Review Article

Concentrations of Polychlorinated Naphthalenes in Food and Human Dietary Exposure: A Review of the Scientific Literature

Jose L Domingo[1]

1. Universitat Rovira I Virgili Tarragona, Spain

In general, for most environmental persistent organic pollutants (POPs), dietary intake is the main way of exposure. Polychlorinated naphthalenes (PCNs) are a family of two-ringed aromatic compounds that are ubiquitous environmental contaminants, being structurally similar to PCDD/Fs and PCBs. Although the production and use of PCNs were banned in the USA and Europe some decades ago, due to their persistent properties, PCNs remain present in the environment, able to enter the food chain. The present paper was aimed at reviewing the results of the studies focused on determining the levels of PCNs in foods. The human dietary intake of these compounds was also reviewed, with the few available data. The information on the levels of PCNs in foodstuffs is currently more abundant than that found in a previous review (Domingo, 2004). Since then, China is the country that has contributed the greatest number of studies. The results of most surveys seem to suggest that human health risks of PCNs due to dietary exposure should not be worrying. However, because of the important differences in the methodology of the published studies, the comparison of the results is not easy, although there seems to be a general trend towards a decrease in the levels of PCNs in foods. In the next few years, a continued reduction of the environmental levels of PCNs is still expected. Therefore, a direct repercussion of the concentrations of these pollutants in foodstuffs must be also noted. Consequently, a reduction of the dietary exposure to PCNs should be expected. Anyway, to establish the tolerable dietary intake of PCNs is a key issue for assessing human health risks of these pollutants.

Corresponding author: Jose L Domingo, joseluis.domingo@urv.cat

1. Introduction

Polychlorinated naphthalenes (PCNs) are a group of aromatic compounds that consist of a molecule of naphthalene that is substituted with one-to-eight atoms of chlorine. Consequently, there are 75 possible congeners of PCNs. The chemical structure of PCNs is relatively like that of other well-known persistent organic pollutants (POPs) -which are also diaromatic hydrocarbons- such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), and polychlorinated biphenyls (PCBs). Among other uses, PCNs were widely employed as dielectrics and flame retardants in the 20th century. With respect to the environmental presence of PCNs, the primary source of emissions of these compounds is due to unintentional production -along with PCDD/Fs and PCBs- from industrial thermal processes (Liu et al., 2014; Li et al., 2023), with the environmental sources of PCNs being various and diverse (Domingo, 2004; Bidleman et al., 2010; Dong et al., 2019; Rigby et al., 2023). Thus, there are reports on the emissions of PCNs by coking industries (Liu et al., 2010), iron ore sintering processes (Liu et al., 2012), waste incinerators (Hu et al., 2013; Zheng et al., 2013), hot dip galvanizing plants (Liu et al., 2015), cement kilns co-processing municipal wastes (Liu et al., 2016; Han et al., 2022), metal smelters (Ba et al., 2010; Wu et al., 2018; Hu et al., 2021; Xu et al., 2020; Yang et al., 2021; Dong et al., 2022a), steelmaking industries (Shen et al., 2021), and electronic waste hubs (Wang et al., 2012; Waheed et al., 2020), among other different and specific emission sources, such as the heating of sucralose (Dong et al., 2019).

In recent decades, numerous studies concerning the environmental sources, distribution, toxicity, and human health risks of POPs - mainly PCDD/Fs and PCBs - have been carried out. However, data on PCNs are much more limited. According to the scientific literature, the first articles summarizing the environmental sources, analysis, and potential toxicity of PCNs were published in the 70s of the last century (Brinkman and Reymer, 1976; Erickson et al., 1978). An interesting review on various issues regarding PCNs was written by Hayward (1998). In that review article, the author reported the history of PCN manufacture and its connection to human and animal toxicity from occupational and experimental exposure. Once already in the 21st century, PCNs have received more attention (Falandysz et al., 2014; Liu et al., 2014; Jin et al., 2022; Klimczak et al., 2023; Olisah et al., 2024). In this sense, it is important to remark that PCNs are currently on the list (Annex A and C) of POPs, for which the Stockholm Convention considered that their use/release must be eliminated/restricted. This is important considering the potential of these pollutants to provoke a wide range of adverse environmental effects, as well as effects on living organisms (Xu et al., 2013; Fernandes et al., 2019; Olisah et al., 2024).

The similarity in the structural configuration of PCNs to those of PCDD/Fs and PCBs is related to human health concerns. Thus, a number of PCN congeners have been reported to contribute to dioxin-like toxicity (Domingo, 2004; Falandysz et al., 2014; Fernandes et al., 2017). The toxicological profile of PCNs was recently reviewed by Fernandes et al. (2022). In their conclusions, the authors remarked that since the restriction of the uses of PCNs and the stricter controls on potential atmospheric emissions, the main route of human exposure was/is currently being the dietary intake. This had already been suggested in previous reviews focused on the human dietary exposure to PCNs (Falandysz, 2003; Domingo, 2004). In this sense, it is worth to remember that the diet is also the most important way of human exposure to POPs with similar chemical structures to those of PCNs: PCDD/Fs, PCBs, or PBDEs, for example (Gasull et al., 2011; Rauscher-Gabernig et al., 2013; De Filippis et al., 2014; Marquès et al., 2022; Lacomba et al., 2024). In relation to exposure to PCNs through food intake, recently, the EFSA (2024) was asked for the opinion on the potential risks for animal and human health due to the presence of PCNs in feed and food. Based on the limitations in the available scientific data, only hexa-CNs were assessed for the human health risks of PCNs. In fact, very limited data were found regarding other PCN congeners/homologues. For hexa-CNs in food, the highest daily exposure for the general population ranged between 0.91 and 29.8 pg/kg body weight (bw). With at least 99% certainty, it was concluded that dietary exposure to hexa-CNs should not raise health concerns (EFSA, 2024).

Studies conducted in recent years have detected the presence of PCNs in several environmental matrices, including air, sediments, and biota (Hanari et al., 2004; Bidleman et al., 2010; McGoldrick et al., 2018; Du et al., 2022; Dong et al., 2023a; Gebru et al., 2024). PCNs have also been detected in human samples of serum, adipose tissue, liver, and breast milk (Weistrand and Noren, 1998; Kunisue et al., 2009; Schiavone et al., 2010; Fernandes et al., 2017). With respect to human exposure to POPs, as above commented, dietary intake has been recognized as the main pathway. Specifically regarding PCNs, the information on the topic is not so abundant as that corresponding to the dietary intake of PCDD/Fs or PCBs. However, according to the scientific literature, in the current century several studies on the presence of PCNs in food samples, as well as on the estimation of the dietary intake of these compounds, have been conducted.

