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Seasonal variation of atmospheric polychlorinated biphenyls and polychlorinated naphthalenes in Japan

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HIGHLIGHTS

- Different seasonality trends of atmospheric PCBs and PCNs were confirmed in Japan.
- The PCBs peaked in summer as expected of normal POPs behavior.
- But the PCNs showed relatively elevated concentrations in winter.
- Seasonality of the PCNs was marked by changes in emission origins of the tri-CNs.

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ABSTRACT

This study investigated the seasonality of atmospheric polychlorinated biphenyls (PCBs) and polychlorinated naphthalenes (PCNs) in Japan. Polyurethane foam (PUF) disk passive air samplers (PAS) were deployed simultaneously at 55 sites in spring 2008, summer 2008 and winter 2008/09. Sampler deployment spanned 8 continuous weeks in each season. The non-outlier ranges of the two pollutants (ng/sample) were as follows; $\sum_{190}$PCBs: 6.5–38.6 (spring), 43.5–220.5 (summer) and 25.9–136 (winter); and $\sum_{63}$PCNs: 0.4–3.9 (spring), 0.7–7.1 (summer) and 1.1–9.2 (winter). The corresponding values in air were $\sum_{190}$PCBs (pg m^{-3}): 33–197 (spring), 222–1125 (summer) and 132–694 (winter); and $\sum_{63}$PCNs (pg m^{-3}): 2.2–20 (spring), 3.5–36 (summer) and 5.7–47 (winter), when sampling rate of 3.5 m^3 day^{-1} was assumed. Thus, the PCBs peaked in summer, while the PCNs mostly peaked in winter; there was an apparent contrast in their seasonality in Japan. For the PCNs, seasonal variability was significant at rural than urban sites. Normally, POPs would show relatively increased air content in summer due to vaporization effect. The PCNs appeared to deviate from such a trend because of overriding input of tri-CNs presumably transported from long range by northwesterly winds in the winter season. The dioxin-like fractions of either pollutant were reduced in winter by about 30–50%.

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1. Introduction

Polychlorinated biphenyls (PCBs) and polychlorinated naphthalenes (PCNs) are organohalogens characterized by high level of persistence, toxicity and bioaccumulativeness and subject to long range transport (UNEP, 2010). PCBs belong to the initial list of chemicals to be designated as persistent organic pollutants (POPs) (the so called "Dirty Dozen"), while the PCNs are presently regarded as candidate POPs. The atmospheric burden of POPs is of interest considering that the atmosphere constitutes a major pathway by which these pollutants are distributed globally (Gouin et al., 2004).

The burden of POPs in the atmosphere is regulated by various meteorological factors, notably temperature and wind. For instance, POPs show temperature-dependent fluxes between the atmosphere and surfaces of soil and water (Cousins et al., 1999; Yan et al., 2008; Nizzetto et al., 2010). They are also distributed by wind to distant places (Gouin et al., 2004; Schmidt, 2010; Hogarh et al., 2012b). As changes occur in these meteorological factors with season, it brings variations to the burden of POPs in the atmosphere.

As a general phenomenon, lighter congeners (lower chlorinated) and heavier congeners (highly chlorinated) POPs respond differently to the effects from meteorological factors. The lower

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chlorinated congeners show greater response to temperature, for instance. They vaporize easily in warm conditions, especially, in summer. PCBs characteristically behave in this manner and show elevated content of tri- and tetra-CBs in summer (Takasuga et al., 2004; Kim and Masunaga, 2005; Anezaki and Yamaguchi, 2011). On the contrary, in few instances when the seasonality of atmospheric PCNs was reported, the burden was relatively greater in winter or mostly outside the summer season (Helm et al., 2004; Takasuga et al., 2004; Manodori et al., 2006b; Wang et al., 2012). Thus, atmospheric PCBs and PCNs seem to show different seasonality. The reason for this is not very clear, but it presupposes that there are still knowledge gaps regarding the seasonality of atmospheric POPs that should be studied. One key question is why PCNs should peak mostly in winter, contrary to the behavior of most POPs. The present study sought to help us understand some of these issues.

Atmospheric PCBs and PCNs were monitored across the entire of Japan in spring, summer and winter 2008/09, applying PUF-disk passive sampling. Previous reports on the seasonality of atmospheric PCBs and PCNs in Japan are few (Takasuga et al., 2004; Kim and Masunaga, 2005; Anezaki and Yamaguchi, 2011), all of which applied active sampling and consequently limited in spatial coverage. With the benefit of extensive spatial coverage, the present study hopefully, should complement the existing information from active sampling studies in explaining the seasonality of atmospheric PCBs and PCNs in Japan.

