A comprehensive assessment of endocrine-disrupting chemicals in an Indian food basket: Levels, dietary intakes, and comparison with European data[☆]

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ABSTRACT

Endocrine-disrupting chemicals (EDCs) in diet are a health concern and their monitoring in food has been introduced in the European Union. In developing countries, EDC dietary exposure data are scarce, especially from areas perceived as pollution hotspots, including industrialized countries like India. Several persistent organic pollutants (POPs) act as EDCs and pose a pressure to human health mainly through dietary exposure. In the present study, a range of organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), dioxins and furans were measured in several food items collected from Indian urban (Delhi) and peri-urban (Dehradun) areas. Food basket contamination data were used to estimate EDC dietary exposure and compare it with that of the average European population estimated from available monitoring data. All the target contaminants were found in most food items, especially in dairies and meat products. OCPs were the main contributers to the measured EDC contamination. Food supplied to Delhi's markets had higher EDC contamination than that supplied to the peri-urban market in Dehradun. Despite lax compliance and control measures, Indian dietary exposure of OCPs and PBDEs were comparable with that of Europe and were lower for PCBs and dioxins. Higher meat consumption in Europe only partly explained this pattern which was driven also by the higher EDC residues in some European food items. A substantial part of endocrine disrupting potential in the diet derives from food and animal feeds internationally traded between developed and developing countries. With increasingly globalized food systems, internationally harmonized policies on EDC content in food can lead to better protection of health in both these contexts.

1. Introduction

Endocrine-disrupting chemicals (EDCs) include a broad group of compounds with a potential for impairing the hormonal system in vertebrates (Diamanti-Kandarakis et al., 2009). Diet is an important path of EDC exposure to organisms at the upper trophic levels, including humans. Among EDCs, several persistent organic pollutants (POPs) have been under scientific and regulatory screening for being ubiquitous in the environment and human samples. These include polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins/-furans (PCDDs/Fs), several organochlorine pesticides (OCPs) (including dichlorodiphenyltrichloroethane (DDT), hexachlorobenzene (HCB) and pentachlorobenzene (PeBC)), and polybrominated diphenyl ethers (PBDEs) that are regulated under the Stockholm Convention (SC). Although the production and use of these POPs are internationally banned or restricted, they have still active sources such as obsolete

materials, old stockpiles or improperly managed waste repositories (Breivik et al., 2002; Sharma et al., 2014a). Moreover, in some developing countries (Including India), DDT is allowed for malaria vector control (UNEP, 2017) and some other OCPs may currently be illegally used. Degradation of POPs in the environment is slow and a substantial fraction of these pollutants emitted in the past recirculate in the environment, ultimately reaching the food web (Nizzetto et al., 2010) and posing a pressure to biota and humans.

There are both scientific and societal demands to conduct monitoring of human exposure to POPs, to enable effective chemical management and the fulfillment of sustainable development goals on human health. Routine monitoring programs for POPs in food are implemented in some developed countries (EFSA, 2014). For example, the 315/93/EEC and 1881/2006 regulations set the frame for controlling chemical contamination in food in the European Union (EU) and give mandate to national governments to carryout routine monitoring of several priority POPs in food products. In developing countries, measures to monitor and reduce human dietary exposure to POPs may be less common, even though results from the WHO/UNEP global survey indicate that some industrialized developing countries, including India, are hotspot of human exposure (van den Berg et al., 2017).

India hosts nearly 18% of the world population with a rapidly changing socioeconomic context, whereby traditional agriculture and traditionally processed food are progressively being replaced with intensive food production systems and industrially processed food. Data on environmental contamination and human exposure on key EDCs are scarce and fragmentary in India. Available reports focusing on POPs have consistently indicated high contamination in the environment and humans in India (Chakraborty et al., 2010, 2013; 2018; Devanathan et al., 2012; Kannan et al., 1992; Sharma et al., 2014a; van den Berg et al., 2017). So far, only a few studies reported concentrations of EDCs in Indian food basket (Battu et al., 2005; Sharma and Parisi, 2016) presenting a seminal yet partial depiction of dietary exposure. Systematic assessments of exposure and health risks of many priority EDCs (e.g. PBDEs) have never been conducted in India. Measuring POPs in the diet through the analysis of food basket contamination is therefore an important step towards understanding human exposure to POPs and guide policies toward an effective protection of human health.

The aim of this research study was to deliver the first comprehensive quality-assured assessment of human dietary exposure to EDCs in India and compare results with dietary exposure of European population. The research was carried out through implementing a large pilot study focusing on food items available from the markets of a cosmopolitan Indian city (Delhi) feeding a large urban population and from peri-urban areas of Dehradun city (Uttarakhand state, Northern India) feeding largely rural population. The assessment distinguishes exposure of groups of people of different age groups and with different dietary choices (vegetarianisms and non-vegetarianism). A secondary objective of the study was to compare EDC exposure of the urban population with that of a reference rural population. This comparison was conceived for enabling an analysis of patterns of dietary exposure along an urban-rural socioeconomic gradient and an assessment of the representativeness of the Delhi's scenario for a broader Indian context.

2. Methods

2.1. Reference sites and data

Food markets in Delhi (hereafter referred as urban market) were chosen to collect food items consumed by the urban population. Delhi has a metropolitan population of over 18.6 million people receiving food supplies from all over India, including traditional and industrially processed stocks (United Nations, 2018). In recent decades, this city has experienced intense urbanization, receiving estimated 1000 migrants belonging to different ethnic groups daily. This diversity is reflected in food choices and patterns (Diehl et al., 2019). Delhi also hosts the largest

wholesale Asian food market (Azadpur mandi), from where the food items are supplied to different parts of India as well as to international markets (Negi and Anand, 2018). The diversity of food consumption in Delhi is a good reflection of that of the entire country, making it a model study area for investigating Indian urban population's dietary exposure to EDCs.

Food collected in the markets of the peri-urban area of Dehradun city in the State of Uttarakhand (Himalayan region) was selected to represent a rural case study. This was done to look for the existence of geographical and/or socioeconomic gradients of dietary exposure or, conversely to assess whether data collected in Delhi were also representative of a rural scenario. In Dehradun's peri-urban markets, food is essentially supplied from local farms operating traditionally. In addition, this market feeds mostly rural population (hereafter referred as rural market) and has shorter food supply chains compared to those from metropolitan cities feeding urban population, and typically does not rely on application of preservatives and pesticides to increase yields and shelf-lives of food items (Halder and Pati, 2011).

2.2. Selection, collection, and pre-treatment of food items

The most frequently consumed food items by food categories (e.g. fruits, vegetables, pulses, cereals, dairies, egg, fish, and meat) were selected based on data from a survey conducted in 2018 by the Quality Council of India. In addition, the National Sample Survey Office (NSSO) of the Ministry of Statistics and Programme Implementation was also referred to as a meta-data to collect information on food habit and dietary intakes of different food items (MoSPI, 2014).