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The purpose of the current paper was to review the scientific literature on the concentrations of PCNs in foods and human exposure to these environmental contaminants through dietary intake. Reports and/or data from studies about risk assessment of PCNs, conducted by regional, national, or international agencies and/or food safety authorities, have not been included here. The scientific literature has been reviewed using the PubMed (https://pubmed.ncbi.nlm.nih.gov/) and Scopus (https://www.scopus.com/search/form.uri?display=basic#basic) databases. The following keywords -and their combinations- were used for the search: "polychlorinated naphthalenes," "PCNs," "food consumption," and "dietary intake." Information on the available studies and their results is next summarized, being presented by specific countries. It must be noted that the year of publication of the reviewed studies does not necessarily correspond with the year in which the study was performed or the samples were collected.

2. Concentrations of PCNs in food and human dietary exposure

2.1. Asian countries

2.1.1. China

According to the scientific databases, in the current century, China has been the country where more studies on the distribution and source identification of PCNs in foods have been conducted. The available results are next summarized following the order of publication from the oldest to the most recent papers. Jiang et al. (2007) carried out a study aimed at measuring the concentrations of PCNs (among other POPs) in seafood collected in the Chinese cities of Guangzhou and Zhoushan. Four species of fish, two species of cephalopods, shrimps, and crabs, as well as three species of bivalves, were purchased from local markets in both cities. PCNs were found in all analyzed samples, with a range of mean concentrations of ∑PCNs between 93.8 (crab of Zhoushan) and 1300 (cephalopod of Guangzhou) pg/g lipid, depending on the sampling location and the specific species. In general, the levels of PCNs in seafood were comparable between both cities. In turn, Li et al. (2015) determined the concentrations of PCNs (also those of PCDD/Fs and PCBs) in two dairy products: cheese and butter. The ∑PCNs were in the ranges 5.6–103 pg/g wet weight (ww) in the cheese samples and 5.0–199 pg/g ww in the butter samples. PCN-73 was the predominant congener, contributing with more than 40% to the PCN TEQ-value. PCN congeners 63, 66/67, 69 were also found at relatively high levels.

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Cui et al. (2018) analyzed the levels of PCNs in 122 marine fish samples, which belonged to 17 species of different trophic levels from a coastal area in a typical industrial Chinese zone: the Bohai Rim Region. Most PCN congeners were detected in most samples. The ∑PCNs concentrations ranged between 7.3 and 214 pg/g ww, with a variation of two orders of magnitude. The highest ∑PCN level was found in ditrema (*Ditrema temminckii*). The predominant PCN congeners were tri-CNs followed by di-CNs, penta-CNs, and tetra-CNs, which contributed respectively with 32%, 23%, 20%, and 15% to the ∑PCN concentration. Interestingly, the authors highlighted that the PCN TEQs were much lower than the PCDD/F and dioxin-like PCB TEQs found in previous studies in fish performed in the Bohai region. The results suggested that toxicological concerns posed by PCNs were rather negligible. On the other hand, Han et al. (2018) measured the concentrations of PCNs in the blood of eight species of animals, whose meats are often consumed. The levels of PCNs (a total of 75 mono- to octa-CN congeners were quantified) were analyzed in whole blood samples of horse, donkey, cattle, sheep, chicken, duck, rabbit, and pig (animals obtained from large farms in the suburbs of Beijing). In the eight species, the median and mean concentrations of PCNs -given on a liquid volume basis- were 826 and 756 pg/L, respectively, with a range between 305 and 987 pg/L. The highest and lowest PCN levels corresponded to the blood of the donkey, 987 pg/L, and the pig, 305 pg/L. The mono-CNs were the predominant homolog group, followed by the di-CNs and tri-CNs. With respect to the dietary intakes of PCNs, it was found that chicken, beef, and pork contributed most to the total TEQs, with percentages of 61%, 27%, and 5.9%, respectively. In turn, Wang et al. (2022a) determined the concentrations of 75 PCN congeners in 102 samples of commercial green tea, a popular beverage in China, which were collected from 10 Chinese provinces and Chongqing City. The mean PCN concentration in the green tea samples was 36.1 pg/g dry weight (dw) (range: 3.62-175 pg/g dw). Similar profiles of homologs and congeners of PCNs were observed for all samples from different areas, with di-CNs, tetra-CNs, and tri-CNs the dominant PCN homologs. It was concluded that PCNs in green tea should pose little risk for consumers. In another study, Qi et al. (2022) investigated the presence and distribution of PCNs in 107 samples of bees, bee pollen, and wax collected in various geographical Chinese regions. A total of 75 PCN congeners could be detected in the samples of bees, pollen, and wax, with their average ΣPCNs concentrations 74.1, 96.3, and 141 pg/g dw, respectively. In relation to the predominant homologues, it was found that mono-to tri-CNs contributed with approximately 80% to the total PCNs in all the samples. Regarding the health risks of PCNs for the consumers of bee pollen, which is a popular food in China, it was found that PCNs exposure through dietary pollen posed low carcinogenic and non-carcinogenic risks for both adults and children. In order to investigate the toxicokinetics and bioaccumulation of tri- to octa-CNs in hen eggs and tissues, Wang et al. (2022b) carried out a feeding experiment with laying hens fed fly ash-contaminated feed. The eggs showed increasing concentrations of PCNs 14 days after oral exposure. Although these levels gradually decreased during the 28-day depuration period, they still exceeded the initial concentrations. The ∑PCNs levels ranged between 48.1 and 40 pg/g lipid weight (lw) in tissues of the low-exposure group, and between 67.4 and 394 pg/g lw in tissues of the high-exposure groups. The transfer rates of PCN congeners ranged between 0.27% and 23.0%, which indicated a potential transfer of PCNs from feed to eggs. Moreover, PCN distribution in the laying hens at the end of the exposure showed tissue-specific accumulation, with the high PCN levels found in the liver, spleen, and ovum. Based on these results, the authors suggested potential risks of exposure to PCNs in the food supply chain.

