lower than in NE Europe reflecting their relative remoteness from sources, and concentrations in Arctic air are 60 times lower than in NE Europe. When assessing those concentration differences it is important to keep in mind that they refer to the average conditions within fairly large global regions (15° by 15°); PCB concentrations measured close to sources will be much higher than what is calculated for cell 62, but will not be reflective of the air contamination relevant for uptake in human food chains on a regional scale (i.e. above agricultural areas and above fished waters). The sequence of seawater contamination is somewhat different from that in air, but also reflects the influence of the proximity to emissions: While NE Europe still displays the highest PCB-153 concentrations, concentrations in Arctic sea water are calculated to be 20 times lower. The reason that the global concentration gradient in seawater is smaller than in air is due to the lower temperatures in the Arctic favouring partitioning from the atmosphere into the ocean. The equatorial waters in Brazil and Indonesia have concentrations about two orders of magnitude lower than those in NE Europe.

Who in the world is most exposed to PCBs? The concentrations of PCB-153 at birth and the cumulative

Figure 1. PCB-153 exposure in typical representatives of six human subpopulation with different dietary habits (illustrated with pictograms) is calculated by using the time-variant air and water contamination calculated for a grid cell of the global fate and transport model BETR-Global as the input to a calculation by the human food chain model ACC-HUMAN. Red shading shows the spatial distribution of the cumulative, historical emissions of PCB-153 from 1930–2010 for each of the 288 BETR-Global gridcells. Gridcells representing the environment of the selected human populations are indicated in blue and with a number. For the copyright status of each image in this figure, please see footnotes 6–18.
Table 1. Comparison of emissions and concentrations of PCB-153 predicted by the BETR-Global model for the selected regions.

| Geographic region          | Cell# | Host to:                       | Emissions[a] | avg. C absorbing[b] | avg. C water[c] |
|----------------------------|-------|--------------------------------|--------------|---------------------|-----------------|
| NE Europe                  | 62    | Swede, Baltic fishermen        | 142763       | 7.71                | 5.39            |
| Central Asia               | 68    | Mongolian herder               | 4421         | 0.031 (32)          | 0.06            |
| Sahel                      | 109   | Sahel herder                   | 2948         | 0.021 (48)          | 0.38            |
| Brazilian Amazon           | 153   | Amazon fisherman               | 8337         | 0.058 (17)          | 0.10            |
| Eastern Indian Ocean       | 163   | Indonesian fisherman           | 3414         | 0.024 (42)          | 0.05            |
| Canadian Arctic Archipelago| 31    | Canadian Inuit                 | 64           | 0.0005 (2231)       | 0.13            |
| International Fishing Grounds | 14, 15, 37, 39, 151, 176, 62 | Modern Swede                 | n.a.         | n.a.                | n.a.            |

Notes:
- [a] Grid cell number in the BETR-Global model.
- [b] Cumulative emissions to grid cell over time period 1930–2090 in units of kg.
- [c] Concentration in the lowest air compartment of the grid cell averaged over the period 1930–2090 in units of pg m$^{-3}$.
- [d] Concentration in the sea water compartment of the grid cell averaged over the period 1930–2090 in units of pg L$^{-1}$.
- [e] Relative to NE-Europe (factor difference within parentheses).
- [f] For the Amazon fishermen, fresh water was used instead of sea water to indicate the contamination of the aquatic food chain.
- [g] Average of multiple cells, weighted based on the contribution to internationally sourced fish sold in Sweden.
- [h] Not applicable.
- [i] Average C$_{water}$ of international fishing grounds relative to C$_{water}$ of NE Europe between years 1980–2010 is 0.7 and this ratio is 0.1 in the year 2010 when locally sourced fish in the modern diet was phased out.

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lifetime exposure of women representing different birth cohorts, as calculated by the human food chain bioaccumulation model ACC-HUMAN when fed with the air and water concentrations generated by BETR-Global, are displayed as a function of birth year in figure 3. Table 2 gives the maximum values for those two exposure metrics, as well as their value relative to that of the Southern Swedish reference population. The relative calculated PCB contamination among the subpopulation is the same irrespective which exposure metric is used and which birth cohort is considered: Canadian Inuit >> Baltic fisherman > Southern Swede >> Mongolian herder > Sahelian Steppe > Brazilian fisherman > Indonesian fisherman. Consistent with earlier studies, [6] those born in the 1950s and 1960s are expected to have suffered the highest lifetime exposure to PCBs, whereas it is their children, i.e. those born in the 1980s, who experienced the highest perinatal exposure. Similar trends of exposure with birth year in the different investigated populations (figure 3) reflect that (i) the time trends in the PCB emissions affecting the different populations are not substantially different, e.g. with respect to the timing of peak emissions and (ii) the process of long range transport and accumulation through the food chain does not introduce a sufficient lag for the more remote population to experience a delayed exposure peak.