In Delhi, samples of food items were collected from wholesale food markets and supermarkets in four zones (North, South, East, and West-Delhi, see the map in supplementary data (Fig. S1; Table S1)). For all food items, representative samples were collected from wholesale markets and supermarkets. In case the selected food item was being sold under different brand names in a supermarket, a composite sample for the desired amount was prepared. Different collection and homogenization process were applied to liquid and solid/semi solid food items. Liquid samples included drinking water and milk. At least 8 samples of 1 L packaged (bottled) and municipal drinking water were collected in pre-cleaned glass bottles from shops and tap-points distributed in each of the four zones. Samples from one zone were mixed to make one composite sample (1 L) from each zone for tap and bottled water, separately. Similarly, milk-packages of 500 ml were collected from at least four milk-booths and supermarkets in each zone and mixed to make a zone-integrated sample.

Solid and semisolid food items were collected from at least five shops in wholesale food markets and five supermarkets and mixed in equal wet mass ratios to obtain zone-aggregated samples. All food items were collected in September 2018 (except for winter vegetables (cabbage, cauliflower, spinach) and fruit (orange) which were collected in February 2019). In total, 24 aggregated samples of the different food items and two types of drinking water samples (municipal supplied and packaged) were collected from the each zone in Delhi.

The same typologies of samples were collected from traditional street shops in the reference rural market in Dehradun city in March 2019 (SM, Fig. S1). Among the food items collected from Dehradun, only pulses and mango were not locally produced. The quantity and description of all the collected food item are presented in Table 1.

2.3. Target compounds

In total, 13 OCPs (*o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDT, *p,p'*-DDT, α-HCH, β-HCH, γ-HCH, δ-HCH, ε-HCH, HCB, and PeCB), 6 indicator PCBs (PCB 28, 52, 101, 138, 153, and 180) and 12 dl-PCBs (PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189), 7 PCDDs (2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, and

Table 1
Description of the collected food items from an Indian food basket and their estimated per capita daily consumption (adopted from the survey of NSSO (MoSPI, 2014)).

Food category	Food item	Sample amount	Description	Per capita estimated consumption (g/person/day)	
				Rural	Urban
Drinking water	Packaged (bottled)	1 L from each zone for each type of water	Supply water was collected from taps of the households or community drinking water tap. Packaged water was collected from local shops (or supermarket)	2500 (ml)	2500 (ml)
	Municipal supplied				
Fruits	Mango	200 g from each zone	Whole product after removal of stems and seed	3.2	5.3
	Apple		Whole product after removal of stems and seeds	3.4	6.6
	Banana		Whole product after removal of stems and peel	17.4	22.9 [a]
	Watermelon		Edible part	1.2	2.9
	Orange		Edible part	4.9	7.4 [b]
Vegetables	Tomato		Whole product	19.6	28.6
	Potato		Whole product after removal of tops and soil	63.9	61
	Onion		Mature bulbs after removal of easily detachable skin and soil	27.9	31.5
	Okra (Ladies' fingers)		Edible part	6.5	9.2
	Cabbage		Edible part	7.6	7.2
	Cauliflower		Edible part	18.6	18.3
	Spinach		Edible part	27.4	16.7
Pulses	Mung beans (*Vigna radiata*) (*Moong*)	250 g from each zone	Whole product	1.2	4.6
	Red lentils (*Lens culinaris*) (*Masoor*)			6.3	4.9
	Pigeon pea (*Cajanus cajan*) (*Arhar*)			5.5	6.9
Cereals	Wheat		After the removal of the hull from paddy	230	182
	Rice			165	69
Dairy products	Milk (packaged)	200 ml from each zone	Whole product based on a fat content of 4% by weight	258	283
	Indian cottage cheese (Paneer)	250 g from each zone	As it is	0.55	1.92
	Yogurt		As it is	1.1	1.9
Animal-based products	Chicken egg		Whole product after removal of the shell	4.3	7.0 [c]
	Chicken meat		Meat after removal of trimmable fat	2.6	7.1
	Goat meat			1.0	1.5
	Fish (Rohu, *Labeo rohita*)		Flesh after removing skin, scales, and fat	–	1.2
	Fish (Rohu, *Labeo rohita*, from local farm)			1.8	–

The average weight assumed for a [a] banana is 120 g, an [b] orange is 131 g, and for a [c] chicken egg is 70 g.

OCDD), 10 PCDFs (2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF, and OCDF), and 10 PBDEs (BDE 28, 47, 66, 85, 99, 100, 153, 154, 183, and 209) were analyzed in the food and drinking water samples.

2.4. Chemical analysis, quality assurance and control

Chemical analyses were conducted into accredited laboratories using validated methods and state-of-the-art mass spectrometry instrumentation at the RECETOX laboratory in Brno, Czech Republic. For the sake of conciseness, a detailed account of all sample preparation, extraction, clean-up, and instrumental analysis is reported in the Supplementary Information (Text S1) along with a description of the quality assurance and control measures.

2.5. Estimation of EDC exposure

Dietary intakes of the target compounds through consumption of contaminated drinking water and food were estimated for Indian population between 3 and 75 years old as follows:

$$\text{EDI}_{\text{water and food}} = \sum_{i,j} \frac{(C_i \cdot Q_j \cdot r_{uptake})}{bw} \quad (1)$$

where $\text{EDI}_{\text{water and food}}$ (ng/kg-bw/day) is the comprehensive estimated daily intake of the full set of analyzed contaminants (calculated for compound family), C_i (in product weight (ng/g) or wet weight (ng/g w. w.) for dairy, fish, meat, and poultry products) is the concentration of the i-th contaminants in drinking water or food items, Q_j (g/day) is the

daily intake of the j-th food items or drinking water, and r_{uptake} is the gastrointestinal uptake factor (a value of 100% was assumed) (Lu et al., 2021; Shen et al., 2016), and bw (kg) is the mean body weight of an individual from a given age group (Table S4). For PCDDs/Fs and dl-PCBs, calculations were done using toxic equivalents (TEQs, Table S3) rather than their masses. Summed TEQs (in ng-TEQ WHO$_{05}$/g or ng-TEQ WHO$_{05}$/g w.w.) were calculated by multiplying their concentrations by corresponding toxic equivalency factors (TEF$_{\text{WHO, 2005}}$). It is acknowledged that TEQ are not specifically assimilating information on endocrine disruption effects. To this regard, the use of relative estrogenicity quotient (EEQ) would have been more appropriate. However, the aim of the present study was to assess and compare exposure rather than estimating the risk from estrogenic activity. Furthermore, EEQs are not available for all compound targeted in this study. Finally, available data from the EU on PCBs and PCDD/Fs dietary exposure are presented in aggregated form using TEQs. Considering these points, the use of TEQ in calculating dietary exposure for dl-PCBs and PCDD/Fs represented the viable option to enable addressing the objectives of the study.

