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Pilot study on long-term simulation of PCB-153 human body burden in the Tibetan Plateau

Libin Liu [a, c], Haiyan Zhang [b], Can Chen [a], Ziguang Li [d], Yue Xu [a, *]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550081, China
[b] The Johns Hopkins University-Nanjing University Center for Chinese and American Studies, Nanjing University, Nanjing, 210093, China
[c] College of Chemistry and Life Sciences, Zhejiang Normal University, Jinhua, 321004, China
[d] Haikou Forest Farm, Kunming Forestry Bureau, Kunming, 650114, China

HIGHLIGHTS

- Long-term Tibetan female burden of PCB-153 was simulated.
- The body burdens were within the range of low-to-moderate levels.
- Grain inflow can be as important as high-lipid diet to exposure risk.
- Chinese sources can contribute half of the PCB-153 burden by grain inflow.

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ABSTRACT

The historical body burden of 2,2′,4,4′,5,5′-Hexachlorobiphenyl (PCB-153) in the Tibet Autonomous Region (TAR) population was simulated on the basis of localized exposure factors and dietary data, which present a preliminary attempt to quantify the influence of high lipid dietary patterns, grain transported from inland China, and atmospheric transport on human exposure to polychlorinated biphenyls (PCBs). Herdsman with large animal-based food consumption exhibited the highest body burden that was comparable with that in inland China. The body burden of other residents was within the range of low-to-moderate level. High-lipid diet of urban residents caused their body burden being 1.5—-2.5 times higher than that of rural residents. The consumption of grain transported from higher polluted areas can also result in 50%—115% increase in the body burden of Tibetan rural residents compared with when local produced grain is consumed, suggesting that the influence of grain logistic can be as important as dietary patterns. The exposure risk for rural residents associated with grain logistic should not be ignored even if they consumed less high-lipid food. By splitting the inventory, over 80% of the PCB-153 pollution in the TAR was identified to be induced by atmospheric transport from foreign countries. However, the grain logistic contributed approximately half of the overall human body burden of Tibetan residents recently if assuming that the grain shortage was supplied by adjacent Sichuan Province. The combined influence of high-lipid diet, atmospheric transport and food logistic highlights the difficulties of risk control in remote regions that accumulate POPs, such as TAR.

1. Introduction

Over several decades, persistent organic pollutants (POPs) have been applied in densely populated and agricultural areas extensively. At present, POPs are at present ubiquitous in the world, accumulated in environmental compartments, and can bio-accumulate and bio-magnify in organisms because of their persistency, semi-volatility, and lipophilicity. Those compounds are threatening human health when consumed or exposed to contaminated environmental media. Many diseases, such as cancer, hypertension, asthma, and birth defects, have been associated with exposure to POPs (Annamalai and Namasivayam, 2015; Bencko et al., 2009; Meng et al., 2016; Perkins et al., 2016), thereby leading to considerable investigations to identify the key factors for exposure control, such as emissions, diet, food origins and exposure

* Corresponding author.
E-mail address: xu-yue@mail.gyig.ac.cn (Y. Xu).

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factors of local residents (Breivik et al., 2010; Zhao et al., 2018).

The Tibetan Plateau (TP) is the highest plateau with an average elevation of 4500 m. Known as the 'roof of the world' and the 'forbidden zone of life', anthropogenic activities in the TP are extremely limited. However, its central location also makes the plateau vulnerable to pollution from surrounding countries in Asia, such as China and India. Sufficient inorganic and organic pollutant observations demonstrated that air pollution around the Plateau has resulted in non-negligible ecological and human risk in the pristine land (Wu et al., 2016). Among the pollutants that were introduced into the TP, POPs are of particular scientific concerns because of their large usage in Asia, potential to be transported to remote areas, and possible long-term adverse effects on human beings. POPs transported by monsoon or the mid-latitude Westerly Jet can cross over the Himalayan Mountains to reach the TP (Sheng et al., 2013; Wang et al., 2015) and accumulate in the cold region with a low annual average temperature of 5 °C (Xu et al., 2013). Therefore, related risk assessment and management in the ecologically-sensitive region should be particularly important. To date, a few studies investigated POPs in the air, water, soil and biota in the TP (Sheng et al., 2013; Yang et al., 2010; Wang et al., 2010). Wu et al. (2016) assessed cancer risks caused by the consumption of fish, food, and water for adults and children and ranked them as low-to-moderate levels. Wang et al. (2016a) identified high-lipid content diet, such as yak butter, as the major contributor to exposure. These assessments, which were based on the monitored data, provided valuable information on health risks in the TP, although some crucial factors, such as long-term exposure variations, food origins, and localized exposure factors, were not included in these evaluations.