The exposure differences between different populations are fairly similar between different cohorts, i.e. lines in figure 3 remain largely equidistant, although the differences between the modern Swede and the reference population is getting more pronounced for the most recent cohorts.

The results that Inuit feeding on marine mammals are most exposed to PCB-153 is in agreement with empirical observations. The mean concentration of PCB-153 in blood collected during 2005–2010 from Inuit from villages in Greenland where the traditional diet still plays an important role was 840 ng g$^{-1}$ lipid [7]. Lipid-normalized concentrations in blood are similar to lipid-normalized concentrations in breast milk, [8] which are more readily available. For comparison, the concentrations in pooled human milk samples from 56 countries analyzed in the context of the WHO/UNEP Global Monitoring Plan ranged from 0.76–260 ng g$^{-1}$ lipid, [9] and in a recent global review of PCBs in human milk encompassing 116 studies the highest mean concentration of PCB-153 (excluding populations living adjacent to major PCB point sources) was 480 ng g$^{-1}$ lipid [10]. To our knowledge comparable or greater concentrations than those measured in Inuit have only been measured in people living close to major PCB point sources, [11] who have been subject to occupational exposure, or who have been poisoned [12, 13]. The good agreement between model and measurements indicates that the model is useful for identifying highly exposed populations. In addition, it offers novel opportunities to study the factors determining human exposure to POPs.

The importance of marine mammal consumption. The high exposure calculated for the Canadian Inuit indicates that very high exposure susceptibility can overcome dilution caused by distance. Even though the average contamination of Arctic air and seawater is about 60 and 20 times lower than in NE-Europe respectively, the Inuit exposure is predicted to be 6–8 times higher than that of the Swede. This is a testament to the remarkable magnification potential of a human food chain that includes warm-blooded carnivores—we had previously estimated that the exposure susceptibility of the Inuit can exceed that of Southern Swedes by as much as three orders of magnitude [1]. In the case of PCB-153 we estimate an ESI of 120 for the Canadian Inuit, i.e. given identical emission rates into their local environment, we would expect Inuit to achieve concentrations that are 120 times higher than those of the Southern Swedish reference population. The ESI of 120 is indeed quite similar to the combined difference in the concentrations in water (20 x lower in Arctic) and in humans (6–8 x higher in Inuit).

The differences in the ESI towards PCB-153 among the populations not eating carnivorous mammals are relatively small, ranging from 0.27–2.35 for the Amazon and Baltic fishermen, respectively, i.e. they deviate by at most a factor of four from the Southern Swedish reference population (the ESI in the Sahel is an exception). Consequently, the calculated exposure of the populations in the South (Amazon, Sahel, Mongolia, Indonesia) to PCB-153 is relatively even lower than might be expected from the contamination of their physical environment. Among those without

![Figure 3. The concentration of PCB-153 at birth (top) and cumulative life-time exposure to PCB-153 (bottom) for different female birth cohorts from eight global human subpopulations calculated with the BETR Global and ACC-HUMAN model. Table 2 indicates the place of residence and the dietary habit of these populations.](image-url)
remote areas, where contamination of the environments whose diet includes marine mammals live in and who live close to sources. A fortunate historical population of individuals whose diet includes marine mammals feature prominently in studies of health effects of PCBs in humans, presumably because such populations eat either fish or meat and dairy products from grazing terrestrial animals, indicating that such large exposure differences cannot be explained by differences in diet. While the exposure susceptibility of those populations for a compound with the properties of PCB-153 is 3–4 times lower than those for the Southern Swedes [1], the large difference in calculated PCB concentrations is due to the diet of the Southern Swedish population being generated in an environment that is more contaminated with PCBs due to the proximity to sources.

In order to illustrate the influence of long-range transport, we performed calculations for two additional indigenous populations eating marine mammals from Arctic shelf seas, namely those residing in BETRGlobal cells 27 (Eastern Alaska) and 40 (Siberian coast South of Novaya Zemlya) (see supplementary figure S1 available at stacks.iop.org/ERL/13/064036/mmedia).
These cells comprise similar subarctic environments with comparable climate and received very similar estimated, cumulative emissions of PCB-153 of 2417 and 2529 kg, respectively. The maximum concentration at birth calculated for the 1980 and 1990 cohorts is an order of magnitude higher for the Siberian population (6000 ng g\(^{-1}\) lipid) than that for the Eastern Alaskan population (500 ng g\(^{-1}\) lipid). The reason is that Siberia is downwind from the PCB emissions in Europe and Russia, resulting in higher concentrations in water in the Barents Sea than the Beaufort Sea. This shows that in areas with low local emission rates, the extent of long range transport can strongly influence, if not control, PCB exposure. Ultimately, this is the motivation for the international regulation of POPs such as PCBs, as it implies that reducing exposure is often not feasible through national efforts.