In this study, it was assumed that Q_i and bw are influenced by geographic/socioeconomic transects (i.e. urban vs rural), dietary practice (vegetarian/non-vegetarian), age, and sex according to the survey of NSSO on Household Consumption of Various Goods and Services in India in 2011–12 (MoSPI, 2014). This survey report provides national- and state-wise monthly per capita consumption of a variety of food items by rural and urban populations (provided in Table 1). We extrapolated these food consumption rates based on different dietary practices, age groups, and sex (Table S4).

Results of dietary exposure estimates calculated based on the EDC

dataset from Delhi were compared with dietary exposure from the European average population. The purpose of this comparison was to estimate difference in dietary exposures to two populations with distinguished food consumption and dietary choices. This was done considering contamination levels of POPs in food item taken from the reports of the European Food Safety Authority (EFSA, 2014; 2012a; 2012b). Consumption rates of individual food groups by an average EU citizen were extracted from two recently published studies (Crenna et al., 2019; Notarnicola et al., 2017). These average food consumption rates were further extrapolated for children, adults, and seniors based on their required calorific intakes (Table S5–S7). EDIs were calculated (following Eq. (1)) for three different age groups (children, adults, and seniors) of EU habitants and compared with their Indian equivalents. To achieve an optimal uniformity between Indian (primary) and European (literature) datasets, EDIs were calculated only for those contaminant groups (DDTs, HCH, HCB, PCBs, PCDDs and dl-PCBs and PBDEs) that were present in both datasets (Indian and European). The rest of the data (e.g. PCDFs) were omitted in this comparison. Average concentrations of the selected compounds in each food categories were used to estimate EDIs for the European populations. Furthermore, measurement units of both Indian and European datasets were made consistent using conversion factors from lipid weight (l.w.) to wet weight (w.w.).

Furthermore, since the data on POP concentrations in individual food items consumed by European population were provided in form of different quantiles (mean concentrations and their confidence intervals, 99th percentiles) (provided in Table S19), we used a bootstrap technique for delivering a dataset with a comparable structure with that of the Indian dataset. For this, 1000 estimates of concentration data for each food items in the EU food basket were randomly drawn from the reported quantiles fitted with a "best likelihood" distribution-fitting algorithm. Next, 10000 individual people were simulated in three different age groups (children, adults, and seniors), randomly assigning to each an age and corresponding body weight (provided in Table S6). For each of these simulated individuals, EDIs were calculated using food consumption rates of each food groups (Table S5) (adjusted by daily calories demand of different age classes provided in Table S7) and one of the estimated values of concentration in each food group. These computed distribution of EDIs were further presented in aggregated form using box plots (depicting median, 25–75 percentiles) and whiskers (depicting 5–95 percentiles) for each age group and plotted against the empirical results of EDIs for the Indian equivalent group of people. All calculations to estimate the daily intakes of EDCs for the Indian and the European populations were performed using R version 3.0.2 (R Core Team, 2013).

3. Results and discussion

3.1. Overview of results on food contamination

Most targeted compounds were detected in all food items, including drinking water. The following are the generally observed patterns: i) OCPs (and especially DDTs and HCHs) are ubiquitous in the analyzed food items; ii) a diffuse occurrence of OCPs (prevalently of γ-HCH and α-HCH) in drinking water samples with a tendency towards higher levels in packaged water compared to municipal supplied water; iii) a prevalence of several contaminants in tomatoes and apples (mostly associated with higher residues of HCH and DDT) compared to other fruit and vegetables; iv) moderate levels of PCBs in all analyzed samples; v) the ubiquitous occurrence of several PBDE congeners in the Delhi food basket and in particular in fish, egg, poultry and goat meat (reported for the first time in this study); and vi) sporadic high concentrations of highly substituted PBDEs and PCDD in animal-based products.

A comparison between present data and previously reported data on POP contamination in Indian food (covering the period between 1992 and 2017) is presented in the Supplementary Information (Text S2, Figure S2). Such a comparison showed that concentrations of some

contaminants (primarily DDTs and OCPs) measured in this study were up to two orders of magnitude lower compared to previous reports.

A detailed description of the observed patterns of food contamination is provided in the following sections organized by group of food items.

3.1.1. Drinking water

Several compounds were detected in drinking water samples from Delhi (Fig. 1A, Table S8). Concentrations were slightly higher in packaged water than in municipal supplied water. γ-HCH was the most abundant contaminant and was found at concentrations up to 55.7 ng/L in municipal supplied water, and 83.9 ng/L in packaged water. These levels were in the same order of magnitude as the USEPA recommended threshold of 200 ng/L (ATSDR, 2005). Concentrations of α-HCH was up to 0.6 ng/L in municipal supplied water and up to 0.8 ng/L in packaged water. High concentrations of both these HCH isomers likely reflect the historically extensive use of both the technical-HCH (mixture of α-, β-, γ-, δ- HCH) (technical-HCH was banned in 1997 in India) and γ-HCH (banned in 2013 in India) in agriculture and their high persistence in soils, from where they can reach water sources (both surface and groundwater reservoirs) through runoff and leaching (Chakraborty et al., 2015; Mutiyar et al., 2011; Singh et al., 2007).

Concerning DDTs, *p,p'*-DDE and *o,p'*-DDT (only found in packaged water) were the most abundant congeners measured in the drinking waters, with concentrations up to 0.3 ng/L. Some PBDE congeners (i.e. PBDE 47, PBDE 99, PBDE 100, PBDE 153, PBDE 154) were routinely detected at trace levels (e.g. between <1 and 62 pg/L). PBDE 209 was measured in some samples of both packaged and municipal drinking water up to 780 pg/L. Indicator PCBs and PCDDs/Fs were typically below detection limits.

Concentrations of DDTs and HCHs measured in the water samples originating from different zones of Delhi were typically within the same order of magnitude. Only for PBDE 209 a substantial variability (up to within 3 order of magnitude) was observed. Some EDCs (β-HCH, PeCB, PBDE 153, and PBDE 209) were detected less frequently in packaged water than in municipal water.

3.1.2. Fruits and vegetables

All the targeted compounds were detected in at least one type of fruits (Fig. 1B, Table S9–S10) collected in Delhi's markets. Generally, bananas, oranges, and watermelons appeared to have the lowest content of the analyzed POPs (in terms of both frequency of detection and concentrations). These fruits are generally supplied to Delhi's food markets from different regions of India, and the difference in concentration profiles of these fruits (especially for OCPs) may reflect local farming practices and the historical use of pesticides, as well as physiology and composition of the different plants.

OCPs were detected in fruits (especially in apples) at the highest concentrations compared to other groups of EDCs. Among them, γ-HCH, *p,p'*-DDE, and PeCB were the most abundant (up to 0.45 ng/g, 0.03 ng/g, and 0.01 ng/g, respectively). The highest measured concentration of HCB was 0.005 ng/g (in apple). Among non-pesticidal EDCs, PBDE 209 was measured up to 0.01 ng/g (in apple), PCB 28 up to 0.01 ng/g (in apple), OCDD and OCDF (7.9×10^{-2} pg/g and 3.01×10^{-2} pg/g, respectively).