Dong et al. (2002b) determined the concentrations and assessed the distribution of 75 PCN congeners in 82 raw cow milk specimens from three different regions in North China, which varied in the intensity of emission sources of PCNs. “A” was the region with the highest emissions, while no PCN emission sources were detected in region “C”. The median and mean concentrations of PCNs in raw cow milk were as follows: 364 and 487 pg/g lw (region A), 150 and 261 pg/g lw (region B), and 171 and 186 pg/g lw (region C). The levels detected in region A (range 214–2050 pg/g lw) were significantly higher than those found in regions B (range 89.6–638 pg/g lw) and C (range 90.2–386 pg/g lw). The homologue profiles of PCNs were dominated by di-CNs and tri-CNs, accounting for approximately 70% of the total PCNs. It was concluded that, in general, the exposure risk posed by PCNs in cow milk for the average Chinese consumer would not be significant. In a subsequent study of the same research group, Dong et al. (2022c) determined the levels and homologue profiles of PCNs in 65 samples of milk from four dairy farms, which were situated within 10 km of an iron smelting plant in Northern China. Although not directly related to the purpose of the current review, it is interesting to note that in that study, samples of excrements, feed, plants, and soils from the farms were also analyzed. The mean and median PCN concentrations in milk were 552 and 519 pg/g lw, respectively. These values were comparable to those found in samples from other regions with important PCN emission sources (mean: 487 pg/g lw, median: 364 pg/g lw). However, they were higher than the levels in milk from less polluted Chinese areas, with no industrial PCN emission sources identified (mean: 186 pg/g lw, median: 171 pg/g lw) (Dong et al., 2022a). In a subsequent study, the same research group (Dong et al., 2023b) measured the concentrations of 75 PCN congeners in beef samples obtained from local markets in Beijing and six Chinese provinces. The total number of analyzed samples was 83, including 26 flanks, 31 rounds, and 26 shank samples. Almost all the 75 PCN congeners could be detected. The total concentrations in the 83 analyzed samples ranged between 9.9 and 239 pg/g ww, with mean and median values of 61.9 and 43.6 pg/g ww, respectively. Tri- and di-CNs were the predominant homologues, with 54% and 30% of the total PCNs, respectively. The estimated daily intakes (EDIs) for PCNs, using the beef intake, ranged between 0.0011 and 0.071 pg TEQ/kg bw, with mean and median values of 0.0089 and 0.0059 pg TEQ/kg bw, respectively. Based on these values, it was concluded that exposure to PCNs through beef consumption by an average person living in China would be minimal. Recently, the same research group (Dong et al., 2023c) measured the concentrations of PCNs in 72 samples of commercial milk-based formula produced in various countries and sold on the Chinese market. The total PCN concentrations in all the commercial cow milk formula samples from different countries were in the range 7.8-30.3 pg/g ww, with mean and median values of 15.5 and 13.6 pg/g ww, respectively. Mono- to tri-CNs were the predominant homologs in these samples, while the penta- and hexa-CNs were below their respective LODs. The TEQ values for PCNs in all the formula samples ranged between 0.0001 and 0.0019 pg TEQ/g ww, with mean and median values of 0.0008 and 0.0007 pg TEQ/g ww, respectively. It was concluded that based on exposure to PCNs, the consumption of formula in China poses a lower risk to infants and toddlers compared with consuming breast milk.

Considering that in Asian countries, the Chinese mitten crab is a popular foodstuff, Wang et al. (2022c) measured the levels of 75 PCN congeners in farmed Chinese mitten crabs (also crab compound feed and sediments) collected from 36 crab farms in Anhui Province and Shanghai. The mean concentration of the individual PCNs in all crab samples was 21.3 pg/g ww (range 5.46–43.8 pg/g ww). Di-CNs, tetra-CNs, penta-CNs, and hepta-CNs accounted for more than 85% of the total PCN concentrations in the samples of crabs collected in Anhui Province and Shanghai. Regarding potential health risks for Chinese consumers, the average EDIs associated with crab consumption were 0.0044–0.0083 and 0.0025–0.0048 pg TEQ/kg bw/day for a 63 kg adult consuming 40–75 g/day in Anhui Province and Shanghai, respectively. The authors concluded that the results would not pose significant health risks to the Chinese population from exposure to PCNs through the consumption of farmed crabs. One of the most comprehensive Chinese studies on the dietary exposure to PCNs and the health risks derived from that exposure was conducted by Li et al. (2022). These researchers determined the levels of 75 PCN congeners in 8 categories of food from 24 provinces of China. In that survey, 192 composite samples - comprising 17,280 subsamples - of fish, meat, eggs, milk, legumes, cereals, vegetables, and potatoes were analyzed. As expected, the levels of PCNs in animal-origin foods were generally higher than those found in plant-origin foods. The highest concentrations of PCNs were found in meat (range: 49.2-174.1 pg/g ww; mean: 92.0 pg/g ww), eggs (range: 7.6–159.0 pg/g ww; mean: 31.4 pg/g ww), and fish (range: 11.1–138.6 pg/g ww; mean: 27.2 pg/g ww). In contrast, among animal-origin foods, the lowest concentration corresponded to milk (range: 8.6–51.7 pg/g ww; mean: 22.2 pg/g ww). On average, it was found that meat, cereals, and vegetables were the most important contributors to the total dietary intake of PCNs, with percentages of contribution of 35.4%, 25.2%, and 19.4%, respectively. The risk assessment did not suggest significant cancer risks or potential non-carcinogenic adverse health effects for the Chinese population. However, it was also concluded that the release of PCNs from industrial thermal processes was still an important factor in contamination by these compounds, and consequently, for human exposure.