In other words, source proximity and long-range transport need to be taken into account in addition to dietary habits when comparing the exposure of different human sub-populations to persistent organic pollutants. A worldwide survey of concentrations in human breast milk [23] identifies exclusively central and Eastern European countries as having elevated PCB concentrations, consistent with the intensity of PCB emissions in this region (figure 1). Concentrations in breast milk in countries in tropical latitudes and the Southern hemispheric are generally much lower, again reflective of the global distribution of historical PCB emissions.

The role of environmental factors. For populations distant from sources, environmental factors can have some influence on exposure. The cumulative PCB exposure estimated for the Mongolian herder is slightly higher than that of the Sahelian herder, both eating identical diets, even though the PCB contamination in the atmosphere in sub-Saharan Africa is more than 6 times that estimated for Central Asia. However, the low concentration in Asian air is compensated by lower temperatures, which result in higher concentrations in soil and grass than in Africa, and the concentrations in the milk and beef cattle are therefore similar in the two regions. The higher human PCB exposure estimated for the Brazilian fisherman relative to the Indonesian fisherman is consistent with concentrations in water that are calculated to be higher for the Amazon than for the Eastern Indian Ocean. Although environmental properties can have some influence on exposure, it is minor compared to the influence of proximity to sources.

The role of the place of origin of food. The assumption of a locally sourced diet is no longer realistic for many human populations, especially in industrialized countries. Increasingly globalized food production means that humans eat animal lipids from terrestrial animals and fish that are often grown or caught in regions very different from their place of residence. Marek et al [24] for example noted similar dietary exposure to PCBs of children living in rural Iowa and urban Chicago, consistent with a nationally integrated food production system in the US. In particular, fish consumed in industrialized countries is now rarely sourced locally. We compared the PCB exposure of a traditional Southern Swedish consumer who continued to eat cod and herring locally caught in the Baltic Sea, with a ‘modern’ Swede who eats fish increasingly from a variety of international sources, including aquaculture. Our calculations predict slightly lower PCB exposure for the modern Swede and increasingly so over time (figure 3). By 2010 the prenatal concentration is predicted to be 1.3 times lower for the modern Swede than that for the traditional Swede. This suggests that PCB exposure of the Swedish population would be higher today if the transition from Baltic fish to internationally sourced fish had not occurred.

The simulated transition from locally to globally sourced fish in Sweden is assumed to have occurred gradually between 1980 and 2010, i.e. during the period of major commercial expansion of aquaculture [25]. The contamination of the international waters producing herring (North Atlantic), cod (Barents Sea) and fish feed for the Norwegian fish farms producing salmon (weighted average Iceland, Peru, Chile, Barents Sea, North Atlantic) for the Swedish market is on average lower by a factor of 5, 6 and 17 than that of the Baltic Sea during 1980–2010. In addition, over time the fraction of proteins and lipids of vegetative origin has increased in fish feed [26] (in our simulations assumed to have negligible concentrations of PCB-153), also lowering the trophic level of farmed fish. At the same time, the lower exposure of farmed and imported wild fish is partly compensated by a higher lipid content of the farmed fish (salmon) in the modern diet. As fish accounts for on average half of the exposure of the traditional Swede during the last decades of the 20th century, the transition to a less contaminated source of fish leads over time to a marked decrease in exposure. It is easy to envisage dietary choices where the origin of food could have an even more profound influence on PCB exposure. For example, a Swedish individual eating primarily meat and fish produced and caught in the southern hemisphere [27] can be expected to have lower PCB exposure than someone who eats the same amounts of dietary lipid sourced in the North.