The contamination profile of vegetables was similar to that of fruits, but with a higher detection frequency (54 total detected substances in vegetables compared to 43 in fruits) (Fig. 1C and D, Table S11–S12). OCPs in vegetables were generally present at higher concentrations compared to other group of compounds. Tomatoes had the highest residues of OCPs (e.g. *p,p'*-DDT up to 2.3 ng/g, γ-HCH up to 0.78 ng/g). Residues of all the targeted OCPs were detected in spinach, tomato, cauliflower, and okra. Non-pesticidal compounds were detected in vegetables with higher frequencies compared to fruits, with spinach showing the highest residues, followed by tomato, okra, cauliflower, cabbage, onion, and potato.
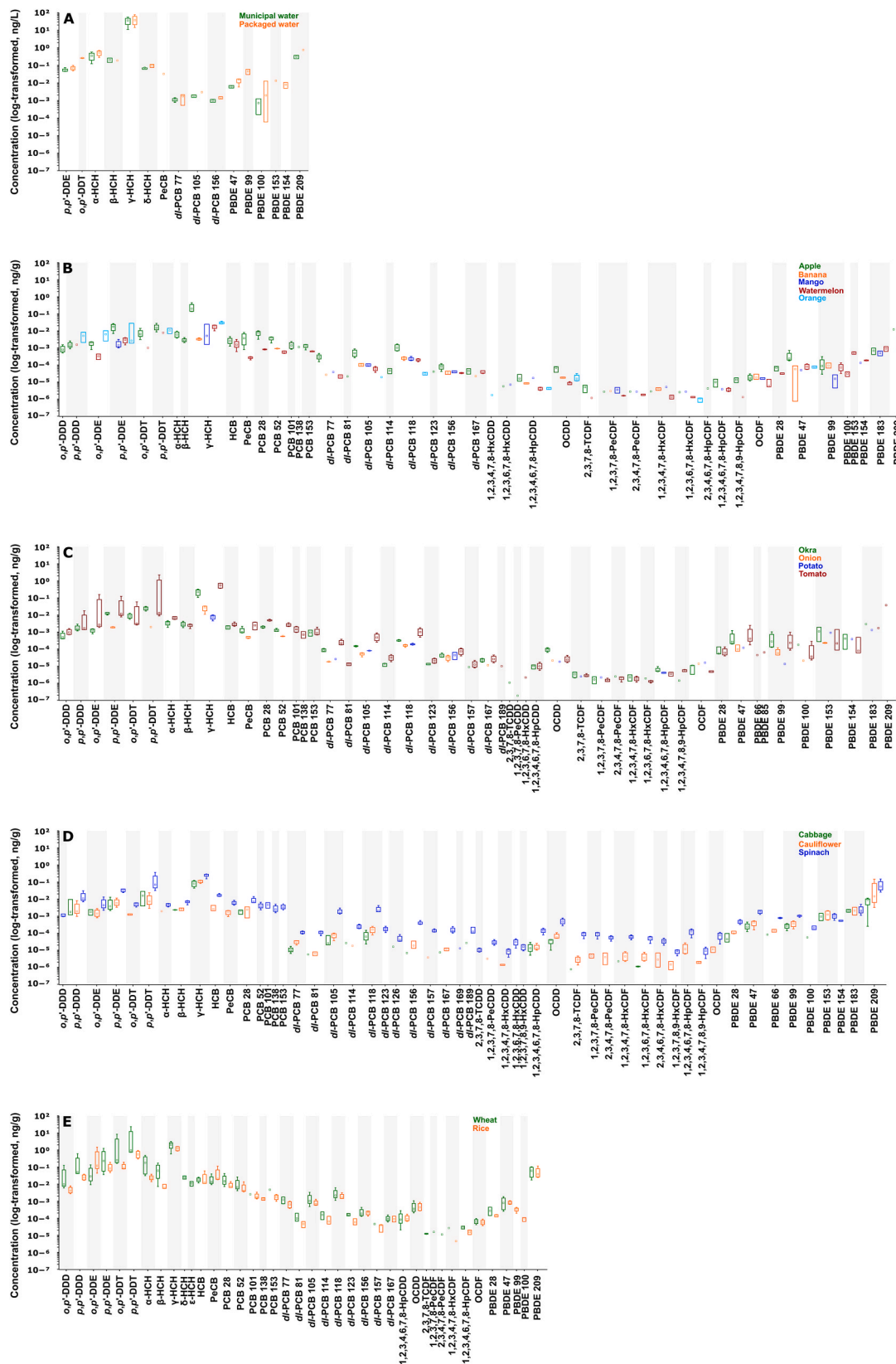
Fig. 1. Log-transformed concentrations of contaminants in drinking water (A), fruits (B) vegetables (C, D), cereal grains (E), pulses (F), dairy products (G), fish (H), and poultry products and goat meat (I) obtained from food markets in Delhi. Concentrations of fruits, vegetables, cereals, and pulses (in Fig. 1 and sections 3.2 and 3.3) are presented in ng/g product weight. The boxes represent median (□) and 25th and 75th percentiles; the whiskers mark non-outlier range (1st–99th percentile). Alternate grey stakes are to distinguish between different chemicals. The complete dataset is reported in supplementary data (Table S8–S17).
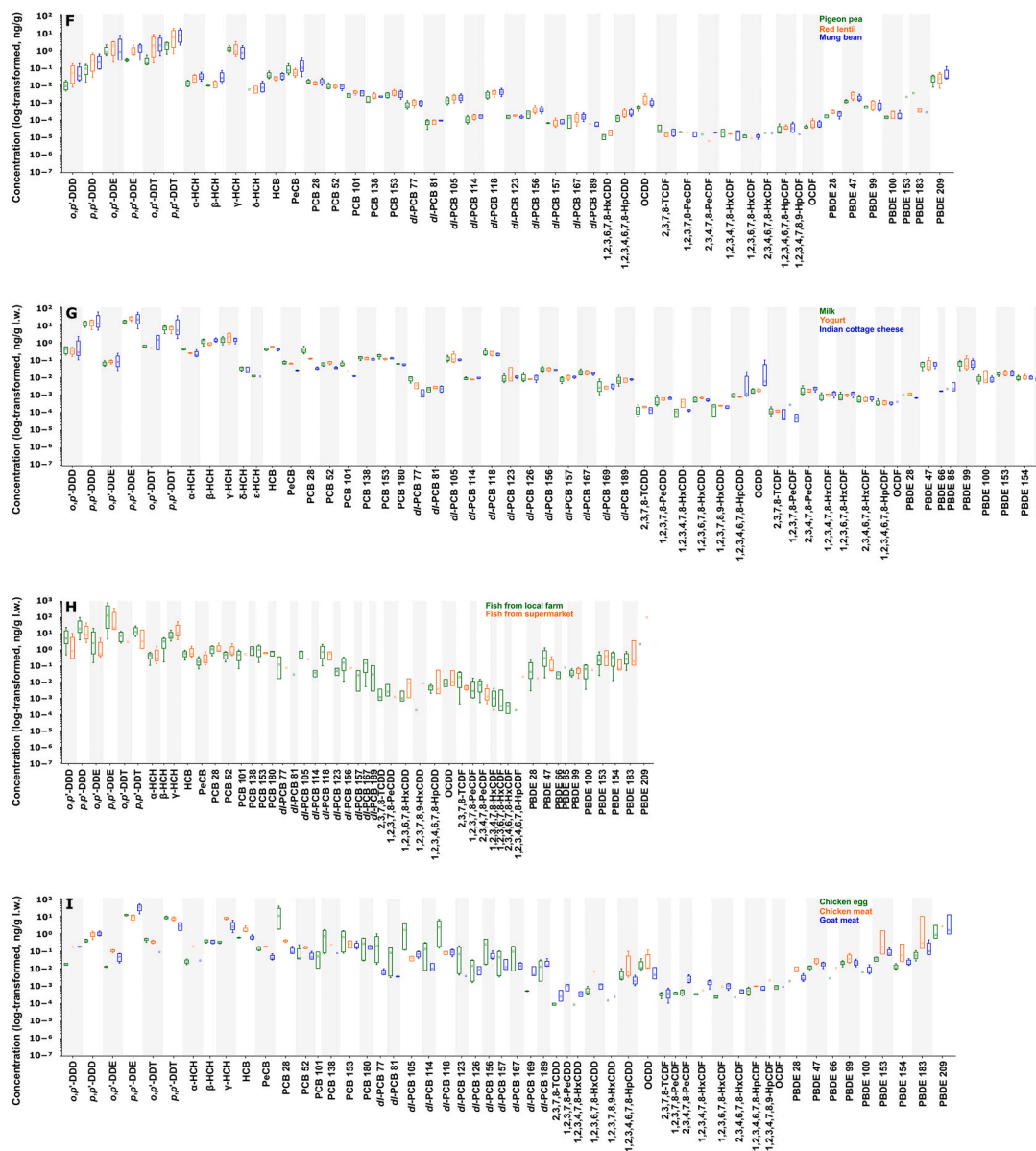
Fig. 1. (*continued*).