2.1.2. Korea

In Korea, Kim et al. (2018) determined the levels of 37 PCN congeners (from tetra- to octa-CNs) in 33 species of fish and seafood, while the dietary intake of PCNs through fish and seafood consumption was also estimated. A total of 165 samples of the edible parts of fish, mollusks, and crustaceans (widely and frequently consumed in the country) were analyzed. In fish samples, the highest concentrations of total PCNs corresponded to sailfin sandfish (110 pg/g ww), herring (96 pg/g ww), and salmon (53 pg/g ww), while in mollusks and crustaceans, the highest level of total PCNs was detected in mussels (29 pg/g ww). In fish samples, penta-CNs was the predominant homolog group, followed by tetra-, hexa-, and hepta-CNs. In turn, tetra-CNs was the dominant homolog group in samples of mollusks and crustaceans. The EDI of total PCNs through the consumption of fish and seafood was 570 pg/kg, with penta-CNs being the highest contributor (310 pg/kg). Regarding the specific species, the highest contribution was due to mackerel (170 pg/day), while other highly consumed seafood species in Korea, such as anchovy, flatfish, and croaker, were also important contributors to the dietary daily intakes of PCNs. In a recent study conducted by the same research group, Heo et al. (2024) investigated the concentrations and congener patterns of PCNs in foodstuffs from Korean markets. A total of 208 food samples belonging to 30 food items were analyzed. The food items followed this distribution: 9 agricultural products, 8 fishery products, and 13 processed foods. The highest mean concentrations of ΣPCNs corresponded to fishery products (70.7 pg/g ww, range 22.2- 275 pg/g ww), while the lowest levels corresponded to agricultural products (9.12 pg/g ww, range ND-39.9 pg/g ww). Regarding the distribution pattern of PCNs, it was found that tri-CNs were the most dominant group in all three food groups. In turn, tetra-CNs and penta-CNs, followed by tri-

CNs, were the dominant homologues in fishery products and processed foods. With respect to the dietary intake of PCNs by the Korean population, the mean intakes of PCN (TEQ) for the Korean population were estimated to be 0.901 and 0.601 pg-TEQ/day, for males and females, respectively.

2.1.3. *Other Asian countries*

In Pakistan, Mahmood et al. (2014) measured the concentrations of PCNs in crops of rice and wheat, while the human dietary exposure to these pollutants via these cereal crops was also estimated. A total of 14 sampling locations were selected for the sampling of cereal crops, locations within a 170 km stretch along the two tributaries of River Chenab, Punjab Province. Twenty-eight samples of rice and wheat were collected from their fields during the harvesting seasons. The average concentrations of ∑PCNs in wheat and rice samples were 0.11 ng/g (range: 0.02–0.21 ng/g) and 0.31 ng/g (range: 0.02–1.21 ng/g), respectively. Among PCN homologs, penta-CNs were predominant, with 37% for wheat samples and 40% for rice samples. Penta-CNs were followed by tetra-CNs, with percentages of 27% and 25%, for wheat and rice samples, respectively. The EDIs of ∑PCNs through consumption of wheat and rice were 0.21 ng/kg bw/day and 0.03 ng/kg bw/day, respectively.

2.2. *Europe*

2.2.1. *Ireland and United Kingdom*

Fernandes et al. (2010) performed a study aimed at examining the occurrence of PCNs in foods and determining the dietary exposure of these compounds for the population of the United Kingdom (UK). A number (44) of samples of various commonly consumed foods - fish, meat, milk and dairy products, eggs, vegetables, etc. - were analyzed for the content of the following PCN congeners: 52, 53, 66/67, 68, 69, 71/72, 73, 74, and 75. The highest levels and the widest range of PCN congeners were found in fish (average, 20 ng/kg whole weight), with the highest concentrations specifically detected in farmed and organically produced salmon: 37.29 and 34.51 ng/kg, respectively. A wide range of congeners was also detected in the samples of eggs/poultry, but the average level was comparatively lower (about 2.4 ng/kg whole weight) for the sum of the measured PCN congeners. Based on the concentrations of PCNs, it was concluded that the average dietary intake of these pollutants should not raise toxicological concerns in the UK. In another study conducted by the same research group, Fernandes et al. (2011) determined in commonly consumed Irish foods the occurrence of PCNs. The resulting human exposure to PCNs was also estimated. One hundred composite samples were analyzed: 17

samples of marine and farmed fish and shellfish, 15 samples of milk and eggs, 21 samples of carcass fat (from beef cattle, pigs, lambs, chickens, and ducks), 12 samples of liver (bovine, porcine, ovine, equine, and avian), 6 dairy products, as well as 14 other miscellaneous foods. PCNs were detected in most foods, with a range of concentrations for the sum of the measured PCNs between 0.09 and 59.3 ng/kg whole weight, for milk and fish samples, respectively. The PCN congeners 66/67, 52, and 73 were the most frequently detected. The calculated PCN TEQ values associated with the reported concentrations ranged between 0.0001 ng/kg TEQ (mainly for vegetable-based foods and some shellfish) and 0.03 ng/kg TEQ for fish. In another investigation carried out by the same research group, Rose et al. (2015) identified groups of the population who might be exposed to various environmental pollutants due to fish consumption from UK freshwater systems. For this, regional and seasonal differences in consumption of freshwater fish were considered. PCNs (together with other POPs, heavy metals, OC pesticides, etc.) were included in that investigation, which was designed to identify UK inland waterways that are likely to be contaminated either by anthropogenic activity or because of the geology of the area. The habits of anglers and others who might consume fish caught from these waters were established. Efforts were made to identify groups of the population that might be exposed to the environmental contaminants, as well as regional and seasonal differences in consumption of freshwater fish. An estimate of the extent of these habits among the various sub-groups of the population was made. The mean and median concentrations of ΣPCNs in freshwater fish were 109.75 and 43.78 μg/kg (range: 1.11-1197.33 μg/kg), based on an upper bound whole weight basis. The results of that survey confirmed the occurrence of a wide range of environmental pollutants, including PCNs, in fish. Within the same research line, but now in the marine waters of the UK, with an extension of the sampling areas to the coasts of Norway and the south of the Algarve (Portugal), a new study was conducted by Fernandes et al. (2018). A total of 182 samples of marine species (sea bass, mackerel, herring, sprats, grey mullet, sardines, turbot, halibut, various shark species, etc.) were collected, mainly from the waters around the UK and the European coastal North Atlantic. The concentrations of various POPs were determined in these samples. Regarding PCNs, the levels were measured in a sub-set of 75 samples belonging to seven species. Concentrations were reported as the sum of 12 measured PCN congeners. They ranged between 0.7 and 265 ng/kg ww, with the highest levels corresponding to sprats and mackerel: means of 67 and 68 ng/kg ww, respectively. The highest PCN levels corresponded to fish samples from the Irish Sea. On the other hand, Zhihua et al. (2019) carried out a study aimed at determining the occurrence and geospatial mapping of PCNs, PTEs (potentially toxic elements), and aflatoxins to provide effective visualization of their distribution

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within geographical regions. The focus was mainly established on the distribution of PCN contamination for different edible marine fish species from locations in the North Atlantic Ocean (mainly, but not exclusively, from waters around the UK, Ireland, and Northern France). Seventy-five samples of sea bass, herring, mackerel, mullet, and sardine were analyzed for PCNs as part of a larger study investigating also other environmental contaminants. The PCN congeners included in that survey were the following: 52/60, 53, 66/67, 68, 69, 71/72, 73, 74, and 75. The highest PCN levels corresponded to sprats and mackerel, with mean summed concentrations of 67 and 68 ng/kg ww, respectively. The geo-spatial visualization approach proposed for food contaminants (including PCNs) should allow rapid interpretation and communication of complex information.

