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Estimating polybrominated diphenyl ether (PBDE) exposure through seafood consumption in Switzerland using international food trade data

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A R T I C L E I N F O

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A B S T R A C T

Seafood is a major source of human exposure to polybrominated diphenyl ethers (PBDEs). The intake of these globally distributed and bioaccumulative contaminants depends on both consumption patterns (which seafoods are consumed) and on their origins. Here, we investigate exposure to PBDEs through seafood consumption as a function of species, origins and consumption levels. We estimate the contribution of seafood consumption to PBDE exposures in the Swiss population using two approaches. The first approach estimates exposures by estimating the composition of the Swiss seafood diet using trade data and national statistics on total seafood consumption. This naïve approach could be used for any country for which no individually reported consumption data are available for a population. The second approach uses dietary survey data provided by the Swiss Federal Statistical Office as part of the menuCH study for exposure estimates. To support region- and species-specific estimates of exposures for both approaches, we built a database of PBDE concentrations in seafood by analysis of published PBDE levels in fish from food markets or freshwater resources from various countries. We find estimated PBDE exposures ranging from 0.15 to 0.65 ng/kg bw/day for the trade data-based diet. These were close to the median exposures of 0.68 ng/kg bw/day for the Swiss population based on the menuCH survey, indicating that the composition and consumption rate derived from trade data are appropriate for calculating exposures in the average adult population. However, it could not account for PBDE exposures of more vulnerable (high seafood consuming) populations captured only by the survey data. All estimates were lower than the PBDE Chronic Oral Reference Doses (RfD's) suggested by the EPA, but could increase substantially to a value of 7 ng/kg bw/day if fish are sourced from the most contaminated origins, as in the case of Vietnamese shrimp/prawn, Norwegian salmon, and Swiss whitefish. Exposures as high as 8.50 ng/kg bw/day are estimated for the survey-based diet, which better captures the variability in consumption by individuals, including extreme high and low values. In general, the most frequently consumed species reported by Swiss consumers are consistent with those predicted using trade data.

1. Introduction

Polybrominated biphenyl ethers (PBDEs) are lipid-soluble compounds (Schecter et al., 2010) used as flame retardants in synthetic fibers like rayon, nylon, polyester (Shin and Baek, 2012) and molded plastics (Kim et al., 2006). There are 209 different PBDE congeners (Darnerud et al., 2001) based on the number (2–10) and configuration of bromines attached to diphenyl rings (EPA-a, 2014). Three technical mixtures of PBDE homologues have been commercialized since the early 1970 s: (CDC, 2016) pentaBDE, octaBDE and decaBDE (EPA-a, 2014), of which decaBDE is the most abundant in the environment (de Wit, 2002). PBDEs are released into the environment during manufacture, use and disposal of products, eventually making their way into ecosystems where they enter food chains, accumulating in fat-rich tissues. The commercial production of pentaBDE and octaBDE ceased in 2004 due to emerging recognition of their bioaccumulative, toxic and persistent nature (Betts, 2008), and in 2008 deca-BDE was also banned by the European Court of Justice (Betts, 2008; The Official Journal of European Union, 2008). Despite the bans on PBDEs in the United States (U.S.) and European Union (E.U.) (EPA-a, 2014) and their inclusion under the Stockholm convention as Persistent Organic Pollutants (POPs) in 2009 (Idowu et al., 2013), PBDEs continue to be a matter of

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concern to human health (Costa et al., 2014; de Wit, 2002; EFSA, 2011) since they are persistent in the environment and are incorporated into materials that may still be in use or releasing PBDEs after disposal. Animal studies have confirmed toxic effects including neurobehavioral changes (e.g. lower IQ), reproductive system damage, and thyroid and liver malfunctions due to PBDE exposure (Akortia et al., 2016; Darnerud et al., 2001; Herbstman et al., 2010).

PBDEs enter human bodies through dust ingestion and inhalation of contaminated air as well as food consumption (Anh et al., 2017; Streets et al., 2006), with the latter being a major source of exposure (Besis and Samara, 2012; Trudel et al., 2011a). Studies have confirmed that fish, meat and dairy products contribute significantly to daily PBDE intake (Trudel et al., 2011b). For investigating fish intake as an exposure pathway, species-specific intake data are crucial. Some national agencies have been successful in conducting dietary surveys to furnish species-specific databases. For instance, the National Health and Nutrition Examination Survey (NHANES) (CDC, 2014) conducted by the Centers for Disease Control and Prevention (CDC) reported 24-h and 30-day species-specific fish consumption frequency for several regions in the United States. Similar surveys have also been conducted in many European countries. For instance the National Diet and Nutrition Survey (NDNS) (NDNS, 2008) in the UK and the Belgium National Food Consumption Survey 2014–2015 (Scientific Institute of Public Health. WIV-ISP, 2015) in Belgium. However, not all countries conduct these surveys, so alternate data sources are needed for generating seafood diets. Additionally, researchers have derived fish consumption patterns for Portugal and Greece among others countries, using information on trade data and fish landings (Willemsen, 2003). However, in our understanding no study has attempted to validate trade-estimated seafood diets by comparing them with survey based dietary data. Here, we evaluate whether widely available trade data can generate reliable dietary estimates using pre-existing survey data for comparison.

Apart from being a tool for providing insight into typical diets for modern populations, international food trade data can also add an important dimension to the chemical exposure landscape: the transfer of contaminants across borders (Ercsey-Ravasz et al., 2012; Ng and von Goetz, 2017). This is particularly appropriate for a globally distributed class of chemicals like PBDEs (Perkins et al., 2014). When in commerce, the majority of PBDEs were synthesized in the E.U., U.S., China, Israel and Japan (Akortia et al., 2016). However, practices like waste management (recycling, disposal or landfilling), emissions from construction materials, and food trade can effectively disseminate these contaminants and may be responsible for the ubiquitous occurrence of PBDEs in the environment (Anh et al., 2017; D'Odorico et al., 2014; Duan et al., 2016). PBDEs contained in electronic waste have been identified as one of the most critical ongoing emissions pathways (Wang et al., 2009). Many developed nations like the U.S. and members of the E.U. export their e-wastes (containing PBDEs) for processing and disposal to developing countries, including India and China (Lee et al., 2018; Perkins et al., 2014). PBDEs emitted from e-waste make their way into the local environment and ultimately into the food chain (Zhao et al., 2009). Many e-waste dumping destinations are also major hubs for global aquaculture production, and actively export seafood to other parts of the world (Ahmed et al., 2018). In 2016, Asia contributed 89% to global aquaculture production, China being the highest producer (61.5% of total aquaculture production), followed by India, Indonesia, and Vietnam (Ahmed et al., 2018; FAO. The State of the World Fisheries and Aquaculture 2018-meeting the sustainable development goals, 2018). Although the concept of e-waste dumping is not new, the impacts of contaminants being transferred across borders are still poorly quantified (D'Odorico et al., 2014; Ercsey-Ravasz et al., 2012) and food as a means of transport has not been explored (Ng and von Goetz, 2017). In this study, we estimate PBDE exposures via dietary intake of internationally traded seafood and compare methods to generate representative diets, using both trade- based data and a pre-existing survey. We calculate PBDE exposures using both trade data from the UN Comtrade Database (UN Comtrade, 2016) and survey data from the menuCH National Nutrition Survey 2014/2015 (SFFSVO, 2018), evaluating the influence of seafood origin on PBDE exposure.

2. Methods

2.1. Study area

We selected Switzerland as our case study based on the role of food trade in its economy and the availability of dietary survey data. Fish consumption has increased substantially in Switzerland over the past decades: approximately 8.8 kg of fish were consumed annually per person in 2014, in comparison to only 6.4 kg in 1984 (FOEN, 2016). Since fish bioaccumulate PBDEs from their surroundings (Barber, 2008; Ng et al., 2018), this 37.5% increase in fish consumption could contribute to increased PBDE exposure. Moreover, Switzerland is among the countries with the highest share of foreign trade in gross domestic product (GDP) (FSO, 2016), implying that integration of seafood trade in our study would be relevant for this population.