Numerical simulation is an economical and convenient risk assessment tool when direct measured data are insufficient. With the recently compiled age-dependent exposure factors (MEP, 2014; MEP, 2016a; MEP, 2016b) and relatively comprehensive food consumption dataset from the Tibet Autonomous Region (TAR) Statistics Bureau, the long-term 2,2′,4,4′,5,5′-hexachlorobiphenyl (PCB-153) human accumulation in the TAR was simulated here as a case study to illustrate the importance of body burden evaluation in the region. Exposure to polychlorinated biphenyls (PCBs) was associated with disorders in endocrine, reproductive, neurological, and immune systems (Ghosh et al., 2015). Previous investigations of the TAR showed that PCBs were the major contributors to the total cancer risk of POPs (Wu et al., 2016), and a declining trend of POPs, except for PCBs, was observed (Wang et al., 2018). Among PCB congeners, PCB-153 exhibited a mean environmental residence time of as long as 110 years (Jonsson et al., 2003), which pose long-term health risk. The level of PCB-153 is an excellent indicator of the total PCB levels in human blood serum (Covaci et al., 2002; Lee et al., 2007) and is therefore widely applied in human body burden estimation (Breivik et al., 2010; Zhao et al., 2018). Although the concentration level in the air was often below detection limits (Wang et al., 2016a), PCB-153 was still the most prevalent PCB congener detected in biota samples, such as the fish and yak tissues collected in the TP (Yang et al., 2010; Wang et al., 2010) and breast milk in China (Zhang et al., 2017), possibly because of its high bioconcentration factors (Wang et al., 2016b). According to the TAR Statistics Bureau, these foods were consumed by TAR residents, especially by urban residents. Those factors make PCB-153 an ideal compound for modeling health risk. In the present study, scenarios that consider dietary variations, grain logistic, and different long-distant source regions, including India and China, were simulated to illustrate the importance of these factors on the PCB-153 body burden. This work contributes to a preliminary understanding of the long-term risk of exposure to POPs in the TAR.

2. Methods

2.1. Numerical simulation procedure

The long-term body burden was estimated by emission, residue, and multimedia exposure models. First, the historical emission inventory of PCB-153 was generated by an emission model. Second, the inventory was processed by a residue model to calculate the environmental concentrations in the air, water, soil and sediment. Finally, a multimedia exposure model was applied to simulate long-term variations of body burden for females in different age groups given that field study suggests that gender differences of body burden was insignificant in China (Zhao et al., 2010). These females were assumed to be born at the beginning of each year.

2.1.1. Emission inventory

PCB-containing product usage in the Tibet was limited (Xu et al., 2018). The PCB residues in the TP were mainly originated from surrounding countries, such as India and China (Sheng et al., 2013; Wang et al., 2015). These potential sources were included in the inventory presented by Breivik et al. (2007). Recently, an improved inventory in China was reported, and its modeled environmental concentrations of PCB-153 fitted better with the historical recorded values (Xu et al., 2018). In the present study, the improved inventory in China with 1/4° latitude × 1/4° longitude resolution and emission inventories from other Asian countries from the study of Breivik et al. (2007) were combined. To remain consistent with the spatial resolution in China, the original inventory of PCB-153 (Breivik et al., 2007) of other countries was spatially interpolated from 1 ° × 1 ° into 1/4° × 1/4° grid using population density data as surrogates. The interpolation may reduce the accuracy of a model outside China, but its influence in the simulation results of the TAR should be minor. PCBs can be released as by-products of manufacturing or thermal processes (Bartlett et al., 2019). The unintentionally released PCB-153 was not included because of the lack of reliable inventories. The inventory domain was set from 0°N to 60°N and from 50°E to 150°E to include potential sources of the TAR. Time step was set as 1 day.

2.1.2. Environmental residue model

PCB-153 concentrations in the air, water, soil and sediment were simulated from 1950 to 2018 by ChnGPERM (Chinese Gridded Pesticide Emission and Residue Model), a gridded transport and transfer model based on fugacity method (Tian et al., 2009). It has been proved to be capable of capturing the spatial and temporal variations of PCB-153 residues in China by comparison with a large monitored dataset. Detailed temperature-dependent physicochemical properties of PCB-153, parameter setting and simulation methods were discussed previously (Xu et al., 2018). Meteorological data were obtained from the United States National Center for Environmental Prediction (NCEP) reanalysis dataset (Kalnay et al., 1996) and interpolated to 1/4° × 1/4° resolution by cubic interpolation.