The role of dietary transitions. The above example relates to a dietary shift from locally toward internationally sourced food, but did not involve a major change in the type of food being consumed (It did involve a change in the type of fish consumed and the trophic level of that fish). Two kinds of dietary transition can have a large impact on human exposure: a major change in the quantity of marine mammal lipid, or a major change in the quantity of other animal lipids in the diet. A Swedish vegetarian with small quantities of animal lipids in the diet has markedly lower exposure susceptibility than the reference Swede on a mixed diet [1]. A particularly complex dietary shift is currently occurring in many Inuit communities, where
younger generations are eating increasingly less traditional food (including marine mammals) and more food imported from the South. This shift thus involves both a change in the place of origin of the food and a major change in the properties of the food web. Specifically, the shift is from food caught in a clean environment to food that is produced in one that is more contaminated. It is also a shift from a diet that includes marine mammals to one that does not. Quinn et al [28] and Binnington et al [29] have used a similar modelling approach to the one adopted here to explore the effect in this dietary shift on PCB exposure. The effect of the change in food web structure is larger than the effect of the proximity of the food source region to PCB sources, i.e. it is expected to result in notable lowering of the exposure to PCBs.

This approach could similarly be applied to quantify the impact of other population-level dietary shifts on PCB exposure. Examples include decreasing fish consumption among Northern Europeans or increasing meat consumption in India and China [30]. These changes are expected to diminish differences in POP exposure between human populations worldwide.

The role of chemical properties. The analysis here is restricted to one particular PCB. In order to assess the extent to which the main conclusions are transferable to other POPs, we need to consider both degradation and partitioning properties. PCB-153 is one of the most persistent POPs, both with respect to environmental persistence and biotransformation half-life. The less persistent a POP, the more source proximity will be controlling population exposure, because less persistence implies low potential for both long-range transport [31] and food chain bioaccumulation [32]. For example, Inuit populations will experience low exposure to non-persistent chemicals, because (i) environmental degradation will prevent the chemical from being efficiently transported over long distances and reaching the Arctic and (ii) biotransformation will prevent it from biomagnifying in the food chain. We had previously indicated that for a biotransformed compound a marine mammal in the food chain acts as a filter rather than an amplifier [1].

Some POPs have a higher long range transport potential than PCB-153 because higher volatility allows for more efficient atmospheric long-range transport [33, 34]. Examples are hexachlorobenzene and α-hexachlorocyclohexane. For such chemicals, air and water concentration ratios between remote and source regions are larger than calculated for PCB-153 (table 1), i.e. concentration gradients with distance from sources are less steep [35, 36]. In a post-ban period those ratios may even reach values above 1, i.e. contamination of the Arctic air and seawater is higher than that of source regions. In such cases, source proximity ceases to control population exposure and the bioaccumulation potential of the human food web becomes dominant. If such chemicals are sufficiently persistent to biomagnify in the food chain, clearly those with the highest exposure susceptibility (as expressed by the ESI) will suffer the highest exposure.

3. Conclusions

The modeling approach introduced and applied here can be used to explore the combined effect of source proximity, global transport, human food web structure and dietary habits on human exposure to POPs. Specifically, we have illustrated that it can be used to identify the most exposed human subpopulations globally and it can explain what factors are responsible for such elevated exposure. In the case of the PCBs, we predict that the presence of a warm-blooded carnivore in the diet is the factor most responsible for elevated PCB exposure in a human subpopulation, strong enough to overcome the dilution caused by even large distances from sources. Populations eating seals, such as the Inuit or subsistence fishermen on Lake Baikal, are thus among the most PCB-exposed populations worldwide. Our approach, however, also highlights that environmental contamination as controlled by source proximity is likely the decisive factor controlling differences in PCB exposure among most other human sub-populations (i.e. those not eating warm-blooded carnivores), as the effect of differences in food web structure (even those among fish vs. meat/dairy eating populations) are generally insufficiently large to overcome the effect of differences in environmental contamination. The quantity of animal lipid in the diet can also be an important factor.

Because of the dominance of the dietary exposure pathway for POPs, it is important to stress that it is the proximity of the area of dietary lipid generation to POP sources that is decisive rather than the proximity of the population itself. In other words, it is the location of the grazing area or the location of the feed growing area (e.g. corn or soy) that determines the contamination of dairy and meat products. And it is the location of the fishing grounds or the location of the production of the fish food (e.g. fish pellets used in aquaculture) that controls the contamination of dietary fish lipids. We have further illustrated that the modeling approach can take the area of food production into account. In particular, because of their dynamic nature, the models can be used to explore the effect of dietary shifts over time on POP exposure. These shifts include changes in dietary composition and changes in food sourcing and production. For POPs displaying strong spatial gradients in environmental concentrations, modeling of human exposure would benefit from improved knowledge of the sourcing of the food and feed supplying animal lipids to the diet.

The tools underlying the current study have applicability in the area of chemical regulation. We demonstrate that the model combination can be used to identify chemicals for regulation under the Stockholm Convention, [37] namely chemicals that as a
consequence of long range transport result in exposure that could cause adverse effects. Annex D of the Convention text [37] explicitly allows for the use of models in this context.