Our study investigated PBDE intakes from seafood consumed by the Swiss population using two different approaches: trade data and survey data. Using trade data, we report here import volumes for individual seafood species (referred hereafter as "species-specific") and by the country of origin (referred hereafter as "origin-specific"). Using the survey data, we calculated daily seafood intakes for individual seafood species, but as origins of the seafood consumed are not reported by respondents in the menuCH survey, these are referred to hereafter as "species-specific but not origin-specific".

2.2. Construction of seafood consumption characteristics

2.2.1. Swiss diet constructed from trade data and domestic catch

Seafood imports to Switzerland from the rest of the world, extracted from the UN Comtrade Database (UN Comtrade, 2016), together with domestic fish catch, reported by the Swiss Federal Office of Fisheries Statistics (FOEN, 2016), were used to build a diet profile. All calculations are based on trade data from 2016. We assume the trade statistics to translate to consumption by adults, in order to compare with the menuCH survey of the adult Swiss population. However, national trade statistics account for the entire population; therefore, there is some uncertainty associated with assigning trade data to the diet of a particular population sector. Note that the term "seafood" is used here for all consumable aquatic species (marine or freshwater) in general.

Imports reported by Switzerland (mass imported; kg/year) were extracted for seafood including fish, mussels, and shrimp (these tend to dominate the Swiss diet) covering fresh, frozen, fresh fillet, and frozen fillet categories (see SI, section S2). Mass exported in kg/year for the same commodity codes as reported by Switzerland's trade partners was also obtained to assess discrepancies between partner-reported exports and Swiss-reported imports (Feenstra et al., 2005) (SI Figure S2).

From the list of total imported commodities, we report here the top 20 seafood types used for calculating "species-specific and origin-specific" PBDE exposures (Table 1; for a complete list of total seafood commodities imported see SI Table S3). We also included the complete list of imported species and not only the top 20 to calculate "species-specific but not origin-specific" PBDE exposures (see details in Section 2.5). Note that in Table 1 and SI Table S3 multiple entries may occur for related species, as reported in the UN Comtrade Database. For example, separate entries exist for Salmon, Trout and Salmonidae, with the Salmonidae entry explicitly stating: "Salmonidae excluding 030211 and 030212", where 030211 and 030212 are entries for common species of Trout and Pacific/Atlantic/Danube Salmon, respectively. Since we have extracted all our trade data from Comtrade, we retained the same nomenclature.

Among the entire range of countries supplying seafood to Switzerland, we focused on the top three exporters for each seafood

Table 1
Traded quantities of species selected for trade-data based diet generation (UN Comtrade, 2016).

*Seafood species/group	Imports (kg/year)	Exports + re-exports (kg/year)	Net quantity (kg/year)
Salmon	9,519,516	52,577	9,466,939
Shrimp	4,609,169	29,276	4,579,893
Catfish	2,802,212	4396	2,797,816
Flatfish	1,595,391	1200	1,594,191
Mussels	1,443,911	No Exports/Re-Exports	1,443,911
Gadiformes	1,400,404	No Exports/Re-Exports	1,400,404
Cod	1,343,495	2586	1,340,909
Seabream	1,199,876	160	1,199,716
Trout	1,046,693	282,359	764,334
Seabass	834,985	No Exports/Re-Exports	834,985
Tilapia	543,341	3695	539,646
Hake	387,259	No Exports/Re-Exports	387,259
Alaska Pollock	301,844	1269	300,575
Tuna	300,673	4547	296,126
Sardines	289,433	5	289,428
Sole	287,062	No Exports/Re-Exports	287,062
Mackerel	260,307	1008	259,299
Coalfish	247,671	630	247,041
Turbot	148,239	No Exports/Re-Exports	148,239
Swordfish	109,512	No Exports/Re-Exports	109,512

* top 20 in descending order of quantity traded.

species/group. Together, these generally amounted to the highest trade quantity for a given seafood by a large margin; for instance, salmon imported from Norway, Denmark and the United Kingdom (UK) alone contributed 52% to the total Swiss imports of salmon from 31 nations. In the event of discrepancies between imported quantities reported by Switzerland and quantities reported by the partner nations as exported to Switzerland, imported quantities were used in diet generation and exposure calculations, since previous studies have found them to be more reliable (Barigozzi et al., 2011; Feenstra et al., 2005). Data on exports and re-exports of seafood from Switzerland were also extracted for comparison. However, these were found to be minimal in comparison to imports (SI Table S3) and therefore were excluded from all calculations.

Although perch fell below the top 20 seafood imports (traded quantity 14842 kg/year) it was added to the list of selected species, because it is both imported and locally caught (FOEN, 2016), a combination not found for any other selected fish. This allowed us to probe whether local or imported perch contributes more to PBDE exposure. Our analysis was therefore inclusive of 23 seafood species in total; 20 imported and 3 local, with both local and imported perch included.

Whitefish, roach and perch dominate the domestic Swiss fish catch (FOEN, 2016), and hence have been included in our analysis for the domestic component of exposure calculations. Data on catch quantity (kg/year) were extracted by the Swiss Federal Office of Fisheries Statistics (FOEN, 2016). As reported, Switzerland caught 1,365,729 kg fish in 2016, contributing only approximately 2% of the country's fish intake (see Supporting Information, SI, section S1). Whitefish (845,917 kg), perch (230,246 kg) and roach (119,176 kg) were the most widely caught fish species, contributing 62%, 17% and 9%, respectively, to the total domestic catch.

To translate the imported and local seafood proportions to amounts of each species consumed we used the average annual fish consumption reported by the Swiss Federal Statistical Bureau: Production and Consumption of fish (FOEN, 2016; Mühlemann and Renggli, 2012). This is equivalent to approximately 23 g/day, assuming that consumption is equally distributed over all days and over the entire Swiss population.

2.2.2. Swiss dietary survey (menuCH)

We received access to the detailed menuCH dietary survey data published by the Swiss Federal Food Safety and Veterinary Office (SFFSVO, Swiss Federal Food Safety and Veterinary Office: National Survey menuCH 2014/2015, 2018). These data represent a single day of consumption (24-hour dietary recall) by 2000 adult participants. On average this amounted to a total fish consumption of approximately 40 g/day for all surveyed participants (consumers and non-consumers) and included the following species: salmon, cod, tuna, shrimp, trout, perch, whitefish, sardines, seabream, pangasius, plaice, herring, flounder, hake, mackerel, sole, crab, mussels, anchovies, cuttlefish, squid, crayfish, oysters, Atlantic halibut, scallops, eel, clams, lobster and whiting. We did not include any processed fish in our calculations due to the unavailability of PBDE concentrations for them.

2.2.3. Additional Origin-Based scenarios

As mentioned above, the transport of e-wastes for disposal and processing plays a key role in dispersing PBDEs into new environments (Wang et al., 2009). At the same time, e-waste receiving nations like China, Vietnam, and Indonesia are also among the major exporters of seafood to Switzerland, based on UN Comtrade trade statistics. To inspect the different dimensions of the e-waste – food trade – PBDE exposure nexus we constructed 3 different extreme scenarios: (i) consumption only of seafood imported from Norway, a country with significant contribution to seafood exports to Switzerland that is also an e-waste source country, where PBDEs may be released during product use; (ii) consumption of only seafood imported from Vietnam, which has significant seafood exports to Switzerland and is an e-waste receiving country, where PBDEs may be released during e-waste disposal and processing (Ahmed et al., 2018; FAO. The State of the World Fisheries and Aquaculture 2018-meeting the sustainable development goals, 2018) (iii) consumption of only locally produced fish from Switzerland, itself an e-waste source country. For these scenarios, 40 g of daily fish consumption by a Swiss adult weighing 72 kg was assumed, based on the average of the survey responses. PBDE concentrations in seafood from Norway did not include Norwegian whitefish since it is not imported at all, as informed by the UN Comtrade Database. For local exposures, we considered only whitefish since measured PBDE concentrations were available for Swiss whitefish, but not for perch or roach.