2.2.2. *Italy*

Kannan et al. (2002) determined the concentrations and congener profiles of PCNs (and other POPs) in samples of fish from the Italian coast of the Mediterranean Sea. Bluefin tuna (liver, muscle, and fat) and swordfish (muscle and liver) were the fish species selected for that survey. The levels of PCNs, which were detected in all analyzed samples, ranged between 6.97 and 552 pg/g in muscle and fat of tuna, and from 14.6 to 62.9 pg/g in muscle and liver of swordfish.

2.2.3. *Finland*

Isosaari et al. (2006) measured the levels of PCDD/Fs, PCBs, dioxin-like PCBs, PCNs, and PBDEs in 156 composite samples of fish collected from the Baltic Sea, as well as three Finnish lakes during the period 2001–2003. The following fish species were collected from the Baltic Sea and from the three lakes, when available: burbot, Baltic herring, sprat, pike-perch, salmon, perch, pike, flounder, vendace, whitefish, river lamprey, smelt, and bream. The levels of PCNs in fish were less than 0.085 ng/g lw except for salmon, river lamprey, and herring. As it could be expected, fish species caught in the Baltic Sea but which can also live in Finnish lakes (pike, perch, pike-perch, burbot, bream, vendace, roach, and whitefish) showed, in general, higher levels of PCNs, as well as the other analyzed POPs.

2.2.4. *Latvia*

Zacs et al. (2021) performed a study aimed at providing data on selected PCNs in foods from Latvia and at estimating the dietary intake of these pollutants by the general population of the country. The analyzed foods, which are widely consumed in Latvia, were the following: milk and dairy products, eggs, meats, fish, cod liver, bread and cereal products, vegetable oils, fish oil as a food supplement (purified from POPs), and baby food samples. Twenty-six tetra- through octa-PCN congeners were determined. The highest mean PCN levels were observed in fish products, with mean and median ∑PCNs concentrations of 45.7 and 27.1 pg/g, respectively. The highest ∑PCNs concentration (286 pg/g) corresponded to a sample of cod liver. The EDIs for ∑PCNs and PCN-TEQ for the general population were 116 and 0.036 pg TEQ/kg bw, respectively. Fish and fish products provided the main contribution to the dietary "dioxin-like" burden of PCNs, with approximately 60% of the total PCN-TEQ intake.

2.2.5. France

Godére et al. (2022) conducted -for the first time in France- a survey aimed at determining the presence of PCNs in fish and seafood products, as well as to assess the health risks for French sea consumers. During the known as CALIPSO study (Leblanc et al., 2006), composite samples of the most consumed fish and seafood (collected between January and April 2005 in 4 French coastal cities: Le Havre, Lorient, La Rochelle, and Toulon) were selected based on a local Total Diet Study (TDS). PCNs were detected in 36 of the 37 samples analyzed. The mean and median concentrations of PCNs were 100 and 34 pg/g ww, with a range between 1.9 and 440 pg/g ww (medium-bound values). Crab and mackerel were the species containing the highest levels of PCNs, followed by sardine, salmon, and sea bass. Regarding the assessment of the potential health risks for consumers, it was found that salmon - followed by mackerel and sardine- made up 50–90% of total intakes of PCNs. The specific PCN-related dietary dioxin-like exposure was 0.028-0.051 pg TEQ/kg bw/week for adults, based on fish and seafood consumption only. In that same line, recently, this research group (Godére et al., 2024) determined also the levels of PCNs in 60 food products, which were acquired in 2021–2022 throughout the French territory. The foodstuffs included meat and meat products, milk and dairy products, eggs, vegetable oils, and baby foods. Simultaneously, the levels of PCDD/Fs and PCBs were also measured to allow the contribution of PCNs to the cocktail of dioxin-like pollutants to be evaluated. The concentrations of ∑PCN ranged from 2.5 to 150 pg/g (ww) (13 to 3900 pg/g lw). The median ∑PCNs concentrations in the different food categories ranged from 12 to 14 pg/g ww (excepting baby food). On a lipid weight basis, the levels were more than 10 times higher in fish and fishery products. In this recent study, Godére et al. (2024) found a low risk from dietary exposure to PCNs, with milk and dairy products being the highest contributors, followed by meat and meat products.

2.2.6. Spain

In recent decades, our research group, commissioned by the Waste Agency of Catalonia (Spain) and the Catalan Food Safety Agency, has conducted several studies focused on establishing the occurrence of various environmental contaminants in foods and the human dietary exposure to these pollutants. These have included surveys on heavy metals, PCDD/Fs, PCBs, PBDEs, PCNs, PAHs, and PCDEs, among others (Domingo et al., 1999; Llobet et al., 2008; Martí-Cid et al., 2008; Perelló et al., 2015; Aznar-Alemany et al., 2017; Martorell et al., 2012; González et al., 2021). Regarding specifically PCNs, in a first study (Domingo et al., 2003), we analyzed the concentrations of tetra- to octa-CNs, with the mean sums of tetra-octa-CN also estimated. A total of 108 composite samples of vegetables, tubers, fruits, cereals, pulses, fish and shellfish, meat and meat products, eggs, milk, dairy products, and oils and fats were analyzed. All foods were randomly acquired in various cities of Catalonia. The highest level of total PCNs was found in oils and fats (447 pg/g). Cereals (71 pg/g), fish and shellfish (39 pg/g), and dairy products (36 pg/g) showed also comparatively high levels of PCNs, while the lowest concentrations of total PCNs corresponded to milk (0.4 pg/g) and fruits (0.7 pg/g). In general, tetra-CNs were the predominant homologue in all food groups. The largest contribution to the daily PCNs intake was due to the consumption of oils and fats, and cereals. The highest and lowest daily dietary intakes of PCNs corresponded to children and seniors: 1.65 and 0.54 ng/kg bw, respectively.