2. Materials and methods

2.1. Study area and sampling process

The study was conducted in Japan with monitoring sites distributed nationwide (Fig. 1). Fifty-five sampling sites, same as in Hogarh et al. (2012a), were monitored in three different seasons – spring 2008, summer 2008 and winter 2008/09. The data for spring 2008 have been reported previously in Hogarh et al. (2012a), but included in the present study for purposes of comparison with the summer and winter seasons. PUF-disk PAS were simultaneously deployed at all 55 sampling sites for 8 continuous weeks in each season. The specific sampling periods were: spring (12th Mar. – 16th May 2008), summer (8th Aug. – 4th Oct. 2008) and winter (26th Nov. 2008–21st Jan. 2009). The sampling process in the respective seasons followed similar procedure as in Hogarh et al. (2012a).

2.2. Chemical analysis and quality control

The chemical analytical procedures have been described in detail in Hogarh et al. (2012a). Briefly, harvested PUFs were Soxhlet extracted with acetone for about 12 h (exceeding 50 cycles of circulation), the extract was taken through various concentration and cleanup steps, and then screened for 202 PCB and 63 PCN congeners using high resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS).

Average percentage recoveries were generally low for the di-CBs, mostly below 50%. Therefore in this report, we concentrated on the tri- to octa-chlorinated homologs of both pollutants. Nona- and deca-PCBs were not screened. So, effectively, the reported levels reflect $\sum_{190}$PCBs and $\sum_{63}$PCNs. Among seasons, the range of the average recoveries of PCB homologs were: tri-CBs (92–143%), tetra-CBs (92–101%), penta-CBs (87–125%), hexa-CBs (88–137%), hepta-CBs (79–121%) and octa-CBs (78–116%); and for PCNs: tetra-CNs (76–97%), penta-CNs (80–93%), hexa-CNs (83–90%), hepta-CNs (108–125%) and octa-CNs (98–125%). Peaks were recognized when signal to noise ratio was greater than 3:1. The method detection limit (MDL) derived from laboratory blanks ranged 0.01–0.17 ng/sample for PCB congeners and 0.001–0.03 pg/sample for the PCNs.

2.3. Statistical analysis

Data were summarized using descriptive statistics and differences among data sets were evaluated applying ANOVA and *t*-test, where appropriate. Differences amounting to p-value < 0.05 were considered significant. Data were also evaluated for outliers applying Statistica 7 software (StatSoft, Tulsa, OK, USA).

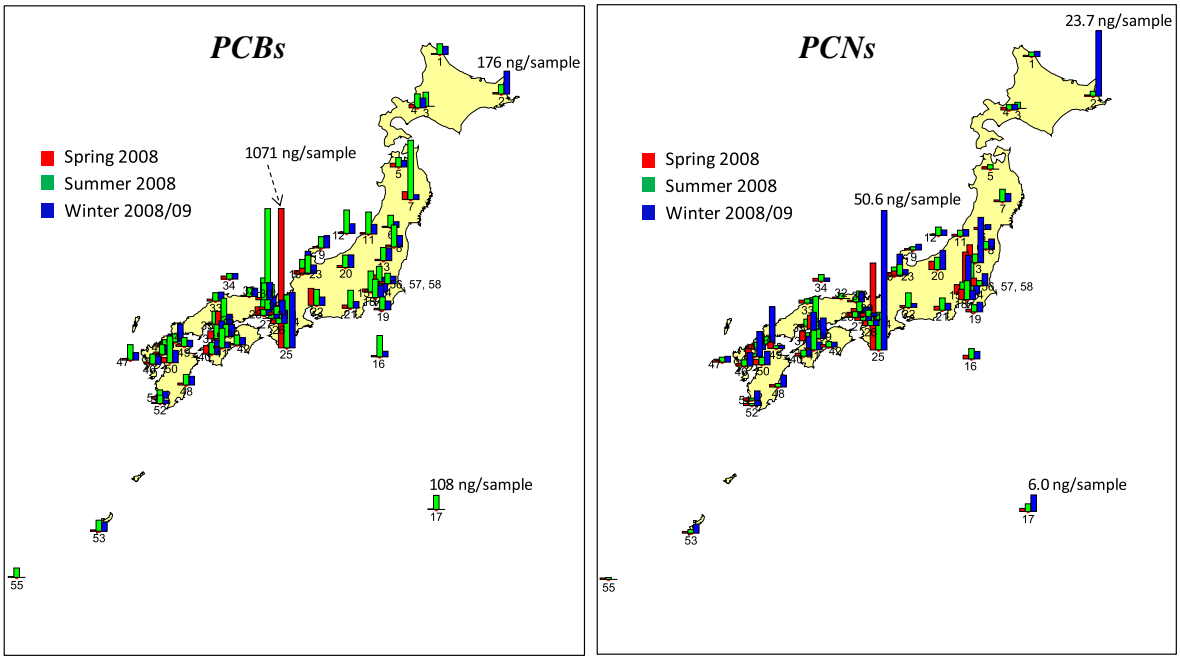

Fig. 1. Spatial distribution of atmospheric PCBs and PCNs in different seasons in Japan (sampling points are indicated by site numbers on map).

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