2.3. Global PBDE levels in seafood

We compiled global PBDE levels from the literature to translate consumption levels to exposures. PBDE concentrations in marine and freshwater species selected for exposure calculations in the current study were collected using two databases, Ei Compendex and Scopus, and two search engines, PubMed and Google Scholar. We used the search terms "PBDE OR polybrominated diphenyl ether AND fish OR market basket study OR seafood intake" Publications from 2000 through 2018 were included. Among the screened papers, only sampling locations from Asia, Africa, North America, and Europe (specifically: Bangladesh, Belgium, Chile, China, Denmark, France, Germany, Greece, Iceland, Indonesia, Italy, Japan, Norway, Netherlands, Poland, Portugal, South Africa, Spain, Thailand, Turkey, UK, USA, and Vietnam) were included for further analysis, as these regions are among the dominant exporters of consumable aquatic species to Switzerland based on the UN Comtrade Database. We included only those studies where sampling was done from either food markets or fish farms. We excluded studies where sampling was done from known contaminated sites or potential point sources (e.g. rivers/lakes near industrial areas or municipal dump sites), because these could represent a biased sample. However, due to the unavailability of any market based study reporting PBDE concentrations in the fish locally caught in Switzerland (whitefish, roach and perch), we decided to include one study reporting PBDE concentrations in whitefish caught from Swiss lakes (Zennegg et al., 2003). Refer to SI, Figure S1 for a Prisma-type flow diagram for this study.

Table 2 shows the origin-specific PBDE levels (pg/g wet weight) in

Table 2
Global PBDE values in seafood.

Seafood species	Locations sampled/Origins	PBDE congeners included in total[a]	Sampling year	ΣPBDE[b] (pg/g wet weight)	Species average ΣPBDE[c] (pg/g wet weight)
Salmon	*Norway (Hites et al., 2004; van Leeuwen et al., 2008)	1, 2, 3, 7, 8, 10, 11, 12, 13, 15, 17, 25, 28, 30, 32, 33, 35, 37, 47, 49, 66, 71, 75, 77, 85, 99, 100, 105, 116, 119, 126, 138, 140, 153, 154, 155, 166, 181, 183, 190, 191, 196, 197, 206, 207, 208, 209	2002, 2007–2008	1783	985
	Belgium (Voorspoels et al., 2007)	28, 47, 99, 100, 153, 154, 183, and 209	2005	1580	
	*USA (Cade et al., 2018; Schecter et al., 2010, 2004)	17, 28, 47, 49, 66, 77, 85, 99, 100, 119, 138, 153, 154, 183, 196, 197, 206, 207, 209	2004, 2009, 2015–2016	1058	
	Japan (Ohta et al., 2002)	28, 47, 99, 100, 153, 154	2002	835.75	
	Spain (Gómara et al., 2006)	17, 28, 47, 66, 85, 99, 100, 153, 154, 183, 184, 191, 196, 197, 209	2003–2005	251	
	Chile (Montory and Barra, 2006)	1, 2, 3, 7,10, 13, 15, 17, 25, 28, 35, 47, 49, 66, 71, 75, 77, 85, 99, 100, 116, 119, 126, 138, 140, 153, 154, 155, 156), 81, 183, 197, 203, 207, 209	2006	1460	
Shrimp/ prawn	Vietnam (Shanmuganathan et al., 2011)	47, 99, 100, 138, 153, 154, 156, 183, 206, 207, 209	2011	25,100	310
	Belgium (Voorspoels et al., 2007)	28, 47, 99, 100, 153, 154, 183, and 209	2005	61	
	*USA(Cade et al., 2018; Schecter et al., 2004)	17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183, 209	2004, 2015–2016	228	
	Japan (Ashizuka et al., 2008)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209	2004–2005	20	
	*China (Guo et al., 2010; Su et al., 2012)	17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, 209	2004–2005, 2006	111	
	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	1140	
Catfish	USA (Schecter et al., 2010, 2004)	17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183, 209	2004, 2009	779	364
	China (Su et al., 2012)	17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, 209	2006	270	
	*Vietnam (Hien et al., 2012; van Leeuwen et al., 2008)	28, 49, 71, 47, 66, 77, 100, 119, 99, 85, 126, 153, 138, 156, 184, 183, 191, 197, 196,208, 206, 209	2007–2008, 2008	229	
Mussels	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	1120	482
	USA (Cade et al., 2018)	17, 28, 47, 99, 100, 153, 154	2015–2016	398.1	
	Belgium (Covaci et al., 2005)	28, 47, 49, 66, 85, 99, 100, 153, 154, 183	2002	690	
	*Spain (Bocio et al., 2003; Domingo et al., 2008)	47, 99, 100, 153, 154, 183	2000, 2006	175	
Cod	Belgium (Voorspoels et al., 2007)	28, 47, 99, 100, 153, 154, 183, 209	2005	48	92
	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	410	
	USA (Schecter et al., 2010)	17, 28, 47, 49, 66, 85, 99, 100, 119, 138, 153, 154, 183, 196, 197, 206, 207, 209	2008	31.8	
	European market (Aznar-Alemany et al., 2017)	47, 99	2014–2015	385.2	
	Norway (van Leeuwen et al., 2008)	28, 49, 71, 47, 66, 77, 100, 119, 99, 85, 126, 153, 138, 156, 184, 183, 191, 197, 196, 208, 206, 209	2007–2008	28	
Trout	Belgium (Voorspoels et al., 2007)	28, 47, 99, 100, 153, 154, 183, 209	2005	270	976
	Switzerland (Zennegg et al., 2003)	28, 47, 99, 100, 153, 154, 183	2003	1300	
	*USA (Cade et al., 2018; Olson, 2001; Schecter et al., 2004)	17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183, 209	1996–1999, 2004, 2015–2016	4375	
	Italy (van Leeuwen et al., 2008)	28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 191, 196, 197, 206, 208, 209	2007–2008	413	
	Denmark (van Leeuwen et al., 2008)	28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 191, 196, 197, 206, 208, 209	2007–2008	355	
	Turkey (van Leeuwen et al., 2008)	28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 191, 196, 197, 206, 208, 209	2007–2008	3831	
Tilapia	*USA (Schecter et al., 2010, 2004)	17, 28, 47, 49, 66, 77, 85, 99, 100, 119, 138, 153, 154, 183, 196, 197, 206, 207, 209	2004, 2009	14	26
	*China (Meng et al., 2007; van Leeuwen et al., 2008)	28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 191, 196, 197, 206, 208, 209	2004–2005, 2007–2008	51	
	Netherlands (van Leeuwen et al., 2008)	28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 191, 196, 197, 206, 208, 209	2007–2008	27	
	Indonesia (van Leeuwen et al., 2008)	28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 191, 196, 197, 206, 208, 209	2007–2008	22.75	
Hake	Spain (Domingo et al., 2008)	47, 99, 100, 153, 154, 183	2006	221.1	221

(continued on next page)

Table 2 (*continued*)