With respect to the dietary intake of environmental pollutants, it is well established that fish and shellfish are -in general terms- the food group showing the highest contributions to human daily exposure to these contaminants. For that reason, we conducted in our laboratory a survey specifically aimed at determining human exposure to PCNs through the consumption of edible marine species (Llobet et al., 2007). Forty-two composite samples of 14 widely consumed species in Catalonia (sardine, tuna, anchovy, mackerel, swordfish, salmon, hake, red mullet, sole, cuttlefish, squid, clam, mussel, and shrimp) were analyzed. The highest levels of total PCNs corresponded to salmon (227 ng/kg ww), mackerel (95 ng/kg ww), and red mullet (68 ng/kg ww). In contrast, cuttlefish (3 ng/kg ww) and shrimp (5 ng/kg ww) were the species showing the lowest levels of total PCNs. Penta-CNs (60%) was the predominant contributor to total PCNs, except for cephalopods and shellfish species, for which tetra-CNs were the predominant group. Regarding the daily dietary intake of PCNs through the consumption of fish and seafood, it was estimated to be 1.53 ng for a standard male adult. The species with the highest contributions to this intake were salmon (0.41 ng/day), sole (0.28 ng/day), and tuna (0.24 ng/day). To know the temporal trend in the dietary intake of PCNs by the population of

Catalonia, in addition to fish and seafood (Llobet et al., 2007), foodstuffs belonging to the rest of the food groups that had been assessed in our first survey (Domingo et al., 2003) were again analyzed. In that new survey (Martí-Cid et al., 2008), 50 food items (meat and meat products, vegetables and tubers, fruits, eggs, cow's milk and dairy products, cereals, pulses, oils and fats, and bakery products) were selected, and composite samples were analyzed. For estimating the dietary intake of PCNs, the results regarding fish and seafood were taken from our previous study (Llobet et al., 2007). Precisely, it was noted that the highest ∑PCNs corresponded to fish and seafood (47.1 ng/g ww), followed by oils and fats, bakery products, and dairy products, with values of ∑PCNs of 21.5, 15.3, and 11.7 ng/kg ww, respectively. For a standard consumer -male adult (70 kg bw)- of Catalonia, the daily dietary intake of PCNs was estimated to be 7.25 ng, which meant an 84% decrease in relation to our previous survey (Domingo et al., 2003). This important reduction was mainly due to the decreases in the contribution of oils and fats and cereals.

2.2.7. *Analysis of PCNs in cod liver*

The occurrence and human exposure to PCNs, PCDD/Fs, and PCBs through the consumption of cod liver products have also been assessed. Falandysz and Fernandes (2020) analyzed a set of persistent and toxicologically significant PCN congeners in samples of cod liver oil and canned cod liver, which had been obtained between 1972 and 2017 from the North Atlantic region, including the Baltic Sea. The analyzed samples consisted of cod liver oil of medical grade, sourced from the Baltic Sea or the North Atlantic, as well as canned cod livers made from Baltic Sea cod. The levels of the following 19 PCNs were analyzed: 13, 27, 42, 52/60, 53, 63, 65, 66/67, 64/80, 69, 70, 71/72, 73, 74, and 75. The highest contributors to ΣPCNs were CNs #42 (TeCN), #52/60 (PeCN), #66/67, #69, #64/68, and #71/72 (HxCN) and #73 (HpCN). The concentrations of ΣPCN congeners in the medicinal grade and other cod liver oils ranged between 2050 and 13,400 pg/g[1] (period 1972–2001). In turn, in canned cod liver foods, the range of ΣPCNs was between 1670 and 2240 pg/g[1] fat (730–1050 pg/g whole weight in 2017). The PCN contribution to dioxin-like toxic equivalence (TEQ) was estimated for these samples to be in the range 1.2–15.9 pg TEQ/g. This range was notable in comparison to the EU regulated value of 1.75 pg TEQ/g for dioxins in fish oils. Falandysz et al. (2019) also carried out a retrospective investigation into the occurrence and human exposure to PCNs (also PCDD/Fs and PCBs) through the consumption of cod liver products. It was found that, over the years covered by the study, cod liver oils and cod liver products from Northern Europe showed relatively high levels of PCNs (also of PCDD/Fs and PCBs). In

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turn, cod liver oils from the Baltic Sea showed higher levels of these pollutants than the North Atlantic samples. Specifically regarding PCNs only, it was noted that the contribution to daily adult TEQ intake was relatively small.

3. Discussion and conclusions

For environmental pollutants such as PCDD/Fs, PCBs, or PBDEs, among other POPs, it is well established that air inhalation is one of the ways of human exposure to these compounds (Tue et al., 2012; Wang et al., 2017; Cai et al., 2020; López et al., 2021; Othman et al., 2022). For PCNs, whose chemical structure is similar to that of PCBs, air is also one of the potential ways of human exposure (Waheed et al., 2020; Yang et al., 2021; Klimczak et al., 2023). However, as it also occurs with other environmental pollutants, for non-occupationally exposed individuals, the diet is -in general- the main source of exposure to these POPs and also to heavy metals.

The present review shows that, in recent years, China is the country for which more scientific literature on the concentrations of PCNs in foods is available. A summary of the most relevant studies carried out in China and other Asian countries is presented in Table 1. Various surveys conducted in China have measured the levels of PCNs in a number of foodstuffs. Notwithstanding, data regarding the complete dietary daily intake of PCNs is limited. Most studies were focused on the EDIs calculated only with the specific food items included in the respective surveys (fish and seafood, milk, meats, etc.). To the best of our knowledge, in the scientific literature, there is not any article reporting the results of studies aimed at estimating the global human dietary exposure to PCNs in China. Despite this, the results of most surveys carried out in that country suggest that dietary exposure to PCNs from the analyzed food items should not mean human health risks. In Korea, recently, Heo et al. (2024) reported the daily dietary intake of PCNs, which was estimated after analyzing their levels in 9 agricultural products, 8 fishery products, and 13 processed foods. To date, that is the first and most complete survey conducted in that country (Table 1).