2.1.3. Multimedia exposure model (MEM)

A MEM applies level-IV fugacity method to simulate chemical residues in plants and animals to calculate human exposure via ingestion of water and food. The simulation generated the concentrations of PCB-153 in biotic and abiotic media, exposure dose, and female body burden in each 1/4° × 1/4° grid cell at a time step of 1 day. The statistical data on the food consumption of Tibetan residents categorized major daily food intake into 10 groups, including grain, beef, pork, poultry, egg, fish, fruit, vegetable, edible oil, and milk. According to the China Statistical Yearbook, rice/barley, beef, pork, chicken, egg, carp, apple, Chinese cabbage, peanut oil and cow milk are the major contributors to the total production of those foods in China. Except for Chinese cabbage and peanut, of which the plant parameters were not well documented, other species were selected as representative food to simulate the PCB-153 concentrations in the 10 major food groups. Pakchoi cabbage and soybean with detailed growth parameters in China were selected to represent vegetable and edible oils. The fugacity equations, growth curve and other parameters of representative plant and animal can be found in Section 1 of Supportive information (SI). Mechanistic description of the kinetic processes of bioaccumulation of lipophilic organic pollutants from air, water and food to humans has been illustrated in a fugacity-based model (ACC-HUMAN) (Czub and McLachlan, 2004). The time-varying PCB burden simulation for females was added into MEM to calculate the spatial and temporal variations of human accumulation in the TAR. The overall fugacity equation for human can be described as:

$$\frac{d(V_H Z_H f_H)}{dt} = E_{OH}\sum_i (D_{UHi} f_{UHi}) - E_{OH} D_{EH} f_H - (D_{perH} + D_{MH} + D_{LH} + D_{chH} + D_{urH}) f_H + D_{reH}(f_A - f_H) \tag{1}$$

where f_H, f_A, f_W, and f_{UHi} are the fugacity of human, air, water, and the ith intake food type, respectively. V_H and Z_H are the volume and fugacity capacity of the whole human body, respectively. E_{OH} is the gastrointestinal absorption efficiency. D_{UHi}, D_{EH}, D_{perH}, D_{MH}, D_{LH}, D_{chH}, D_{urH}, and D_{reH} are mass transfer coefficients via food intake, egestion, percutaneous excretion, metabolism, nursing, childbirth, urine excretion, and respiration, respectively (Czub and McLachlan, 2004). To improve the efficiency and accuracy of the model, analytical solutions of fugacity equations were presented in Section 1.3 of SI. Age-dependent parameters, such as most D-values, were calculated using the method previously presented by Czub and McLachlan (2004), with localized food and water intake rate (Section 2 in SI).

2.1.4. Dietary patterns, exposure factors, and grain logistic in the TAR

Oral food intake is the major route for OCP and PCB exposure on TAR residents (Wu et al., 2016). Historical dietary pattern in China changed substantially, especially in the last 40 years. Thus, the dietary data in the TAR is obviously necessary. A relatively comprehensive food consumption dataset, including the annual food consumption of urban and rural residents in the TAR from 1980 to 2018 were applied in the present study (Fig. S3). National consumption dataset before 1980 were interpolated using the food consumption of each province in 1980 as surrogate to attain the corresponding data in the TAR (Section 2.1 of SI), assuming that the dietary preferences in Chinese provinces remained the same from 1950 to 1980. Unlike the original ACC-HUMAN, food intake here was categorized into grain, beef, pork, chicken, egg, fish, fruit, vegetable, edible oil, and milk, thereby covering the most of food types in the daily consumption recorded by the TAR Statistics Bureau. That is, the simulated human body burden in MEM could be higher than ACC-HUMAN when the environmental concentrations were the same.