Seafood species	Locations sampled/Origins	PBDE congeners included in total[a]	Sampling year	ΣPBDE[b] (pg/g wet weight)	Species average ΣPBDE[c] (pg/g wet weight)
Sardines	Spain (Domingo et al., 2008)	47, 99, 100, 153, 154, 183	2006	710	169
	Japan (Ashizuka et al., 2008)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209	2004–2005	130	
	Belgium (Voorspoels et al., 2007)	28, 47, 99, 100, 153, 154, 183, 209	2005	52	
Sole	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	440	731
	Spain (Domingo et al., 2008)	47, 99, 100, 153, 154, 183	2006	241.5	
	USA (Cade et al., 2018)	17, 28, 47, 99, 100, 153, 154	2015–2016	3680	
Tuna	*Japan (Ashizuka et al., 2008; Ohta et al., 2002)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209; 28, 47, 99, 100, 153, 154	2002, 2004–2005	29	55
	Spain (Domingo et al., 2008)	47, 99, 100, 153, 154, 183	2006	558.3	
	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	10	
Mackerel	Belgium (Voorspoels et al., 2007)	28, 47, 99, 100, 153, 154, 183, and 209	2005	200	876
	*Japan (Ashizuka et al., 2008; Ohta et al., 2002)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209; 28, 47, 99, 100, 153, 154	2002, 2004–2005	950	
	Spain (Domingo et al., 2008)	47, 99, 100, 153, 154, 183	2006	1123.7	
	Ireland (van Leeuwen and de Boer, 2008)	28, 47, 49, 66, 71, 75, 77, 85, 99, 100, 119, 138, 154, 183, 190, 209	2003	2100	
	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	1150	
Swordfish	Spain (Domingo et al., 2008)	47, 99, 100, 153, 154, 183	2006	977.7	978
Herring	Central North Sea (van Leeuwen and de Boer, 2008)	28, 47, 49, 66, 71, 75, 77, 85, 99, 100, 119, 138, 154, 183, 190, 209	2003	7600	6046
	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	4810	
Whitefish	**Switzerland (Zennegg et al., 2003)	28, 47, 99, 100, 153, 154, 183	2003	4500	75,000
	USA (Johansen and Olson, 2001)	99, 100	1996–1999	1,250,000	
Alaska Pollock	PBDE data unavailable within the inclusive criteria				
Seabream	Greece (Aznar-Alemany et al., 2017)	47, 99	2014–2015	4780	1157
	Japan (Ashizuka et al., 2008)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209	2004–2005	280	
Eel	Netherlands(Bakker et al., 2008)	47, 99, 100, 153, 154	2003	4160	1767
	Belgium (Covaci et al., 2005)	28, 47, 49, 66, 85, 99, 100, 153, 154, 183	2002	5525	
	Japan (Ashizuka et al., 2008)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209	2004–2005	240	
Perch	USA (Brown et al., 2006)	47, 99, 100, 153, 154	2000–2001	9301	9301
Plaice	North Sea (Aznar-Alemany et al., 2017)	47, 99	2014–2015	514.29	454
	Netherlands (Bakker et al., 2008)	47, 99, 100, 153, 154	2003	400	
Halibut	PBDE data unavailable within the inclusive criteria				
Crab	Thailand (Shanmuganathan et al., 2011)	47, 99, 100, 138, 153, 154, 156, 183, 206, 207, 209	2011	3750	1285
	China (Guo et al., 2010)	28, 47, 66, 99, 100, 138, 153, 154, 183, 209	2004–2005	440	
Clams	Japan (Ohta et al., 2002)	28, 47, 99, 100, 153, 154	2002	52.4	126
	USA (Cade et al., 2018)	17, 28, 47, 99, 100, 153, 154	2015–2016	303	
Scallop	Japan (Shanmuganathan et al., 2011)	47, 99, 100, 138, 153, 154, 156, 183, 206, 207, 209	2011	5720	1057
	USA (Cade et al., 2018)	17, 28, 47, 99, 100, 153, 154	2015–2016	195.5	
Flounder	Netherlands (van Leeuwen and de Boer, 2008)	28, 47, 49, 66, 71, 75, 77, 85, 99, 100, 119, 138, 154, 183, 190, 209	2003	15,100	777
	Japan (Ashizuka et al., 2008)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209	2004–2005	40	
Coalfish	Netherlands(Bakker et al., 2008)	47, 99, 100, 153, 154	2003	410	410
Squid	China (Shanmuganathan et al., 2011)	47, 99, 100, 138, 153, 154, 156, 183, 206, 207, 209	2011	19,420	19,420
Carp	*China (Meng et al., 2007; Su et al., 2012)	17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190, 209	2004–2005, 2006	87	575
	Belgium (Covaci et al., 2005)	28, 47, 49, 66, 85, 99, 100, 153, 154, 183	2002	3800	
Seabass	Japan (Ashizuka et al., 2008)	17, 28, 47, 49, 66, 77, 99, 100, 119, 153, 154, 183, 184, 196, 197, 206, 207, 209	2004–2005	330	330

[a] PBDE congeners measured by the study.
[b] Average total PBDEs reported (pg/g wet weight).
[c] PBDEs (pg/g wet weight) as geometric mean of values reported in column 4, rounded to the nearest whole number.
* Multiple studies reporting PBDE concentrations from the same origin, geometric mean concentration used.
** Swiss whitefish data from Swiss lakes, not market.

seafood used in our analysis. Origin-based, species-specific exposure estimates were calculated using origin-specific PBDE concentrations (Table 2, column 4). In cases where a seafood species was associated with more than one concentration from the same origin (e.g. salmon from USA and Norway, shrimp from USA and China, catfish from USA and Vietnam, mussels from Spain, trout from USA, tilapia from USA and China, tuna from Japan, mackerel from Japan and carp from China), we used the geometric mean of PBDEs across a single origin in the final exposure calculations for that origin. For exposures where we did not consider origins, we used the geometric mean of PBDEs over all the available origins. For example, species average PBDE levels for salmon were calculated as the geometric mean of values reported in Norway, Belgium, USA, Japan, Spain and Chile (985 pg/g wet weight), which was then used for calculating PBDE exposures from salmon intake irrespective of origin (termed "species-specific but not origin-specific" exposure estimates).

The total PBDE concentrations for most studies (92%) were predominantly congeners 28, 47, 99, 100, 153, and 154. Since the congener profiles were in general similar across the selected studies, we used the sum of all PBDE congeners, referred to hereafter as total PBDEs. However, high BDE-209 concentrations were detected in a few studies (Ashizuka et al., 2008; Gómara et al., 2006; Schecter et al., 2010; Shanmuganathan et al., 2011). This could potentially bias results for total PBDE exposure, since BDE-209 is considered less bioaccumulative and toxic than lower-brominated congeners. The only study for which this may be a concern is in Vietnamese shrimp, where BDE-209 was 46% of the total reported concentration (Shanmuganathan et al., 2011), and this was also a seafood-origin pair with one of the highest total PBDE concentrations. For the other studies in which BDE-209 was a dominant congener, catfish and tilapia from the USA and salmon from Spain, the total PBDE levels in these particular seafood-origin pairs were relatively low, as shown in Table 2. In all cases, for non-origin-specific scenarios the use of geometric mean values to represent species averages minimized any undue influence from high BDE 209 contributions. For origin-specific calculations, the presence of high amounts of BDE 209 would only substantially affect exposures attributed to Vietnamese shrimp. Other congeners were frequently below the limit of detection.

The primary objective of the literature review was to find the PBDE levels measured in origins and species of interest. However, PBDE data were missing for some combinations of species and origins. In order to estimate PBDE concentrations for all fish and all origins considered in our analysis we made a number of assumptions. In the absence of data for a particular combination of origin country and seafood type we used either lipid-normalized PBDE concentrations (ng/g wet weight/lipid percent) for the same fish but another region in close proximity (FAO, 2016, 2015) or PBDE values reported for the same origin country but for another fish having similar taxonomy to the fish of concern. Refer to Table 3 column 3 for the PBDE data substitutes (if used) within seafood species or origins and Table 3 column 4 for the lipid-normalized concentrations which were used for extrapolations across species. For exposure calculations we used wet weight concentrations (Table 3; column 5), since these are more representative of fish as consumed. Refer to SI, section S3 (Table S4 and Table S5) for details on species- and origin-specific assumptions and extrapolations.