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Country/region/city	Food samples analyzed	Concentrations of PCNs	Remarks	Reference
China, cities of Guangzhou and Zhoushan	Fish, cephalopods, shrimps, crabs, bivalves	∑PCNs, range: 93.8-1300 pg/g lipid	PCNs were detected in all seafood samples	Jiang et al. (2015)
China	Cheese, butter	∑PCNs, ranges: cheese, 5.6-103 pg/g ww and butter, 5.0-199.9 pg/g ww	PCN 73 was the predominant congener followed by PCNs 63, 66/67 and 69	Li et al. (2015)
China, Bohai Rim Region	17 marine fish species	∑PCNs, range: 7.3-214 pg/g ww. The highest level was found in ditrema samples	Predominant congeners were tri-CNs. Toxicological concerns posed by PCNs were considered negligible for the consumers	Cui et al. (2018)
China, Beijing	Blood samples of 8 species of animals, whose meats are commonly consumed	Median and mean: 826 and 756 pg/L (range: 305 pg/L in donkey-987 pg/L in pig).	Mono-CNs were the predominant group of homolgues. Chicken, beef and pork were the main contributors to total TEQs (61%, 27% and 5.9%, respectively)	Han et al. (2018)
North China (3 regions)	Raw cow milk	Means: 487, 261 and 186 pg/g lw, for the most to the less contaminated region	Di-CNs and tri-CNs were the dominant homologues. Exposure to PCNs from milk consumption would not be of concern	Dong et al. (2022b)
North China	Milk from 4 dairy farms near an	Mean and median values: 552 and 519 og/g lw	PCN levels were comparable to those found in milk samples	Dong et al. (2022c)

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Country/region/city	Food samples analyzed	Concentrations of PCNs	Remarks	Reference
	iron smelting plant		collected in Chinese areas with important emission of PCNs	
China, Beijing and six Provinces	Beef (flanks, round and shank samples) from local markets	∑PCNs, mean and median values: 61.9 and 43.6 pg/g ww (range: 9.9-239 pg/g ww)	Tri and di-CNs were the predominant homologues. Using beef consumption, the EDIs ranged: 0.0011-0.071 pg TEQ/kg bw	Dong et al. (2023b)
Chinese market	Commercial milk-based formulas	∑PCNs, mean and median values: 15.5 and 13.6 pg/g ww (range: 7.8-30.3 pg/g ww)	Mono- and tri-CNs were the predominant homologues. TEQ values ranged between 0.0001 and 0.0019 pg TEQ/g ww	Dong et al. (2023c)
China, Anhui Province and Shanghai	Mitten crabs	Mean value: 21.3 pg/g ww (range 5.46–43.8 pg/g ww)	No significant health risks were associated with exposure to PCNs through farmed Chinese mitten crab consumption.	Wang et al. (2002c)
China, 24 Provinces	Fish, meat, eggs, milk, legumes, cereals, vegetables and potatoes	The highest concentrations were found in meat (range: 49.2-174.1 pg/g ww; mean: 92.0 pg/g ww), eggs (range: 7.6–159.0 pg/g ww; mean: 31.4 pg/g wwt) and fish (range: 11.1–138.6 pg/g ww; mean: 27.2 pg/g ww).	The risk assessment did not show significant cancer risks or potential non-carcinogenic adverse health effects for the Chinese population.	Li et al. (2022)
Korea, various cities	Fish, mollusks and crustaceans	The highest ∑PCNs to sailfin sandfish (110 pg/g ww),	The EDI of total PCNs through the	Kim et al. (2018)

Country/region/city	Food samples analyzed	Concentrations of PCNs	Remarks	Reference
		herring (96 pg/g ww), and salmon (53 pg/g ww). In mollusks and crustaceans, the highest ∑PCNs was detected in mussels (29 pg/g ww)	consumption of fish and seafood was 570 pg/kg, being penta-CNs the highest contributor (310 pg/kg).	
Korea, various markets	Nine agricultural products, 8 fishery products, and 13 processed foods	The highest mean ΣPCNs corresponded to fishery products (70.7 pg/g ww, range 22.2- 275 pg/g ww). The lowest levels corresponded to agricultural products (mean 9.12 pg/g ww, range ND-39.9 pg/g ww)	The estimated mean intakes of PCN for the Korean population were 0.901 and 0.601 pg-TEQ/day (males and females, respectively)	Heo et al. (2024)
Pakistan, Punjab Province	Wheat and rice crops	The average ∑PCNs in wheat and rice samples were 0.11 ng/g (range: 0.02–0.21 ng/g) and 0.31 ng/g (range: 0.02–1.21 ng/g), respectively.	The EDIs of ∑PCNs through consumption of wheat and rice were 0.21 and 0.03 ng/kgbw/day, respectively	Mahmood et al. (2014)

Table 1. A summary of some recent publications on the occurrence of PCNs in foods consumed by the Asian population

Table 2 summarizes information on studies conducted in European countries. Most surveys were also performed in the current century. Just a few studies were conducted in the 1990s, while a few others were published in 2000, being focused on determining the levels of PCNs in fish and shellfish samples (Domingo et al., 2004). The current information on the concentrations of PCNs in food and their intake through the diet is now more abundant than that available in our previous review (Domingo, 2004). However, to draw conclusions on the health risks that the dietary exposure to PCNs can mean remains still difficult. In this sense, the recent EFSA report on the presence of PCNs in feed and food and the health risks for animals and humans has not provided an acceptable safety value for human daily dietary intake (or allowable dietary exposure) of PCNs (EFSA, 2024). It is due to the scarce available information on that topic. On the other hand, the lack of published data from complete market-based studies in most countries is notorious, while there is a total absence of available scientific publications in American countries, including the USA, Canada, Mexico, or Brazil, for example. There is no data on human total daily intake through food consumption conducted in other relevant countries such as India, Australia, or Japan, among many others, including all the African countries.