Similar to water and fruits, PBDE 209 (up to 0.15 ng/g) had the highest concentrations among PBDEs but was only detected in spinach and tomato. PCBs were measured at trace levels especially in tomato and spinach (e.g. up to 10 pg/g of PCB 118). OCDD and OCDF were the most abundant PCDD/Fs (0.77 pg/g and 0.11 pg/g, respectively, in spinach).

3.1.3. Cereals and pulses

Wheat was found with higher residues of most contaminants compared to rice, (with the exception of *o,p'*-DDE, HCB, PeCB, PCB 101, 138, PBDE 99, 100) (Fig. 1E, Table S13). Similar to vegetables, *p,p'*-DDT in wheat (23.2 ng/g) and rice (0.9 ng/g) were the most abundant contaminants measured in cereals and pulses, probably reflecting recent uses of DDT in both farming (not allowed under the SC) and as control of vector borne diseases (permitted use under the SC) in many parts of India. The composition profile of different groups of contaminants in wheat and rice was similar to that found in other food items, especially in vegetables and pulses. Higher number of compounds (46) were detected in pulses compared to cereal grains (40) (Table S14). The contamination profiles of the three typologies of pulses sampled in this study were similar (Fig. 1F).

3.1.4. Dairy products

As expected, the concentrations of the targeted POPs in food products of animal origin were higher than plant-based food items. In total, up to 55 compounds were detected in milk, yogurt, and Indian cottage cheese (Fig. 1G, Table S15) with similar levels and profiles among these products. DDT congeners had the highest levels with a prevalence of *p, p'*-DDE (up to 54.8 ng/g l.w. in cottage cheese). All isomers of HCH were detected in at least one type of dairy product. The highest concentration was observed for γ-HCH (3.6 ng/g l.w.) in yogurt. Among PCBs, the highest concentration was detected for PCB 28 in milk (0.6 ng/g l.w.). Among PBDEs, PBDE 99 was detected with the highest concentration of 0.2 ng/g l.w. in yogurt. PBDE 209 (typically the most abundant among PBDEs in other food items) could not be detected in any of the dairy products owing to the high detection limits achieved (i.e. 101–1830 pg/g l.w.). In addition to the detection limits, lower presence of PBDE 209 in dairy products can be due to its high molecular mass leading to its lower bioaccumulation compared to other PBDE congeners with lower molecular mass, moreover, PBDE 209 has the capacity to breakdown in more toxic and easily absorbed congeners such as those existing in penta- and octa-BDEs (McDonald, 2002; O'Driscoll et al., 2016).

3.1.5. Fish

The highest concentrations recorded in this study were observed in fish fillet. Among the target compounds, *p,p'*-DDE (up to 813 ng/g l.w. in fish) was the most abundant contaminant (Fig. 1H, Table S16). DDT was found at higher concentrations in locally farmed fish compared to fish from supermarkets (originating from different parts of India and essentially from commercial fish farms). γ-HCH was the prevalent compound among the HCH isomers with concentrations up to 55 ng/g l. w. α-HCH, HCB and PECB were detected in nearly all fish samples at levels ranging 0.06–0.7 ng/g l.w. Several PCBs and PBDEs congeners were routinely detected with higher substituted ones being the most abundant (e.g. PCB 153 ranging 0.16–1.44 ng/g l.w. and PBDE-183 ranging 50–3740 pg/g l.w.). PBDE 209 was the most abundant PBDE congener, however, due to high detection limits we could quantitatively measure it only in one aggregated sample of fish collected in the North zone (at 99 ng/g l.w.). Concerning dioxins, 2,3,7,8-TCDD, 1,2,3,7,8-HxCDD, and 1,2,3,6,8-HxCDD were routinely detected especially in locally farmed fish at levels ranging 0.7–7.2 pg/g l.w., while they were typically below detection limits in fish sampled from supermarkets. 1,2,3,4,6,7,8-HpCDD and OCDD were found in all fish samples (regardless the provenience) at concentrations ranging between 2 and 50 pg/g l.w. Only tetra-to hexa-CDFs were detected prevalently in locally farmed fish at concentrations ranging 0.1–46 pg/g l.w.