2.4. Exposure calculations

PBDE exposures for the trade-data based approach were calculated using Eq. (1) for both imported (20 species from top 3 exporters) and locally produced (3 species) seafood, as well as overall imported seafood (34 species; average PBDE concentrations over all origins). Total exposure (ΣE), whatever the species or origin scenario, is reported in ng PBDEs/kg body weight (bw)/day. Calculations assumed an average Swiss adult weighs 72 kg (menuCH survey average weight of surveyed individuals). Because this is the trade-data based approach that could be used in the absence of specific reported consumption data (i.e. without a dietary survey available), we used the national-statistics-based estimate of 23 g of fish consumed daily per person in Switzerland (C_d; *dailyconsumption*) (FOEN, 2016; Mühlemann and Renggli, 2012).

$$\sum E = \sum_{i=1}^{n} \frac{\left(\frac{Q_i}{Q_t} \times 100\right) \times p \times C_d \times \sum PBDE}{BW} \tag{1}$$

where $\frac{Q_i}{Q_t}$ = proportion of total imports **(%)**; $\left(\frac{Q_i}{Q_t} \times 100\right) \times p$ = proportion of diet (%) and $\left(\frac{Q_i}{Q_t} \times 100\right) \times p \times C_d$ = daily seafood consumption $\left(\frac{g}{day}\right)$

Here, $\sum PBDE$ refers to the total (sum of individual PBDE congeners) average PBDE concentration in a particular seafood species. Although different PBDE congeners may be included in these sums, based on what was measured in specific studies cited in Table 2, we will refer hereafter only to total PBDEs. Q_i is the quantity imported or locally produced (in units of kg/year) for a species i ranging from 1 to n, and the total quantity imported is Q_t which is 47,969,288 kg for 2016 (SI, table S1). The quantity $\frac{Q_i}{Q_t}$ for a single seafood species represents its percent of total imports. When multiplied by the parameter p, this yields the proportion occupied by each seafood species with respect to total seafood consumption. Here, the parameter p takes the value of 0.98 or 0.02 to represent the percent of the Swiss seafood diet that is composed of imports or local products, respectively.

For the dietary survey-based approach, we calculated the PBDE exposure as the product of reported daily consumption by species and the average ΣPBDE concentration in that species (Table 2, column 5) calculated as the geometric mean of PBDE concentrations across all origins (because the survey did not include any information on seafood origin). Calculations were done using an average Swiss body weight of 72 kg as reported in menuCH. We also calculated PBDE exposures for each person (survey correspondent) for the fish species being consumed (here we used the individual body weights and amounts of seafood consumed), from which we constructed the distribution of PBDE exposures across individual fish consumers in Switzerland.

Note that all exposure estimates are for Swiss adults. The exposures were compared to available Chronic Oral Reference Doses (RfD) for PBDEs, representing the maximum acceptable oral dose in units of mg dose per kg body weight per day. We used a range of RfDs for PBDEs (100 ng/kg bw/day to 7000 ng/kg bw/day) representing the allowable doses for the most abundant PBDE congeners (penta, hexa, octa and deca-BDE) as suggested by the EPA (EFSA, 2011; EPA-a, 2014; EPA-b, 2018).

2.5. Uncertainty assessment

Since our analysis is based on a number of assumptions, we considered the uncertainty that could be introduced by each component of our exposure estimation.

2.5.1. Diet generation

The trade-estimated seafood diet we generated is simplified by including only the top 3 exporters (origins) for each speies and only the top 20 seafood imports (species). Using the sum of all imports and total fish import data, we account for fish species or quantities neglected by our analysis and investigate whether this introduces significant uncertainty to the outputs.

2.5.2. Daily fish intake

We assume the reported average daily consumption of 23 g of fish per person as a part in the Swiss diet (FOEN, 2016; Mühlemann and Renggli, 2012) all consists of fresh or frozen whole or fillet forms of imported and domestic fish. We further consider only the top 3 largest exporters of each seaffod type to Switzerland and 23 types of seafood

Table 3
Species–origin combinations and ΣPBDE data used as input for analysis.

Seafood	Top exporters	Origin - species source for PBDE data used[a]	Lipid normalized ΣPBDE[b]	ΣPBDE[c]	Lipid %
Salmon	Norway	Norway – Salmon	5.31	1.78	33.59 (Hites et al., 2004)
	Denmark	Belgium – Salmon	12.12	1.58	13 (Voorspoels et al., 2007)
	UK	Belgium – Salmon	12.12	1.58	13 (Voorspoels et al., 2007)
Shrimp/prawn	Vietnam	Vietnam – Shrimp	1930.77	25.10	1.3 (Voorspoels et al., 2007)
	Bangladesh	China – Shrimp	13	0.11	1.3 (Su et al., 2012)
	Belgium	Belgium – Shrimp	4.69	0.06	1.3 (Voorspoels et al., 2007)
Catfish	Vietnam	Vietnam – Catfish	5.78	0.22	3.8 (Minh et al., 2006)
	Netherlands	Netherlands – Herring	28.29	4.81	17 (Bakker et al., 2008)
	Italy	Spain – Sardines	10.00	0.71	7.1 (Voorspoels et al., 2007)
Flatfish	Netherlands	Netherlands – Sole	44.00	0.44	1 (Bakker et al., 2008)
	Poland	Netherlands – Sole	44.00	0.44	
	Germany	Netherlands – Sole	44.00	0.44	
Mussels	Netherlands	Netherlands – Mussels	61.00	1.12	2 (Bakker et al., 2008)
	France	Spain – Mussels	6.25	0.17	2.8 (Bocio et al., 2003)
	Italy	Spain – Mussels	6.25	0.17	2.8 (Bocio et al., 2003)
Gadiformes	Iceland	Norway – Salmon	5.31	1.78	33.59 (Hites et al., 2004)
	France	Spain – Swordfish	13.81	0.98	7.08 (USDA, 2018)
	Denmark	Belgium – Salmon	12.12	1.58	13 (Voorspoels et al., 2007)
Cod	China	China – Tilapia	0.69	0.051	7.3 (Luo et al., 2007)
	Portugal	Spain – Swordfish	13.84	0.98	7.08 (USDA, 2018)
	Denmark	European market – Cod	107	0.385	0.36 (Aznar-Alemany et al., 2017)
Seabream	Greece	Greece – Seabream	179.00	4.78	2.67 (Aznar-Alemany et al., 2017)
	France	Greece – Seabream	179.00	4.78	
	Italy	Greece – Seabream	179.00	4.78	
Trout	Italy	Italy – Trout	13.32	0.41	3.1(Voorspoels et al., 2007)
	France	Italy – Trout	13.32	0.41	
	Germany	Belgium – Trout	8.71	0.27	
Seabass	France	Spain – Hake	31.42	0.22	0.7 (Murray and Burt, 2001)
	Italy	Spain – Hake	31.42	0.22	
	Greece	Spain – Hake	31.42	0.22	
Tilapia	Vietnam	Indonesia – Tilapia	0.31	0.02	7.3 (Luo et al., 2007)
	China	China – Tilapia	0.69	0.051	7.3 (Luo et al., 2007)
	Indonesia	Indonesia – Tilapia	0.31	0.02	7.3 (Luo et al., 2007)
Hake	South Africa	Spain – Hake	31.59	0.22	0.7 (Murray and Burt, 2001)
	Portugal	Spain – Hake	31.59	0.22	
	Germany	European Market – Cod	107	0.385	0.36 (Aznar-Alemany et al., 2017)
Alaska Pollock	China	China – Tilapia	0.69	0.051	7.3 (Luo et al., 2007)
	Germany	European Market – Cod	107	0.385	0.36 (Aznar-Alemany et al., 2017)
	Denmark	European Market – Cod	107	0.385	
Tuna	Vietnam	Japan – Tuna	1.89	0.02	1.1 (Ohta et al., 2002)
	Netherlands	Netherlands – Tuna	1.00	0.01	1 (Bakker et al., 2008)
	UK	Netherlands – Tuna	1.00	0.01	1 (Bakker et al., 2008)
Sardines	Portugal	Spain – Sardines	10	0.71	7.1 (Voorspoels et al., 2007)
	France	Spain – Sardines	10	0.71	
	Spain	Spain – Sardines	10	0.71	
Sole	Netherlands	Netherlands – Sole	44	0.44	1 (Bakker et al., 2008)
	France	Spain – Sole	24	0.24	
	UK	Netherlands – Sole	44	0.44	
Mackerel	Spain	Spain – Mackerel	7.49	1.12	15 (van Leeuwen and de Boer, 2008)
	Portugal	Spain – Mackerel	7.49	1.12	
	Netherlands	Netherlands – Mackerel	10.45	1.15	11 (Bakker et al., 2008)
Coalfish	Germany	Netherlands – Coalfish	41	0.41	1 (Bakker et al., 2008)
	China	China – Tilapia	0.69	0.51	7.3 (Luo et al., 2007)
	Poland	Netherlands – Coalfish	41	0.41	1 (Bakker et al., 2008)
Turbots	Netherlands	Netherlands – Sole	44	0.44	1 (Bakker et al., 2008)
	Spain	Spain – Sole	24	0.24	
	France	Spain – Sole	24	0.24	
Swordfish	Sri Lanka	Data unavailable			
	Italy	Spain – Swordfish	13.81	0.98	7.08 (USDA, 2018)
	France	Spain – Swordfish	13.81	0.98	
Perch	Netherlands	Netherlands – Herring	28.29	4.81	17 (Bakker et al., 2008)
	Germany	Netherlands – Herring	28.29	4.81	
	Indonesia	Data Unavailable			
	Domestic	Netherlands – Herring	28.29	4.81	17 (Bakker et al., 2008)
Whitefish	Domestic	Switzerland – Whitefish	103.45	4.50	4.35 (Zennegg et al., 2003)
Roach	Domestic	Netherlands – Herring	28.29	4.81	17 (Bakker et al., 2008)