Country/region/city	Food samples analyzed	Concentrations of PCNs	Remarks	Reference
United Kingdom (UK)	Commonly consumed foods: fish, meat, milk and dairy products, eggs, vegetables, etc.,	The highest levels and of PCN congeners were found in fish (average, 20 ng/kg whole weight), being the highest concentrations detected in farmed and organically produced salmon: 37.29 and 34.51 ng/kg, respectively	The average dietary intake of PCNs should not mean toxicological concerns in the UK	Fernades et al. (2010)
Ireland	Marine and farmed fish and shellfish, milk, eggs, carcass fat (from beef cattle, pigs, lambs, chickens and duck), liver (bovine, porcine, ovine, equine and avian), dairy products, and other miscellaneous foods.	∑PCNs ranged between 0.09 and 59.3 ng/kg whole weight for milk and fish samples, respectively	PCN TEQ values ranged between 0.0001 ng/kg TEQ (mainly for vegetable-based foods and some shellfish) and 0.03 ng/kg TEQ for fish	Fernandes et al. (2011)
UK marine regions, Norway coast, and south Algarve (Portugal)	Seven edible marine species	∑PCNs ranged between 0.7 and 265 ng/kg ww. The highest levels corresponded to sprats and mackerel, with means of 67 and 68 ng/kg ww, respectively.	Considering the precedence of the sample, the highest PCN levels corresponded to fish from the Irish Sea	Fernades et al. (2018)
Italy	Bluefin tuna (liver, muscle and fat) and swordfish (muscle and liver)	The levels ranged between 6.97 and 552 pg/ in muscle and fat of tuna, and from 14.6 to 62.9	Exposure to PCNs should not mean health risks	Kannan et al. (2002)

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Country/region/city	Food samples analyzed	Concentrations of PCNs	Remarks	Reference
		pg/g in muscle and liver of swordfish.		
Finland (Baltic Sea and 3 Finish Lakes)	Fish: burbot, Baltic herring, sprat, pike-perch, salmon, perch, pike, flounder, vendace, whitefish, river lamprey, smelt and bream.	The levels of PCNs in fish were lower than 0.085 ng/g lw except for salmon, river lamprey, and herring	Fish species caught in the Baltic Sea, showed, in general, higher levels of PCNs	Isosaari et al. (2006)
Latvia	Milk and dairy products, eggs, meats, fish, cod liver, bread and cereal products, vegetable oils, fish oil as a food supplement, and baby foods.	Mean and median ∑PCNs were 45.7 and 27.1 pg/g, respectively. The highest ∑PCNs concentration (286 pg/g) corresponded to a sample of cod liver.	The EDIs for ∑PCNs and PCN-TEQ for the general population were 116 and 0.036 pg TEQ/kg bw, respectively. Fish and fish products provided the main contribution (60%)	Zacs et al. (2021)
France	Various species of widely consumed fish and seafood	Mean and median ∑PCNs were 100 and 34 pg/g ww, with a range between 1.9 and 440 pg/g ww. Crab and mackerel were the species containing the highest levels of PCNs	The PCN related dietary dioxin-like exposure was 0.028-0.051 pg TEQ/kg bw/week for adults	Godére et al. (2022)
France	Meat and meat products, milk and dairy products, eggs, vegetable oils, and baby foods	∑PCN ranged from 2.5 to 150 pg/g (ww). The median ∑PCNs in the different food categories ranged from 12 to 14 pg/g ww	Milk and dairy products were the highest contributors, followed by meat and meat products.	Godére et al. (2024)

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Country/region/city	Food samples analyzed	Concentrations of PCNs	Remarks	Reference
Spain, Catalonia	Vegetables, tubers, fruits, cereals, pulses, fish and shellfish, meat and meat products, eggs, milk, dairy products, and oils and fats	The highest ∑PCNs was found in oils and fats (447 pg/g), followed by cereals (71 pg/g), fish and shellfish (39 pg/g), and dairy products (36 pg/g)	The highest and lowest daily dietary intakes of PCNs corresponded to these age groups: children and seniors, with 1.65 and 0.54 ng/kg bw, respectively.	Domingo et al. (2003
Spain, Catalonia	Sardine, tuna, anchovy, mackerel, swordfish, salmon, hake, red mullet, sole, cuttlefish, squid, clam, mussel and shrimp	The highest levels of ∑PCNs corresponded to salmon (227 ng/kg ww), mackerel (95 ng/kg ww), and red mullet (68 ng/kg ww).	The daily dietary intake of PCNs through the consumption of fish and seafood was estimated to be 1.53 ng for a standard male adult.	Llobet et al. (2007)
Spain, Catalonia	Meat and meat products, vegetables and tubers, fruits, eggs, cow's milk and dairy products, cereals, pulses, oils and fats, and bakery products	Oils and fats, bakery products, and dairy products, showed the highest ∑PCNs: 21.5, 15.3 and 11.7 ng/kg ww, respectively	In relation to the daily dietary intake of PCNs it was estimated to be 7.25 ng, which meant an 84% of decrease in relation to the previous survey by Domingo et al. (2003).	Martí-Cid et al. (2008)

Table 2. A summary of some recent publications on the occurrence of PCNs in foods consumed by the European population

In the current review, differences among those countries/regions for which results are available have been observed. Doubtless, it is due to the different origins of the food items included in the respective surveys, which belong to regions/areas potentially contaminated by PCNs at different levels. Another important difference is due to the number of individual PCN congeners analyzed in the respective surveys (not only in the specific analyzed PCNs, but also in their total number), and even the analytical techniques, including the detection limits. With the potential existence of 75 PCN congeners, most PCNs were only analyzed in a few studies, while other surveys included only some congeners.

In summary, although the available information regarding human health risks of PCNs due to dietary exposure is still currently rather limited, the results of the studies conducted in the present century in various countries (mainly China) seem to suggest that the risks are not significant. On the other hand, considering that PCNs are included in the list of POPs under the Stockholm Convention (aimed at protecting human health and the environment from the effects of these POPs), it must be expected that environmental levels of PCNs will continue to reduce. This should have a direct repercussion on the concentrations of these pollutants in foodstuffs and, therefore, on a consequent reduction in dietary exposure to PCNs. An example of this would be the result of a study conducted some years ago aimed at evaluating the temporal trend of the dietary intake of PCNs (Martí-Cid et al., 2008). It showed a reduction of the daily dietary intake of PCNs of 84% in relation to a previous survey conducted in the same country (Domingo et al., 2003). Although in Catalonia, no data are available since then, the reductions in the environmental concentrations of PCNs, and consequently the levels in foods, are expected to have been relevant.

Statements and Declarations

Competing Interests

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.

Data Availability

Data will be made available on request.

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