Exposure parameters are basic data that describe the characteristics and behavior of human exposure to pollutants that are essential for the exposure and risk assessment. Given that the dietary habit and physiological features of Tibetan residents were significantly different from those in western countries or inland China, exposure factors, such as D-values should be recalculated on the basis of the Exposure Factors Handbook of Chinese Population (MEP, 2014; MEP, 2016a; MEP, 2016b). We adopted female data in Western China, which includes Tibetan residents, because exposure factors in the TAR were not reported separately in the handbook. Age-dependent inhalation rates and drinking water, cereal, vegetable, fruit, milk, meat, fish, egg, and edible oil ingestion rates of residents in urban and rural areas were collected from the handbook. Age-dependent body weight, lipid content, and body water content data of Tibetan residents were adopted from literatures (Fig. S4—S14). A significant increase in adolescence was found for many factors like food ingestion and lipid weight. It possibly reflects the improved nutrient supply of adolescent children. Because only the annual average dietary data from 1980 to 2018 and age-dependent exposure factors in 2002 were available, those data were further interpolated into the annual consumption for each age group using age-dependent exposure factors as surrogates. The dietary data in the TAR before 1980 was interpolated by the national data and used as spin-up period of the model (Section 2.1 of SI). Thus, discussions will mainly focus on the result from 1980 to 2018. Details of those age-dependent data were included in Section 2.2 of SI. However, the contribution of each food category to the overall intake exposure in the following discussion was calculated using average annual food intake rate rather than age-dependent intake rate.

The grain flow data in the TAR were not recorded. Given that most of the grain transported to the region came from adjacent province Sichuan (Duan et al., 2019), we applied a simple mass-balance approach to estimate grain flow. Annual grain production in the TAR was interpolated into each model grid using cropland density from ChnGPERM as surrogates, whereas the consumption corresponded to the product of population and per capita annual consumption of grain (SI). The grain transported from Sichuan was calculated as the differences between the total grain consumption and total production in the TAR. In each model grid, the grain demand was first supplied by local production. Extra grain in model grids was assumed to be moved to an imaginary granary and mixed with the grain transported from inland with average PCB-153 concentrations in Sichuan. The mixed grain was then transported to grids with grain shortage in the model.

2.1.5. Scenario setup and data analysis

In the present study, the contribution of atmospheric transport, grain logistic and dietary pattern on human exposure should be considered. To investigate the potential influences of these factors on human body burden, numerical simulations have been performed for five model scenarios. The first scenario (s1) is a baseline scenario with all the emission sources and no grain flow. In addition to the historical dietary patterns of urban and rural residents, the high-lipid diet of herdsman in rural areas was also estimated in s1. The second scenario (s2) only considers historical emission sources from PCB-containing products in the TAR the presents the influence of local emission on the overall body burden of PCB-153. Scenarios 3 to 5 were conducted to evaluate the relative importance of atmospheric transport from foreign countries and grain logistic from inland China. The third scenario (s3) includes all the emission sources, time-varying dietary patterns and grain trade. Considering that atmospheric transport from India has been considered the major contributor of the local pollution of the TP (Wu et al., 2016), the fourth one (s4) only includes the emission sources in India and grain trade to represent the contribution of atmospheric transport. Grain deficiency in the TAR was mainly supplied by inland China. Thus, fifth one (s5) includes PCB-153 emission in China and grain trade to represent the contribution of grain logistic. These model scenarios are summarized in Table S5.

For simplicity, only body burden in the TAR were presented and discussed. The average body burden in this study refers to the population-weighted average body burden of female residents. Scenarios 4 to 5 represented body burden influenced by different sources. Therefore, we defined the contribution of certain source as:

$$R_{si} = \frac{C_{si}}{C_{s3}} \tag{3}$$

where C_{si} and C_{s3} are the population-weighted average body burden of 30-year-old females in the TAR of the fourth or fifth scenario to that of the third scenario, respectively.