[a] Refer to SI section S3; Tables S4-5 provide details of assumptions for species and PBDE combinations used; [b]in units of ng/g lipid weight; [c]in units of ng/g wet weight.

(local and imported) by weight. To assess if this point of uncertainty could be relevant, we calculated the daily consumption based only upon the quantity of imported and locally produced fish.

$$C_d^* = \frac{Q_{(Im+Lp)}}{P} \tag{2}$$

The analysis based on fish consumption (C_d^*) was given by the ratio of the total fish quantity [imported (im) and locally produced (lp), $Q_{(Im+Lp)}$;$kg/year$] and the population of Switzerland in the same year (P;million people). This was compared with the reported fish consumption (C_d) and any deviations were studied. Fish forms not included in our analysis (e.g. processed fish, fish products etc.) were considered responsible for any observed asymmetry in daily fish consumption.

2.5.3. PBDE concentration in fish

As mentioned earlier, we use assumptions to fill PBDE data gaps, which included estimating PBDE concentrations in target fish from data for other fish from similar origins. When comparing across species, we used lipid-normalized total PBDE concentrations (ng/g lipid weight), which we could convert back and forth from fresh weight for exposure calculations using Eq. (4).

$$\sum PBDE = \frac{\frac{ng\ PBDE}{g\ fish,\ \text{wet weight}}}{\frac{g\ lipid}{g\ fish,\ \text{wet weight}}} \tag{4}$$

3. Results and discussion

3.1. Trade-based seafood diet

The Swiss seafood diet constructed using data from the UN Comtrade database and national statistics on domestic catch is shown in Fig. 1. Combined with population-level consumption statistics this suggests that, on average, the Swiss population consumes around 10 g/day of salmon, shrimp and cod alone, out of the total 23 g/day. Closer analysis of the top exporters to Switzerland indicates Vietnamese shrimp was the most consumed seafood type from a single exporting country, followed by Vietnamese catfish and Norwegian salmon. Native whitefish was also among the top 10 most consumed fish.

3.1.1. Sensitivity and uncertainty related to diet generation

Our trade-based analysis considers only the top 20 fish and their top 3 origins. These in total made up 20,919,367 kg in 2016, contributing 44% to the total imports. This implies that the remaining 56% of imports (species imported in smaller quantities and exporters beyond the top 3), collectively contribute a significant proportion to the Swiss seafood diet, adding uncertainty to our analysis. However, our approach could identify the most important traded commodities and, even for this restricted set, identification of species- and origin-appropriate PBDE data was a major challenge.

3.1.2. Sensitivity and uncertainty related to daily fish intake

Based only on the total imports for selected fish commodities (SI Section S2) and locally caught fish (FOEN, 2016), daily fish

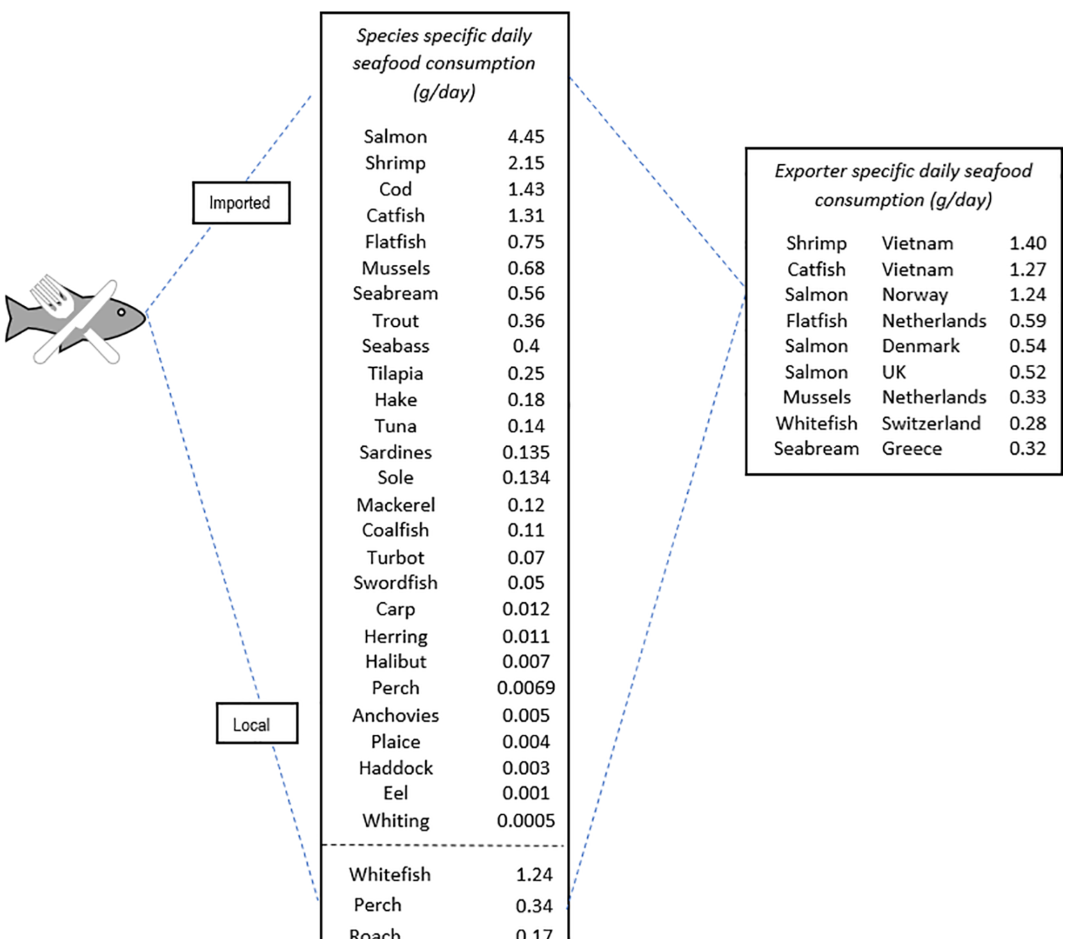

Fig. 1. Species- and origin-specific seafood consumption in Switzerland based on international trade and domestic catch data.

