

# OCCURRENCES AND ESTIMATED DIETARY INTAKES OF POLYCHLORINATED NAPHTHALENES IN AQUATIC FOODS

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## Introduction

Polychlorinated naphthalenes (PCNs) is a group of highly toxic persistent organic pollutants including 75 congeners. Structure of PCNs feature with one to eight chlorine atoms as substituents on the naphthalene rings. Reported toxicities<sup>1-10</sup> of PCNs include dermal lesions, oxidative stress, carcinogenicity, neurotoxicity, chloracne, liver dysfunction, and even death. PCNs have been detected in a wide variety of environmental samples<sup>11-18</sup>. A few studies also reported PCNs concentrations in human tissues samples, which indicated the presence of human exposure to PCNs<sup>19-28</sup>. Due to their ubiquitous presence in different environmental matrices and potential toxicities to human body, PCNs were listed in the Annex A and C of Stockholm Convention in 2015.

Oral intake via foodstuffs is considered as the most important PCNs exposure route of the general population due to the bioaccumulation of PCNs in food web. Occurrences of PCNs in the commonly consumed foods and marine and aquatic biota have been reported<sup>29-39</sup>. Domingo et al.<sup>30</sup> determined the concentration of TetraCN to OctaCN in 11 food groups randomly acquired in seven cities of Catalonia in 2000. The highest concentration was 447 ng/kg wet weight (oils and fats), followed by 71 ng/kg wet weight (cereals), 39 ng/kg wet weight (fish and shellfish), and 36 ng/kg wet weight (dairy products). In another study from Domingo et al.<sup>38</sup> also corresponded to Catalonia in 2006, they found a different distribution of PCNs concentration in foods, in which the highest concentration was 47 ng/kg wet weight (fish and seafood), followed by 22 ng/kg wet weight (oils and fats), 15 ng/kg wet weight (bakery products), and 12 ng/kg wet weight (dairy products). Accumulation of PCNs in edible biota increased the possibility of human exposure. However, occurrences and health risks of PCNs in dietary foods from China on a national scale have never been reported.

In the present study, the concentrations of 75 PCN congeners in aquatic foods from 23 provinces of China were measured by isotope dilution high resolution gas chromatography-high resolution mass spectrometry (HRGC-HRMS). We characterize PCN concentrations and congener profiles in aquatic foods from China and estimated the daily intakes of PCNs via food intake for general population on a national scale. It was expected that this study could provide significant information on health risk assessment of PCN exposure in China.

## Materials and methods

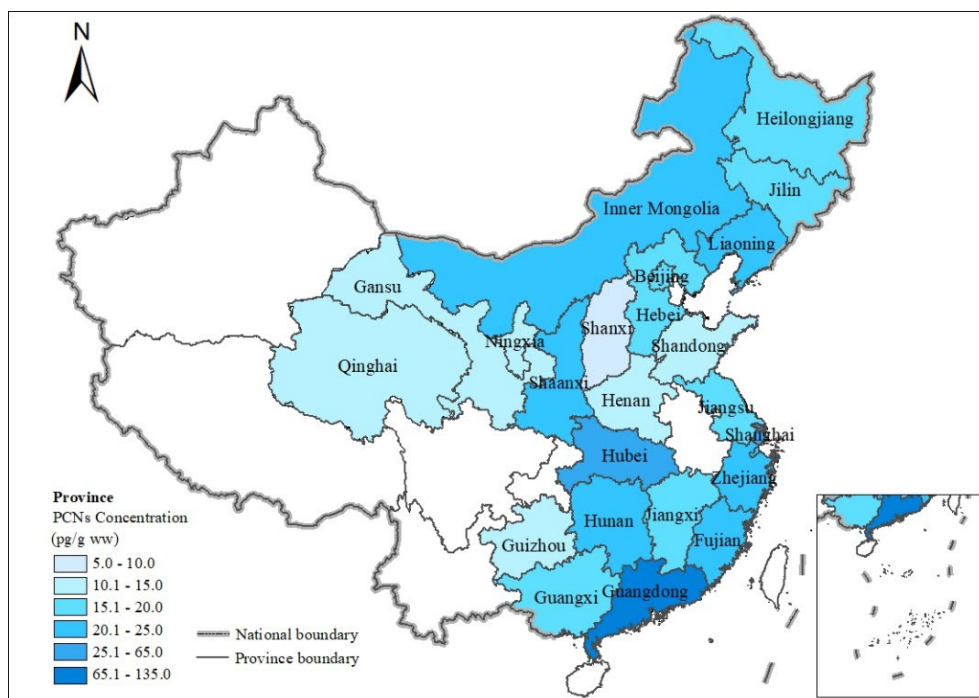
This study analyzed pooled aquatic food samples from 23 provinces in China. The pooled samples were obtained from the China National Center for Food Safety Risk Assessment and were part of the sixth Chinese TDS. Sample collection and daily food consumption surveys were conducted according to a standard procedures<sup>40</sup>.

Aquatic foods at each sampling site were locally collected and cooked. Individual samples from the same province were then pooled into one pooled sample. Each individual in a household participated in a three-day dietary recall over survey. The average of the individual aquatic food consumption in the same province was considered as the aquatic food consumption for a “standard” Chinese man (18–45 years old, 63 kg body weight, and employed in light physical work) at the province. Results of PCNs concentrations detected in this study multiple by daily food consumption from the TDS survey was considered as the estimated dietary intakes of PCNs for a “standard” Chinese man<sup>41</sup>.