2.2. Model evaluation

Measured PCB-153 concentrations in the soil and peat core samples have been compared with the ChnGPERM output in previous simulations (Xu et al., 2018). Squaring the Pearson's correlation coefficient (R^2, the coefficient of determination) of 0.5 suggests that the transport and transfer model captured the spatial trends of PCBs in the environment well. Concentrations in biota samples should be compared to evaluate the accuracy of exposure model. Wang et al. (2016b) reported that the average grain concentration in the TP in 2011 was 0.8 pg/g (dry weight), which is comparable with the modeled value of 0.98 pg/g. Given that PCB-153 contents in human tissues have not been reported in the TAR yet, we compared the PCB-153 concentration levels in Tibetan yak milk butter (Wang et al., 2010), fish samples from fresh water, and those in the breast milk of Chinese females (Zhang et al., 2017). The butter and breast milk datasets were documented in the unit of ng/g lipid. We assumed that the default lipid content was 3.2% for breast milk (Zhao et al., 2018) and 6.5% for yak milk (Wu et al., 1998) to convert the model results. The fish dataset was recorded in ng/g based on wet weight. Figs. S15–S17 compared the modeled and measured PCB-153 concentrations in the three datasets, respectively. The modeled concentrations were generally within the similar levels of measured data. The R^2 of 0.38 for fish and yak butter samples suggests that the present simulation can reflect the average PCB-153 concentrations in food produced in the TAR. The R^2 were relatively low, which may be related to the uncertainties of multimedia exposure model, in which the parameters of 10 plant and animal species were applied to represent all the food consumed by the TAR residents. As a matter of fact, the growth curve, species, age, and other biological factors may influence the PCB-153 concentrations in biota. The parameters presented in SI only selected the food with large consumption, which may not reflect the local food properly. The R^2 between modeled and measured breast milk concentrations was 0.43, suggesting the human accumulation module (Eq. (1)) worked well for the Chinese population. A major factor influences model accuracy is that the improved inventory in China assumed that PCB-containing products were disposed locally, which overestimated the residue levels in megacities where waste could be transported out and relatively clean food can be transported in considerably. Uncertainties of inventory and foodstuff made the correlation between modeled and measured data weak in many exposure studies (Zhao et al., 2018). These issues are beyond the scope of the present study and should be investigated in the future.

3. Results and discussion

3.1. Human body burden of PCB-153 in the TAR

Temporal trends of the average body burden of Tibetan females born at 10-year intervals in s1 were plotted in Fig. S18. The PCB-153 body burden peaked at approximately 8-year-old. After that age, lipid weight increased fast (Fig. S4) and 'diluted' the concentration of pollutants. When the lipid weight reached a peak at 18-year-old, the body burden also decreased to a low level in spite of the high food intake rate at that age (Figs. S7–S14). With the decline in lipid weight at 25-year-old, the body burden gradually increased. In their 30s, body burden decreased to another trough because childbirth and lactation can reduce the body burden of a mother significantly and deliver PCBs to her baby (Czub and McLachlan, 2004). The similar pattern of the '10-year-old peak' and '30-year-old trough' were found in many human body burden studies (Zhao et al., 2018). In the TAR, the PCB-153 body burden of females born before the 1990s was generally low because of the increasing emission in Asia after 1990s. After their thirties, the body burden gradually increased to a relatively high level. After the 2000s, primary emission of PCBs began to decline in Asia, therefore the body burden of individuals born after the 2000s hardly rise again.

The human body burden of 30-years-old female has been adopted as an indicator to illustrate body burden variations in China (Zhao et al., 2018). Thus, we also applied it for comparison in the following discussion. Previously estimated body burden in central China, where PCB-containing products were widely used, ranged from several to tens of ng/g lipid (Zhao et al., 2018). The simulated average body burden of Tibetan females in s1 was approximately an order of magnitude lower than that of other Chinese and more than two orders of magnitude lower than that of European (Zhao et al., 2018). The level was also lower than the observed data in other Chinese provinces (Zhang et al., 2017), which is consistent with the conclusion from observations that the risk of exposure to POPs in the TP were in the range of low-to-moderate levels (Wu et al., 2016). Notably, the historical usage of PCBs in China mainly concentrated in the eastern and central regions. Emissions in the western areas of China were significantly low. For example, the body burden of 30-year-old females of Sichuan Province on the eastern edge of the TAR was also within the low-to-moderate range, which is approximately twice of that in the TAR (Fig. 1). The accumulated PCB-153 emission of the two provinces accounted for 4% and 0.06% of the national total, respectively. However, their female body burden levels were within the same order of magnitude. If only local emissions and average historical dietary consumption were considered (s2), then the female body burden in the TAR was at least two orders of magnitude lower than the other scenarios. This observation indicates that some processes, including high-lipid dietary patterns, grain transported from other Chinese provinces, and atmospheric transport from adjacent regions may contribute to the body burden increases in the TAR.