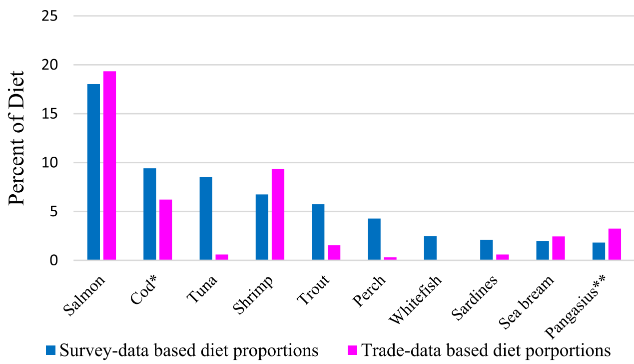

Fig. 2. Proportion of most consumed seafood species based on the dietary survey (blue bars) compared to proportions based on trade data and local production (pink bars). *Cod also includes Alaska pollock and Gadiformes. **UN Comtrade Database reports pangasius within the catfish category. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

consumption calculated using Eq. (2) amounted to 16.5 g per person daily. This is less than the value of 23 g (from total annual seafood consumption for the entire population) used as an input for the trade-based exposure calculations. The missing 7 g represents the species and/or origins not included in our analysis.

3.2. Seafood diet based on direct diet survey

Most of the commonly consumed seafood species identified using the trade data were also found via the menuCH survey. Fig. 2 shows proportions of seafood commodities most consumed in Switzerland according to the survey compared with those estimated using trade data. Refer to SI section S4 for a complete list of seafood species with their daily consumption and proportion of diet for both the survey-based (Table S6) and trade-based (Table S7) diets.

Although the annual average statistics-based seafood consumption (23 g) and 24-h recall survey-based seafood consumption (40 g) differ in total amount, a comparison of the seafood diet structure shows strong similarities in the proportions occupied by various seafood species. As anticipated according to the trade-data-based diet, salmon was the most consumed fish in the country. Our results show that in the absence of available dietary data for a population, widely available food import data and national production statistics can serve as effective tools for constructing an estimated diet.

3.3. Input data for exposure analysis

Table 3 shows the list of selected fish (imported and domestic) and the mean ΣPBDE (ng/g wet weight) reported in them by origin. Table 3 also provides the lipid-normalized ΣPBDE concentrations used for species substitutions. No species- and region-specific data were

Table 4
Origin – Specific PBDE Exposures based on Trade Data.

Fish Type	Top Exporters	Percent of Diet	PBDE Exposure (ng/kg bw/day)
Shrimp/prawn	Vietnam	6.12	0.4914
Salmon	Norway	5.37	0.0306
Seabream	Greece	1.41	0.0216
Whitefish	Domestic	1.24	0.0178
Salmon	Denmark	2.35	0.0119
Salmon	UK	2.30	0.0116
Seabream	France	0.40	0.0063
Gadiformes	Iceland	1.09	0.0062
Seabream	Italy	0.35	0.0054
Mussels	Netherlands	1.45	0.0052

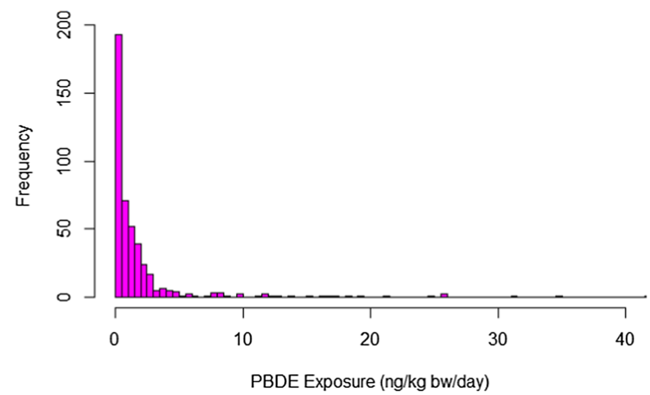

Fig. 3. PBDE exposure range across fish consumers in Switzerland.

available for swordfish from Sri Lanka and perch from Vietnam, so they were not included in final exposure calculations.

3.4. PBDE exposure calculations

3.4.1. Trade-based approach

Calculated PBDE exposures from the trade-based diet are shown in Table 4. The table shows the top 10 exposure values for imported or domestic fish and their origins (for a complete list see SI Table S9), indicating that shrimp imported from Vietnam contributes the most to PBDE exposure in the Swiss population (75% of the total exposures), congruent with the fact that it is exported in largest quantities. This is contrary to exposures as low as 0.004 ng/kg bw/day from Vietnamese catfish which, even after being the second-highest exported quantity, contributes less than many other seafood commodities (SI Table S10) due to low reported PBDE concentrations (Table 2). European exporters were also found to have major contributions to PBDE exposures, as they are among the largest exporters of seafood to Switzerland. It is notable that domestic whitefish is also among the highest contributors to exposures contributing 3 percent to the total exposure estimates. Tilapia from Indonesia, sole from UK, and tuna from Vietnam and the UK were found to have the lowest species- and origin-specific PBDE contributions (Table S9).

3.4.2. Survey-based approach

The PBDE exposures estimated across the surveyed fish consumers in Switzerland ranged between 0.011and 43.42 ng/kg bw/day (Fig. 3). The median exposure (50th percentile) is 0.68 ng/kg bw/day. In comparison, the calculated origin-specific trade-data based exposure is 0.65 ng/kg bw/day, surprisingly close to this value. This suggests the trade data are in fact a good proxy for the average exposure. We also find that the 95th percentile of the surveyed Swiss population is exposed to PBDE levels as high as 8.5 ng/kg bw/day. The analysis of survey data thus allows us to capture exposures of the more at-risk sectors of the population.

Species-specific but not origin-specific PBDE exposures were estimated to be 0.15 ng/kg bw/day using trade data. One reason for this low number is the fact that when we average PBDE concentrations across all origins, the overall PBDE concentration is reduced. To illustrate, Fig. 4 shows the PBDE concentrations reported globally in salmon, shrimp and mussels, as well as their geometric means. Fig. 4 also highlights the PBDEs that were used in our analysis. For instance, origin-based exposure estimates for salmon only account for Norway and Belgium, with individual values of 1783 pg/kg bw/day and 1580 ng/kg bw/day respectively. On the other hand, total trade-based estimates for salmon account for the average PBDE level of 985 ng/kg bw/day across Norway, Belgium, Chile, USA, Japan and Spain. We could therefore conclude that quantifying exposures according to