3.1.6. Poultry products and meat

In general, poultry products (both chicken egg and meat) and goat meat (Fig. 1H and I, Table S17) had lower concentrations compared to fish. OCPs was the group with the highest concentrations (especially *p, p'*-DDE (up to 53.2 ng/g l.w.in goat meat). HCB, PeCB, and γ-HCH were routinely detected in egg and meat with poultry meat showing the highest concentrations of HCB (e.g. 0.6–2.8 ng/g l.w.). PeCB was instead more abundant in chicken egg and meat compared to goat meat. Low chlorinated PCBs were also routinely detected in egg and meat samples, while higher chlorinated PCBs such as PCB 138, PCB 153 and PCB 180 were predominantly found in egg and to a lesser extent in goat meat. Concerning PBDEs, a clear trend was observed with increasing concentrations at increasing level of halogenation, likely reflecting a biomagnification-related pattern. In one chicken meat sample, PBDE 209 was found more than 300 ng/g d.w. A similar pattern was observed for dioxins where 1,2,3,4,6,7,8-HpCDD (2–100 pg/g l.w.) and OCDD (4.5–120 pg/g l.w.) were the most frequently detected compounds in chicken meat. Lower chlorinated furans were instead frequently detected, especially in eggs and goat meat at concentrations ranging 0.1–1.8 pg/g l.w.

3.2. Comparison of EDC concentrations in food items collected in urban vs rural markets

Differences in the contaminant concentrations and frequency of detection in the food items collected from markets in Delhi and Dehradun (Fig. S2B–S2E) were observed for some food items. Concerning drinking water, the levels of γ-HCH and α-HCH (the most frequently detected contaminants in these samples) were a factor of 10 higher in Delhi than in Dehradun (Fig. S2A). In contrast, several PBDE at low and intermediate level of halogenation were significantly higher (up to 3 orders of magnitude) in municipal drinking water in Dehradun (ranging 70–760 ng/L depending on the congener) compared to Delhi (Table S8). Observed traces of POPs in drinking water (particularly in municipal supplied drinking water) indicate local contamination sources in both reference sites and warrant further detailed assessments on fate and distribution of POPs in Indian metropolitan cities.

POPs were detected with a lower frequency in fruits, vegetables, cereals, and pulses samples from the rural market (Fig. S2B, S2C, and S2D) especially concerning DDTs, PCBs, and PBDEs. For instance, the concentration of *p,p'*-DDT and many PCB and PBDEs congeners in spinach and tomato samples from the rural market were up to 6-folds

and 2 orders of magnitude lower than in urban market samples, respectively.

Concerning dairy and animal-based products, we observed no differences between samples from Delhi and Dehradun. Similarly, no substantial differences were observed in the level of contamination of fish samples from Delhi and Dehradun for any group of compounds. In chicken eggs, levels of *p,p'*-DDT, *p,p'*-DDE, *o,p'*-DDT, *o,p'*-DDE as well as most PCBs and OCDD were lower by a factor typically of 2–3 (and sporadically of a factor exceeding one order of magnitude) in Dehradun compared to Delhi. In chicken meat, *p,p'*-DDT, γ-HCH, HCB showed a similar pattern with slightly higher levels in the samples collected from Delhi (Fig. S2E). This, however, was not the case for goat meat where no obvious differences were observed. A similar pattern occurred for PCBs, where chicken meat from Dehradun had lower levels of several congeners compared to samples from Delhi, but these differences were not observed in goat meat. In this case, in contrast, higher levels of PCBs (up to 1 order of magnitude) were observed in the samples from Dehradun.

We tested the significance of difference between contamination in food from Delhi and Dehradun markets, using Wilcoxon signed-rank test (Wilcoxon, 1945). The difference was significant only for α-HCH (p = 0.03), γ-HCH (p = 0.001), HCB (p = 0.025), Σdl-PCBs (p = 0.01), ΣPCDDs (p = 0.02), and ΣPCDFs (p = 0.005).

3.3. Assessment of dietary exposure

As a result of typically higher POP concentrations in the food items sampled in the urban setting (Delhi), EDIs for the urban population tended to be higher than those calculated for the rural population (based on samples from Dehradun) (Fig. 2). The highest differences were observed for dl-PCB (up to 2 orders of magnitude) and PCBs, PCDDs/Fs (one order of magnitude), differences for other groups (i.e. OCPs and PBDEs) were within 1 order of magnitude and can be considered non-significant considering the resolution of the estimation model. This pattern stems from the following facts i) industrially processed food, or food produced and/or processed in proximity of the large urban and industrial conglomerations (available in the large city food markets) are more likely to be contaminated by non-pesticidal POPs and ii) urban population has a higher consumption of animal-based products (egg, meat, and dairies) compared to the rural one.

Among different contaminant groups, OCPs were the largest contributors to EDIs (up to 296 ng/kg-bw/day) both for the urban and rural populations. The lowest EDIs were observed for dl-PCBs. This is not surprising as PCBs were not produced in India and had little historical use. Primary sources capable of producing PCB contamination hotspots are therefore supposedly rare, especially in rural areas. Children had the highest EDIs of all the contaminant groups both in rural and urban settings. Precisely, EDIs of male-vegetarian population were the highest followed by male-non-vegetarian, female-vegetarian, and female-non-vegetarian populations. Differences between vegetarian/non-vegetarian diet were minimal. This finding might appear surprising as meat products had higher concentrations for several highly hydrophobic and bioaccumulative compounds. The Indian vegetarian diet is however rich in dairy products as a substitute of meat and based on our results dairy products had comparable levels of POPs as meat and fish products.