3.2. Dietary influences on body burden

The body burden variations among urban residents, rural residents and rural herdsman in s1 were compared to illustrate the influence of dietary pattern. Generally, environmental pollution levels were considered higher in urban areas. Field observations found that Tibetan soil concentrations in urban areas were

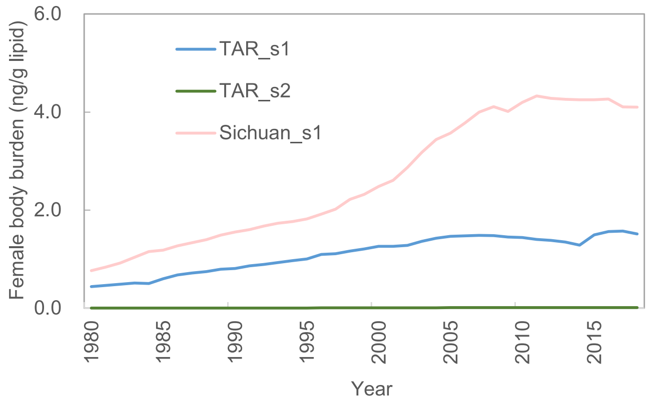

Fig. 1. Average body burden of 30-year-old TAR and Sichuan females in s1 and the values of TAR residents in s2.

comparable to or somewhat higher than those in rural areas (Wang et al., 2009). In scenarios with all the potential emission sources, modeled concentrations in populated areas exhibited a similar spatial trend, thereby leaving food consumption the major contributor to the differences in body burden of Tibetan residents. Dietary intake variations can influence the overall human exposure (Huang et al., 2016). The growth of non-staple food consumption in the TAR was significant after the 1980s, especially for urban residents (Fig. S3). Grain and yak butter were the major contributor to the rural resident's PCB-153 intake, whereas yak butter and fish were the predominant oral intake route for urban residents (Fig. S20). Although Tibetan residents consumed less fish than other Chinese, urban residents in the TAR still consumed a considerable amount of fish according to the data from the National Bureau of Statistic (Fig. S3). As a kind of food with high PCB-153 loadings (Llbobet et al., 2003), fish can contribute a significant part of body burden on urban residents. The body burden of 30-year-old female urban residents became 1.5 to 2.5 times higher than those of rural residents because of consuming more animal-based food.

Notably, a group of high-risk rural residents, namely, herdsman, consumed more animal-based food than urban residents did. The estimated body burden of herdsman were approximately 4–8 times of other residents in the TAR and even higher than the levels in adjacent Sichuan Province (Fig. 2). The body burden of 30-year-old female herdsman in the TAR was comparable to the body burden that ranged from several to tens of ng/g lipid in central China (Zhao et al., 2018), thereby suggesting that their dietary pattern posed a significant risk on PCB-153 exposure. The traditional Tibetan diet consists of superfluous meat because growing vegetables or fruits in this area is difficult. High PCB-contaminated food like fish was rarely consumed by rural residents in the TAR because of previous religious disciplines. However, the harsh environment of the TP made it necessary for local residents to ingest food that is high in protein and lipid (Li et al., 2013). The average beef, milk, and butter consumptions in China were the highest. If considering the consumption of sheep produced food with higher contamination levels than cow (Sethi et al., 2017), the estimated body burden will be higher (Fig. S19). As a matter of fact, the recent grain, meat, and edible oil intake of Tibetan residents has already exceeded the upper limits of the recommended values from the Chinese dietary guidelines, whereas the consumption of other food, such as fruits and vegetables, were still lower than the lower limits (Table S7). As the living standard improved, animal-based food consumption are expected to increase in the future. The food habits of herdsman are neither conducive to nutritional balance nor to the reduction of the human body load of POPs. The high-risk population in the TAR should be included in risk evaluation.

3.3. *Body burden enhanced by grain logistic*

Food logistic has been suggested as another crucial but not well quantified factor contributed to exposure risk (Huang et al., 2020; Zhao et al., 2018). Food transport between Tibet and inland China is difficult. Most animal-based food was produced locally, but the grain demand in the TAR is large. Most of the grain shortage was supplied by the grain from Sichuan Province (Duan et al., 2019). Assuming that all the grain shortage of the TAR was satisfied by those transported from Sichuan, then it will lead to a PCB-153 body burden enhancement in the TAR. Fig. 3 compared the body burden 30-years-old urban and rural females in the scenarios without (s1) and with grain trade (s3). Rural residents with relatively high grain consumption exhibited the largest body burden increase rate that ranges from 50% to 115%, whereas the increase rates for urban residents were only 10%–20%. Dietary differences can lead to 1.5–2.5 times of the body burden increase between urban and rural residents. However, the urban-rural differences were reduced by grain logistic. Implicated in those increase rates is that risk of rural residents should not be ignored even if they consumed less high-lipid food. Grain intake can also contribute a significant proportion to the overall exposure for the TAR residents.