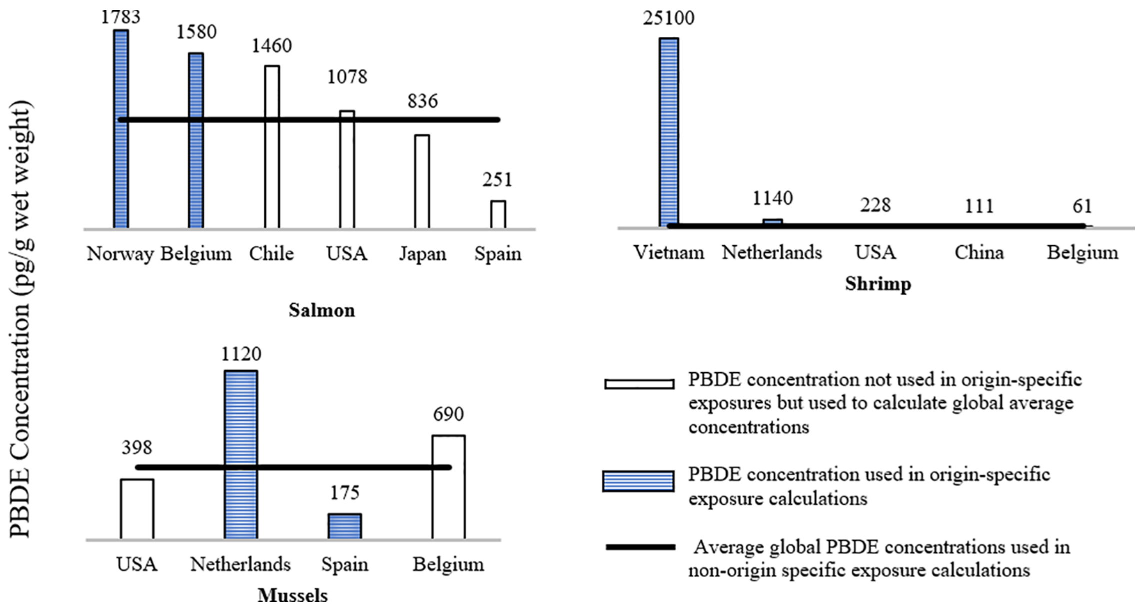

Fig. 4. Difference between PBDE data used for species-specific origin-specific vs non-origin-specific exposures.

Table 5
Species-specific trade-based diet versus survey-based diet PBDE exposures.

Seafood Species	Trade-based PBDE Exposure (ng/kg bw/day)	Survey-based PBDE Exposure (ng/kg bw/day)
Salmon	0.0612	0.0986
Whitefish	0.0181	0.0624
Perch	0.0109	0.221
Shrimp	0.0093	0.0116
Seabream	0.0089	0.0127
Flatfish	0.0076	0.0036*
Trout	0.0067	0.0311
Catfish	0.0066	0.0036
Mussels	0.0045	0.0018
Roach	0.0033	Not reported as consumed

* Value for flounder.

origins gives us a more realistic understanding of a particular community's risk from PBDE exposure.

3.4.3. Comparison of Trade-based and Survey-based approaches

Finally, we compare PBDE exposures by seafood species (irrespective of origin) based on geometric means of the global PBDE concentrations using both trade-based and survey-based diets. Table 5 shows the calculated PBDE intakes (top 10 exposures only based on trade-based diet and the corresponding exposures for the survey-based diet; refer to SI Tables S8 – S9 for a complete list).

Salmon, perch, shrimp, trout and whitefish appear to be the most contaminated species for both the trade-based and survey-based diets. The high exposure in the survey-based diet is driven by higher amounts of seafood eaten by some consumers that pushes up the average exposure from each species. This highlights a potential pitfall of using general annual statistics, since diets vary within populations and hence the risk of PBDE exposure may increase for groups that eat more trout, shrimp, perch or salmon (all having higher PBDE exposures) or have above average daily fish consumption. This was also illustrated by the distribution of PBDE exposures in survey respondents (Fig. 4). However, all the exposures were found to be lower than the RfD range of 100 ng/kg bw/day to 7000 ng/kg bw/day.

3.5. Origin-Specific scenarios

Table 6 shows PBDE exposures estimated for our three origin-specific scenarios. When considering the exporting e-waste source and sink countries selected for this analysis, seafood imports from Vietnam contribute most to PBDE exposure of the Swiss population. Although lower than the allowable reference dose range, these exposures surpass even the total PBDE exposure calculated using the top 3 exporters (0.65 ng/kg bw/day). The scenarios revealed that if Swiss adults consume only seafood imported from an e-waste sink country, as in the case of Vietnam, exposure can be as high as 7 ng/kg bw/day, which is very close to the PBDE exposure for high-risk consumers informed by the survey data (95th percentile, 8.5 ng/kg bw/day) Hence, origin-specific scenarios help provide us with a worst-case perspective on PBDE exposures.

The impact of Norwegian seafood alone was also found to be very close to the median PBDE exposures of 0.68 ng/kg bw/day as reported

Table 6
Scenario-specific PBDE exposures for Swiss adults.

Parameters	Consumption of fish originating from e-waste source/ origin	Consumption of fish originating from e-waste dumping site/sink	Consumption of only local fish
Region	Norway	Vietnam	Switzerland
Species Included	Salmon and cod	Shrimp and catfish	Whitefish
ΣPBDE concentration in selected seafood	Salmon (1.783); cod (0.028) ng/g wet weight	Shrimp (25.100); catfish (0.779) ng/g wet weight	4.50 ng/g wet weight
PBDE Exposure from consuming the scenario specific species	0.50 ng/kg bw/day	7.18 ng/kg bw/day	2.5 ng/kg bw/day

Note: All exposure calculations are for an adult weighing 72 kg and consuming 40 g of selected seafood daily in equal proportions. The three scenarios thus assume a daily intake of 20 g each of salmon and cod, 20 g each of shrimp and catfish, or 40 g of whitefish, respectively.

by the survey data. Norway recycles almost 80% of its e-waste in-state (Streicher-Porte, 2006), which reduces environmental impacts of e-waste exports, but also maintains the PBDEs in these products in circulation. Hence, the risk of exposures within Norway continues.

The consumption of only domestic whitefish (40 g per day) would lead to a lower PBDE exposure than consumption of seafood from Vietnam (20 g each of shrimp and catfish). This is consistent with the fact that Switzerland, like many other European nations, recycles only around 25% of its e-waste; the remainder is either untraced or sent out of state for disposal or processing (Streicher-Porte, 2006). Our analyses illustrate how choices around international seafood trade could result in increases or reductions in PBDE exposure, depending on the origins considered.

4. Conclusions

PBDE exposures as high as 8.5 ng/kg bw/day (for the 95th percentile of the population) were found for the survey-based diet, where consumption amounts reflect more realistic averages for adult seafood consumers than the per capita consumption reported by national statistics. PBDE exposures from the trade-data based diet (origin-specific measures) were found to be very close to the median exposures of 0.68 ng/kg bw/day for the Swiss population, indicating that the per capita food balance derived from trade data is a good proxy for the average exposure, even though it could not account for the population variability captured by the survey data. However, in the absence of dietary survey data, the key species predicted using trade data were found to be consistent with those reported by Swiss consumers. Our analysis showed that tuna, sole and tilapia imported from the UK and Indonesia, were least contaminated with PBDEs. Vietnamese shrimp/prawn, Norwegian salmon and Swiss whitefish were found to be the most contaminated species–origin combinations. From the perspective of import-related exposures, our analysis identified Vietnam, Italy, Norway, and Greece as potential hot spots in the international seafood trade network, playing pivotal roles in bringing diet-borne PBDEs to Switzerland. Thus, if of sufficient quality, readily available trade data can provide important insights when specific data are lacking, and at the same time provides important information on the origin of foods.

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CRediT authorship contribution statement

Megha Bedi: Methodology, Formal analysis, Investigation, Writing - original draft, Visualization. **Natalie Von Goetz:** Conceptualization, Methodology, Writing - review & editing. **Carla Ng:** Conceptualization, Methodology, Writing - review & editing, Supervision.

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 material

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