EDIs calculated for the Indian population in the present study were lower than those reported in earlier studies from India. A previous study estimating dietary intakes based on total diet in the State of Punjab, India reported EDIs of HCHs and DDTs to be 1362 and 453 ng/kg-bw/day, respectively (Battu et al., 2005). Similarly, another study reported EDIs of HCHs and DDTs among middle-income Indian population to be totaled 19671 ng/kg-bw/day (Betsy et al., 2014). For OCPs, EDIs estimated in this study were found higher than those reported for the Chinese population (27 ng/kg-bw/day) (Zhou et al., 2012). Contrary to OCPs, EDIs of PBDEs reported for the Chinese population were found up to 3 orders of magnitude higher (957.2 ng/kg-bw/day) than estimates for the Indian population in this study (Labunska et al., 2014). Similarly,
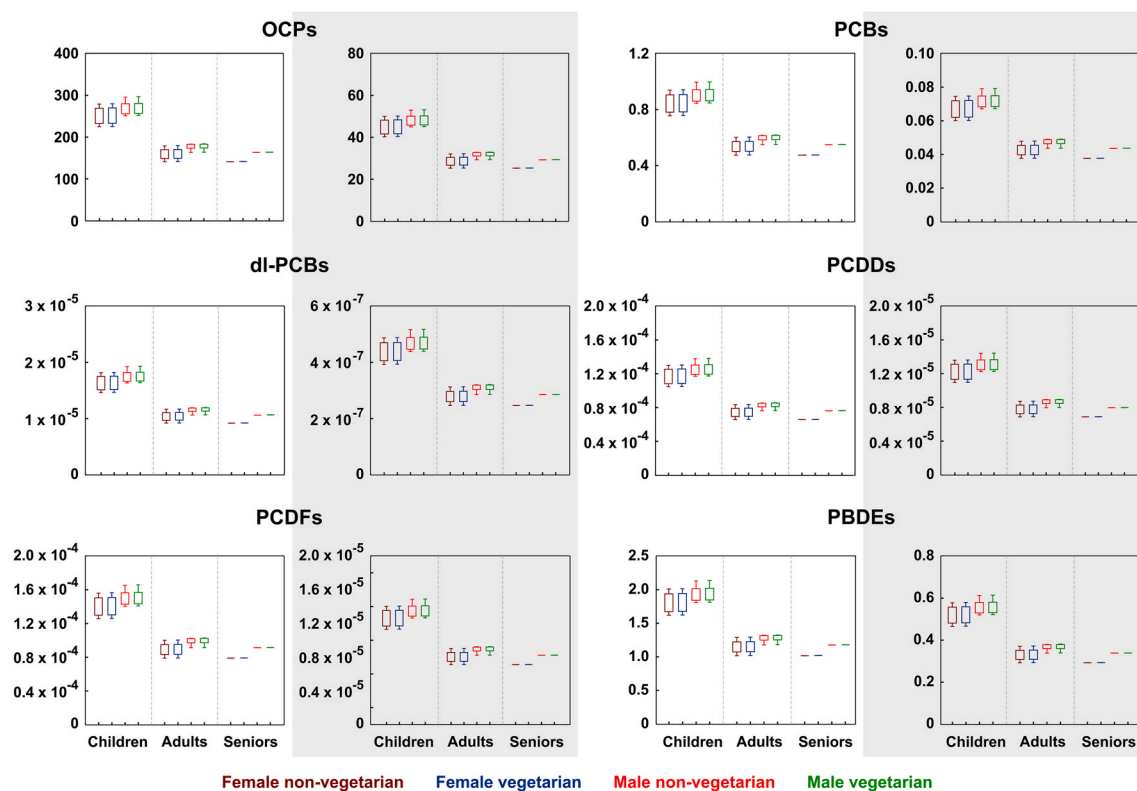
Fig. 2. Daily dietary intakes of contaminants (by group) based on a comprehensive food basket analysis. The x-axis displays the age groups (children (3–12 years), adults (18–60 years), seniors (>61–75 years)) and the y-axis displays EDIs through food consumption (including drinking water) (in ng/kg-bw/day; for dl-PCBs, PCDDs, and PCDFs in ng-TEQ WHO$_{05}$/kg-bw/day). The boxes represent 25th and 75th percentiles and the whiskers mark non-outlier range (1st–99th percentile). The plots on white background show dietary intakes for the urban population while the plots on grey background show intakes for the rural population.

EDIs of dl-PCBs and PCDDs/Fs calculated in this study were remarkably lower than the estimates for the Chinese population (up to 0.003 ng-TEQ/Kg-bw/day) (Zhang et al., 2013). No previous study reported total diet based EDIs of PCBs, dl-PCBs, PCDDs/Fs, and PBDEs for the Indian population. Nevertheless, high estimates of dietary intakes based on individual food groups were previously reported for the Indian population. For example, dietary intake of PCBs through fish consumption in middle-income Indian population was 6.4 ng/kg-bw/day (Ahmed et al., 2016). Dietary exposure to selected POPs in Indian population is even confirmed by several studies focusing on human biomonitoring which found high concentrations DDT, HCH, and PCBs in human blood and breast milk samples from various locations in India (Bawa et al., 2018; Jaacks et al., 2019; Sharma et al., 2014; Sharma et al., 2014b). The new findings provided in the present study endorse a reconsideration of the current understanding of Indian people's dietary exposure to POPs, suggesting a substantially lower pressure from these groups of EDCs compared to what some of the earlier studies had presented.

3.4. Comparison of dietary exposure between India and Europe

Mean EDIs of OCPs calculated in the present study for the European population were slightly higher than EDIs reported in earlier studies (Coelho et al., 2016; Dirtu and Covaci, 2010; Fromberg et al., 2011; Xu et al., 2017) although they are contained within the 95% boundaries of the data distribution. Mean EDIs for PCBs and PCDDs were instead consistent (between 4.3 and 25.7 ng/kg-bw/day for PCBs and average up to 0.002 ng-TEQ WHO05/kg-bw/day for dl-PCBs and PCDDs) (Malisch and Kotz, 2014; Törnkvist et al., 2011). Earlier investigations have also reported EDIs of PBDEs for the European population to be between 0.15 and 3.5 ng/kg-bw/day (Bedi et al., 2020; Pardo et al., 2020; Törnkvist et al., 2011; Xu et al., 2017), in agreement with

estimates in the present study. The differences observed for OCPs may derive from the fact that previous reports focused on local or national case studies and a limited number of samples rather than using the complete figures from EFSA datasets (as done here).

In the present study, the EDIs of OCPs and PBDEs were comparable between Indian and average European population (Fig. 3). In contrast, EDIs were higher for the European population (up to 2 orders of magnitude) for PCBs and (up to 3 orders of magnitude) for the sum of dl-PCBs and PCDDs (Fig. 3 Upper panel). The difference in concentrations of these two groups of contaminants in Indian and European food was also significant (p = 0.03). Evidences of high PCBs and PCDD/Fs dietary exposure in Europe were previously suggested by a study based on global surveys on human breastmilk (van den Berg et al., 2017).

Higher overall exposure of the average European population can be partly explained based on dietary choices, historical use of POPs like PCBs, and relative environmental occurrence of pollutants in these two regions (Kanan and Samara, 2017; Weber et al., 2018). The typical European diet is rich in meat and animal-based products. On average, an EU citizen consumes 80 kg of meat each year (Ritchie and Roser, 2017). In contrast, Indian diet is dominantly lactovegetarian, and the average meat consumption is only about 4 kg/year (Devi et al., 2014; Rammohan et al., 2012). This is expected to reduce the exposure of highly bioaccumulative compounds. Differences in food baskets and dietary choices appears therefore to be a determinant of lower dietary exposure of the Indian population compared to average Europeans.