Wang et al. (2016b) estimated the grain intake contributed 9.3% to the total herdsman oral intake based on the PCB-153 concentrations of local food. In s1, the estimated contribution of grain is

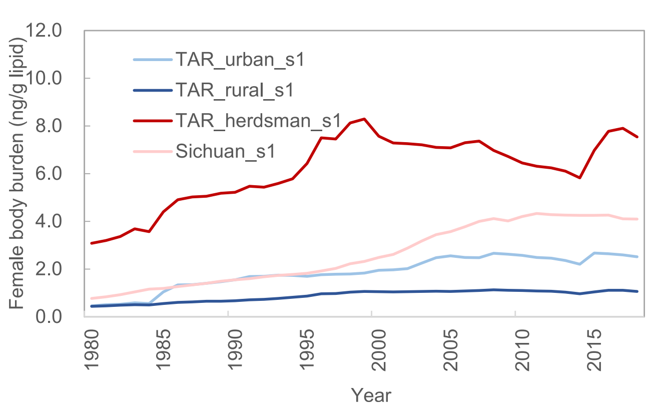

Fig. 2. Average body burden of 30-year-old females of TAR urban residents, rural residents, herdsman, and Sichuan residents in s1.

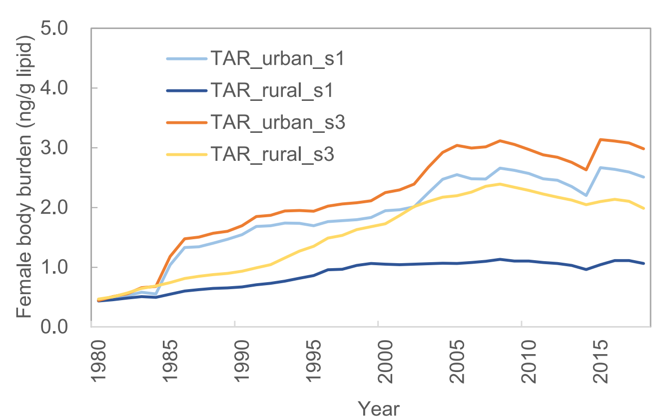

Fig. 3. Comparison of body burden of TAR urban and rural residents without grain flow (s1) and with grain flow (s3).

12% for herdsman, comparable with that estimation. Evidently, the importance of grain was lower than those of other animal-based food, such as yak milk butter. However, local grain production in the TAR cannot meet its grain demand. About 8%–40% of the grain consumption was supplied by inland China (Duan et al., 2019). In scenarios with grain trade (s3), the ratios between annual grain concentrations in Sichuan and the TAR ranged from 1.2 to 10.9 after Chinese source emissions in 1965 (Fig. S21). Consumption of grain produced in Sichuan will increase exposure risk associated with staple food. Until 2018, the average contribution of grain intake to the total oral intake has already increased to 65%, and 34% for rural and urban females, respectively (Fig. S22). It suggests that grain transported from relatively polluted areas could increase the exposure risk associated with staple food consumption considerably. Although the staple food intake decreased with the living standard improvement in the TAR, it still contributed a significant portion to Tibetan diet. That intake route was not routinely included in previous human body burden estimation. Without considering grain trade, the exposure risk in areas with substantial grain shortage, such as the TAR, could be underestimated.

3.4. Contributions of atmospheric transport and grain logistic

The estimated contribution of Indian source on the TP soil can reach 90% for other POPs because of prevailing Indian monsoon in rainy season (Xu et al., 2013). As discussed above, the grain logistic from inland China may also account for PCB-153 exposure to human. To evaluate the contributions of atmospheric transport and grain logistic on female body burden of PCB-153 in the TAR, the numerical simulations were conducted with Indian (s4) and Chinese (s5) emissions only. The air concentrations in s4 and s5 were normalized by those with all the sources (s2). During the whole modeling period, Indian emissions were responsible for more than 80% of PCB-153 in the air of TAR, whereas Chinese emissions contributed less than 10% (Fig. S23). It is consistent to the conclusion of previous simulation of α-HCH in Asia (Xu et al., 2013) and many field observations in the TP (Wu et al., 2016).