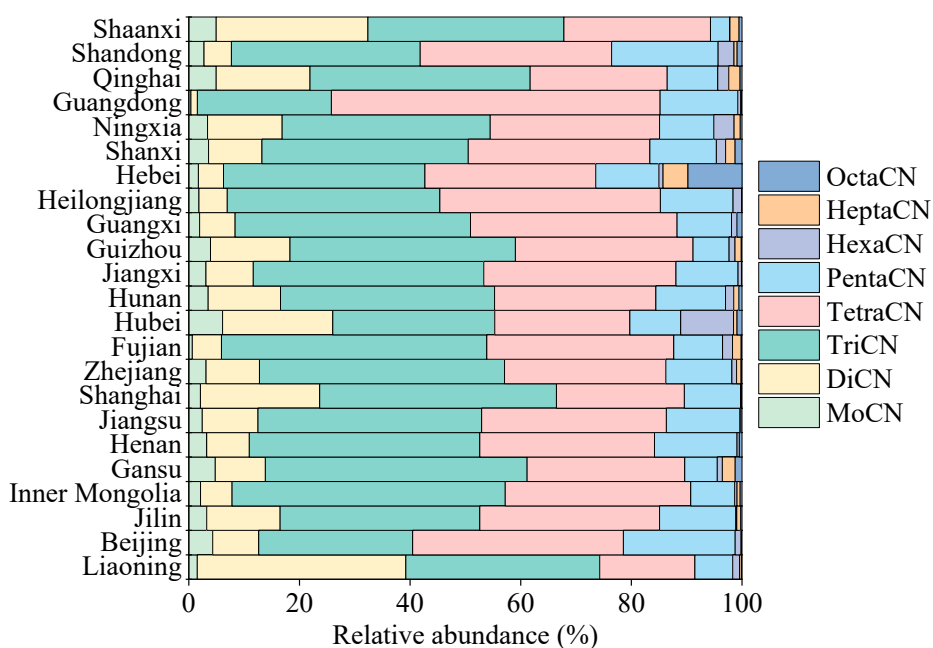
All samples were stored in a refrigerator at –20°C until analysis. Approximately 3 g dry weight was extracted using ASE 350 (Thermo Fisher Scientific, Waltham, MA, USA). The extract is concentrated to about 1–2 mL and purified with an acid silica gel column, a multi-layer silica column, and a basic alumina column. Finally, the extracts were analyzed using high-resolution gas chromatography combined with high-resolution mass spectrometry (Thermo Fisher Scientific, Waltham, MA, USA). Procedural blanks were simultaneously analyzed along with the aquatic samples using the same method. A small amount of MoCN, DiCN, and TriCN was found in the blank samples (lower than 20% of PCNs found in the aquatic food samples). The average recoveries of <sup>13</sup>C<sub>10</sub>-labeled congeners were in the range of 52%–84%.

## Results and discussion

PCNs were detected in all the 23 pooled aquatic samples analyzed in this study. **Figure 1** shows the summed concentrations of 75 PCN congeners in aquatic food samples from 23 provinces of China. Summed concentrations of 75 PCN congeners were in the range of 9.4 pg/g wet weight to 133.1 pg/g wet weight with an average of 24.5 pg/g wet weight, which is comparable to the concentrations of PCNs in fish and seafood samples from Spain reported by Domingo et al<sup>30</sup>. **Figure 2** illustrated the homolog profiles of PCNs in pooled aquatic food samples from 23 provinces of China. TriCN and TetraCN were the dominated PCN congeners in aquatic food samples.



**Figure 1.** Sum concentration of 75 polychlorinated naphthalenes (PCNs) in pooled aquatic food samples from 23 provinces of China.



**Figure 2.** Homolog profiles of polychlorinated naphthalenes (PCNs) in pooled aquatic food samples from 23 provinces of China.

Jiang et al.<sup>39</sup> conducted a research on the concentration of PCNs in various seafood from Guangzhou (the capital city of Guangdong province of China), in which they found that concentrations of PCNs in seafood ranged from 93.8 pg/g lipid weight (crabs) to 1300 pg/g lipid weight (cephalopods) and 545 pg/g lipid weight (fish). The average lipid content of the aquatic food samples in this study was 10.1%. In this case, the average lipid weight concentration of the aquatic food samples was 242.6 pg/g lipid weight. The average concentration of PCNs in pooled aquatic food samples was at a relatively lower scale when compared to Jiang's report. However, it should be noticed that the concentration of the pooled aquatic food samples in Guangdong province was the highest of the studied provinces.

Dietary intake was considered as a major pathway for exposure of humans to PCNs. The dietary intakes of PCNs via aquatic foods in this study were estimated using the PCNs concentrations we detected and data on daily food consumption survey from the TDS. The average daily consumption of the 23 provinces was 30.0 g/day. The average estimated daily consumption of PCNs via aquatic foods for the general population from 23 provinces was 919.2 pg/g-day. Overall, the concentrations, congener profiles, and estimated dietary intake of PCNs in aquatic foods reported in our study could provide significant information on health risk assessment of PCN exposure in China.

### Acknowledgements

We gratefully acknowledge support from the National Key R&D Program of China (Grant No. 2017YFC1600500), CAMS Innovation Fund for Medical Science (Grant No. CIFMS 2019-I2M-5-024), the second Tibetan Plateau Scientific Expedition and Research Program (STEP) (Grant No. 2019QZKK0605) and CAS Interdisciplinary Innovation Team (Grant No. JCTD-2019-03).

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