To further investigate the role of this determinant, however, EDIs were computed and compared specifically for the vegetarian groups in both Europe and India (Fig. 3 Lower panel). As expected, the regional differences in EDIs were remarkably lower in this case. For example, difference for dl-PCBs and PCDDs declined from up to 3 orders of magnitude to 1 order of magnitude, however, results from the comparison of the vegetarian food baskets also confirmed that European
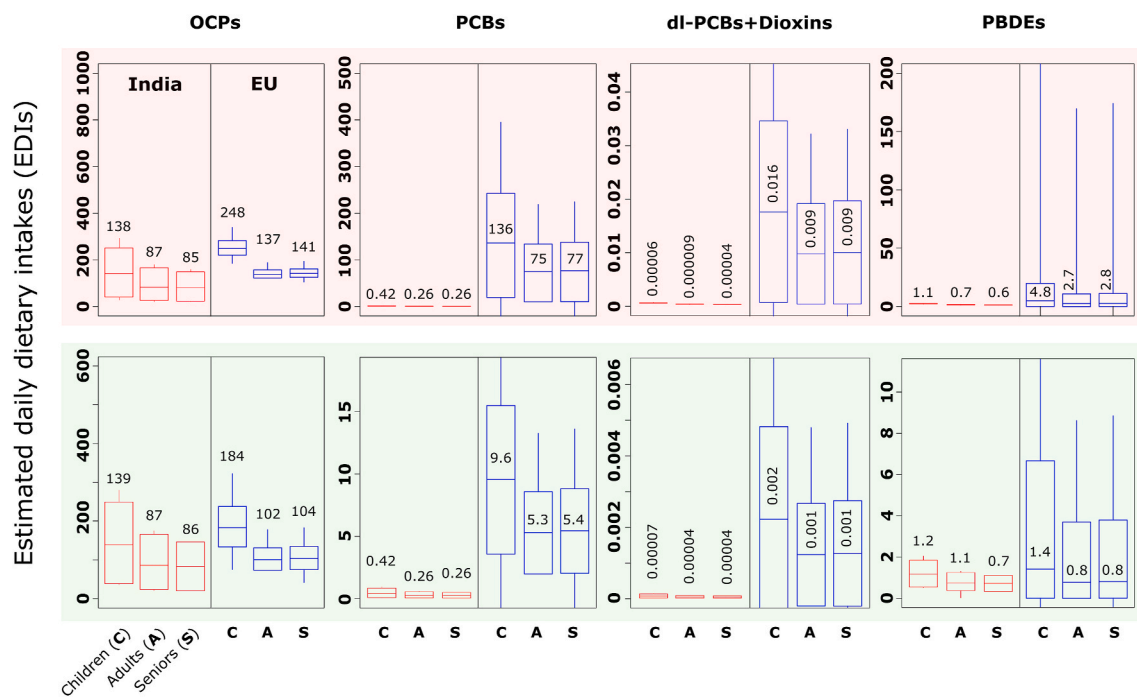
Fig. 3. Comparison between estimated daily dietary intakes (EDIs) of selected contaminant groups by Indian and European populations. For OCPs, PCBs, and PBDEs, EDIs are presented in ng/kg-bw/day, whereas EDIs for the sum of dl-PCB and PCDDs are presented in ng-TEQ WHO$_{05}$/kg-bw/day. The boxes represent median (horizontal line) and 25th and 75th percentiles; the whiskers mark non-outlier range (5th–95th percentile). Median EDI values for individual population groups are depicted above or inside the boxes in each plot. **Upper panel** (pale red background): Comparison between non-vegetarian populations, **Lower panel** (pale green background): Comparison between vegetarian populations.(For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

population may have a inherently higher exposure to POPs, essential due to higher residues of POPs in food. Concerns regarding high exposure of these EDCs to the European population have been raised in several previous studies (Hoogenboom et al., 2015; Malisch and Kotz, 2014; van den Berg et al., 2017; Weber et al., 2018). Despite the higher focus and investment in pollution control and food safety assessment in Europe, such a higher exposure of the European average population may reflect the legacy of historical environmental contamination. Such a result is notable especially considering the comparison was conducted here with India, a country generally considered a hotspot of legacy POP pollution, with incomplete implementation of the SC and meager pollution control measures and regulation.

3.5. Significance and limitations of the study

The study described here represents the most comprehensive assessment of EDC levels in Indian diet available to date. The analyzed food basket and the list of target compounds in this study certainly did not cover the full spectrum of food consumed in India, nor the full list of EDCs relevant for human health. However, the study covered the most common and important food items consumed in the average Indian diet and described the most inclusive food item list ever analyzed in any available study on dietary exposure in India. Also, the number and diversity of EDCs studied here (with a focus on POPs) was omitted in previous studies from this region.

Considering that India is among the top global food exporting countries (APEDA, 2020), comprehensive information on EDC contamination in Indian food basket is quintessential for the development of new markets (nationally and internationally) where priority is given to the food quality and safety.

The comparison of dietary exposure between India and Europe is useful to shed light on the role of effective chemical management. An important limitation inherent to this comparison is the heterogeneity in

structure and data density between the datasets from Europe (data from EFSA synthesis of national reports) and India (data produced in this study). The European dataset arises from a very large international monitoring effort accounting for a comprehensive list of items sampled in several food markets across all European nations. Such an effort is lacking in India (as well in many developing countries). The present study addressed this gap by providing a pilot that may serve to endorse and guide the development of a national food safety monitoring program in India.

4. Conclusions

This was the first study to provide comprehensive data on levels of several important groups of EDCs in Indian food baskets, to estimate dietary intakes of several groups of POPs, and to compare results with historical Indian and European data. Although it has been more than a decade since their ban and/or restriction, HCH and DDT still represent main contributors to POP content in the Indian diet. HCHs were found in drinking water at levels comparable to the USEPA safety threshold. Results however suggested that the levels of OCPs in food items in India has likely dropped during the last three decades. No major difference in the EDIs were found in India between vegetarian and non-vegetarian groups. Children appear to be the most exposed group. The comparison with European data suggests a higher dietary exposure in Europe especially of non-pesticidal POPs (e.g. PCBs and PCDDs), which is only partly explained by higher consumption of animal-based products in Europe.

These results challenge the common opinion of India being a general hotspot for POP contamination. A probable explanation behind this perspective is that earlier studies have mostly focused on hotspot areas overlooking the broader perspective. Findings reported in this study show that Indian and European population's exposure to the analyzed EDCs are similar, or that Indian population have even a lower dietary

exposure to some non-pesticidal POPs such as PCBs and PCDD/Fs. In both cases, the group of PBDEs represent a concern, considering the elevated levels found in some animal-based products (including in the rural setting).

A substantial part of endocrine disrupting potential in the diet derives from food and animal feeds internationally traded between developed and developing countries. Comparing food basket contamination across countries with a vocation for food export can enable a harmonized development of pollution control and food safety regulation in both contexts. With increasingly globalized food systems, internationally harmonized policies on EDC in food can lead to better protection of health in both the global North and South.

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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.117750.

Credit author statement

BMS: methodology, investigation, data analysis, modelling, visualization, results and discussion, writing original draft; GKB: project coordination in India, sampling, sample preparation, results and discussion, manuscript editing; PC: sampling, sample preparation, result discussion, editing; JM: Chemical analysis; OA: chemical analysis; PK: chemical analysis; PP: chemical analysis; PKK: sampling, sample preparation; AS: literature review, inventory of data from literature, manuscript editing; JK: data analysis and modelling; EHS: discussion of results, manuscript editing, supporting project coordination; LN: principal investigator, scientific case definition, discussion of results, writing, manuscript editing.

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