As for female body burden, the contributions of atmospheric transport and grain logistic varied annually. In Fig. 4, the relative contributions of the two processes to body burden of 30-year-old females in the TAR were plotted. Before the usage of PCB-containing products in China, atmospheric transport dominated the human exposure of PCB-153 in the TAR. When China began to use PCBs in 1965, the average concentration of grain in Sichuan

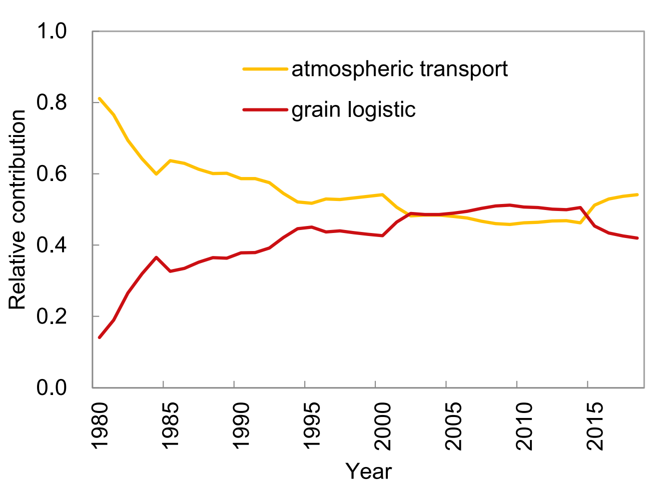

Fig. 4. Relative contribution of atmospheric transport and grain logistic on body burden of TAR residents when grain transportation from Sichuan was considered.

exceeded that in the TAR (Fig. S21). The influence of grain logistic increased simultaneously associated with grain transportation from Sichuan. After the 1980s, the contribution of grain logistic exhibited an increasing trend. About half of the grain demand in the TAR was supplied by grain from inland China and the grain PCB-153 concentration differences between Sichuan and the TAR increased. The influence of grain logistic increased again. The contribution exceeded that of atmospheric transport during the 2000s. In the 2010s, the pollution level in Sichuan grain declined because of emission source control and then the contribution of atmospheric transport bounced back.

4. Conclusion

Estimation of human exposure to POPs is a comprehensive work because of many influential factors, such as emissions, atmospheric transport from potential sources, dietary pattern and food origins. We simulated the long-term variations of body burden of PCB-153 in the TAR with relatively localized exposure factors as an example to illustrate the relative importance of those factors on the overall exposure risk. The estimated body burden of herdsman was comparable with the average level of adjacent Sichuan residents. Yak butter can be considered as high-risk food. High animal-based food diet of urban residents led to a 50%–150% body burden increase compared with those of rural residents. Grain transported from inland China is an equally important factor that elevates the body burden of PCB-153 in the TAR, especially for rural residents. Rural residents either continue their high grain diet (ingesting grain from inland China) or move to urban areas (adopting high lipid diet) are facing an enhanced PCB-153 body burden. Atmospheric transport can only be managed by source control. Given the location and harsh environmental conditions of the TAR, changing high lipid/grain diet or stopping grain logistic is also difficult. The study highlights the difficulties for risk control of POPs in the TAR.

Certain limitations of the present study should be noted. First, the grain shortage in the TAR was assumed to be satisfied by the supply from Sichuan Province because of lack of reliable grain logistic data. Second, grain transport from other provinces could be underestimated. The body burden of herdsman was also not calculated in s3 to s5 because the number of herdsman and other rural residents, which is crucial for grain demand calculation, was not recorded in the statistics dataset. Meanwhile, the prediction was constructed on the basis of the current emission inventory and model frame. Recent studies show that many sources, such as combustion, industrial processes, pigment, and polymer sealant, may contribute to the unexpected high abundance of PCB and other POPs emissions. Whether those sources in adjacent areas influence the TAR needs further research. Other factors, such as PCB distributions among different organs, historical trend of age-dependent exposure factors, high risk related to offal intake, and the association between body burden and diseases, which are also important for risk assessment, were not included in the present study because of data deficiency. The human body burden of POPs is still worthy of evaluation at remote regions, such as the TP, in the future.

Author contribution

Libin Liu: Conceptualization, Investigation, Writing - original draft, Writing - review& editing, Formal analysis. Haiyan Zhang: Conceptualization, Investigation, Writing - review & editing, Supervision. Can Chen: Methodology, data collection and validation, Writing - review & editing. Ziguang Li: Methodology, data collection. Yue Xu: Conceptualization, Methodology, Software, Writing - review & editing, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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