4. Methods

Overview. We used global, historical emission estimates [38] in combination with the global fate and transport model BETR-Global [39] to calculate spatially resolved, long term time trends in the contamination of air and sea water with PCB-153. PCB concentrations in air predicted by the BETR-Global model using this emission scenario show satisfactory agreement with long term monitoring data [40]. Each grid cell in BETR-Global contains seven compartments: upper air (free troposphere), lower air (boundary layer), vegetation, freshwater, ocean water, soil and freshwater sediments. We fed the BETR-Global generated fugacities in gridcells of the global model that represent the environment of the selected human populations as input into the human food chain bioaccumulation model ACC-HUMAN, [41] which had been parameterized for different human sub-populations as in Undeman et al [1].

Global environmental fate calculations. Global emissions of PCB-153 from the ‘higher scenario’ of Breivik et al [38] were pooled into the 15° by 15° grid cells of BETR-Global to generate annual average emission rates into every cell from 1930–2010. Emissions were assumed to be into the lower air compartment. Equilibrium partition coefficients and their temperature dependence were taken from [42] and degradation half-lives in water, soil and sediment were taken from [43]. The degradation half-life in air and the temperature dependence of degradation half-lives were taken as provided in BETR-Global. Changes made to the BETR-Global model prior to the simulations are described in detail in the supporting information.

Food chain bioaccumulation calculations. Food chain bioaccumulation was calculated using ACC-HUMAN [41] with additional marine mammal [44] and plant [45] models included as described in Undeman et al [1]. The composition of the human diet was identical to those defined for the six generic human populations described in Undeman et al [1]. While the Swedish reference population has a mixed diet of fish, beef and dairy, the fishermen eat predominantly fish, whereas the herders eat predominantly meat and dairy of a terrestrial herbivore (see supplementary table S2). The Inuit diet includes a ringed seal. These populations were assigned to BETR-Global grid cells as indicated in figure 1 and table 1.

In addition to the reference Swedish populations with a mixed diet, we also considered a Baltic fisherman with a diet comparable to that of the tropical fishermen as well as a Swede with a mixed diet, whose fish is internationally sourced (as opposed to Baltic Sea cod and herring). The calculation of the dietary contamination of the Swede eating internationally sourced fish (‘modern Swede’) was done as follows: Three further simulations of the reference scenario were conducted where the fish in the diet were simulated as (i) Northern Atlantic herring, (ii) Barents Sea cod, (iii) farmed salmon fed with internationally sourced fish meal and oil (from Peru, Chile, Iceland, Norway and the Barents Sea). We then calculated an average of the human body burden from the three scenarios weighted according to the fish types’ contribution to the Swedish diet [46]. A more detailed description is given in the supporting information.

For each population we calculated the concentration of PCB-153 in females representing different birth cohorts (1920, 1930, 1940, … 2010), both at birth (representing peri-natal exposure) and throughout their life (representing exposure over a 80 year lifespan). A dynamic, time-variant approach is necessary because of the long residence time of PCB-153 in the human body. To aid in the interpretation of the results we further used the environmental fate model CoZMoPOP II [47] and ACC-HUMAN to calculate the different subpopulations’ exposure susceptibility index (ESI) to PCB-153, as defined in Undeman et al [1]. The ESI expresses a population’s accumulation potential relative to the Southern Swedish reference population; a value above one indicates that, given identical emissions, a human living in an ecosystem of interest and sourcing food locally can accumulate a higher body burden of a contaminant than a typical Southern Swede.

The present approach does not account for subpopulations that were accidentally or occupationally exposed to PCBs [12] or experienced exposure from local contamination sources (e.g. in the vicinity of a PCB production facilities [13], or in indoor environments with PCB-containing sealants).

Model evaluation. The modelling approach adopted here has previously been shown to provide reasonable estimation of the PCB contamination of various human populations around the globe, living either in source proximity or in remote regions. Specifically, the combination of ACC-HUMAN with regional environmental fate model calculations has been successful in predicting PCB exposure in Scandinavian populations [48–50]. The combination of global fate calculations with the global fate model Globo-POP and an Arctic version of ACC-HUMAN predicted PCB concentrations that matched well with those recorded in the Inuit Health Survey in Canada [51]. PCB exposure predicted with the combination of BETR-Global and ACC-HUMAN matched that of US populations as sampled within NHANES [52]. Finally, the combination of historical emissions scenario, fate model and bioaccumulation/exposure model used in this paper was recently evaluated against human milk data from 56 countries from the UNEP/WHO Global Monitoring Plan and predicted PCB-153 concentrations to within a factor of 4 for 49 of 78 observations [53].
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