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Does an analysis of polychlorinated biphenyl (PCB) distribution in mountain soils across China reveal a latitudinal fractionation paradox?

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A B S T R A C T

Organic and mineral soil horizons from forests in 30 mountains across China were analysed for polychlorinated biphenyl (PCB). Soil total organic carbon (TOC) content was a key determinant of PCB distribution explaining over 90% of the differences between organic and mineral soils, and between 30% and 60% of the variance along altitudinal and regional transects. The residual variance (after normalization by TOC) was small. Tri- to tetra-CB levels were higher in the South in relation to high source density and precipitation. Heavier congeners were instead more abundant at mid/high-latitudes where the advection pattern was mainly from long range transport. This resulted in a latitudinal fractionation opposite to theoretical expectations. The study showed that exposure to sources with different characteristics, and possibly accumulation/degradation trends of different congeners in soils being out-of-phase at different latitudes, can lead to an unsteady large scale distribution scenario conflicting with the thermodynamic equilibrium perception.

1. Introduction

Persistent organic pollutants (POPs) have been the focus of research and regulation due to their persistence, ubiquitous distribution and bioaccumulative and toxic properties (Aichner et al., 2013; Jones and de Voogt, 1999; Klanova et al., 2011; Zhang et al., 2008b). Polychlorinated biphenyls (PCBs) represent a class of POPs with a broad range of physical–chemical properties (Bozlaker et al., 2008), ideal for investigating the mechanisms involved in the control of fate and global distribution of POPs in general (Aichner et al., 2013).

Although PCBs are banned in most parts of the world, primary emissions continue mostly from old equipment and inadequate waste management (Breivik et al., 2002; Li et al., 2009). Secondary emissions from environmental reservoirs such as soils, vegetation and oceans hosting PCBs burdens emitted during the past, also significantly contribute in feeding long-range atmospheric transport (e.g. (Lammel and Stemmler, 2012)).

Predicting the future of POP exposure requires understanding the dynamic balance between atmospheric sources, atmospheric depositions, environmental degradation and remobilization from environmental reservoirs. Analyzing the current regional and global distribution of the environmental burden of POPs in the major reservoir compartments (such as soils) represent an effective approach to gather information useful for predicting possible future exposure conditions (Meijer et al., 2003).

China has 134 million hectares of forested land including a range of different biomes including subtropical forests, boreal forests, and semi-arid environments (Fang et al., 2001). Large geographic scale, climate variability, complex topography, ecosystem diversity, and

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densely populated conglomerates in well-defined areas make China a very interesting case study (Jiang et al., 1999).

In order to provide an assessment of anthropological and environmental controls on POP distribution, the patterns of PCBs in forest soils of several mountain sites in China were investigated in this study, considering gradients such as soil depth, soil type, altitude, latitude and distance from urban/industrial clusters.

2. Materials and methods

2.1. Monitoring design

Thirty mountain sites across China were chosen (shown in Fig. 1). In each mountain site, up to four sampling locations were selected along the same aspect on the altitudinal transect. In total 159 forest-soil samples were collected in 82 locations from the 16th of May 2012 to the 15th of March 2013. Major gradients included: latitude ranging between 21° and 53°; altitude ranging between 200 m and 3800 m; yearly mean temperature (*ymt*) and averaged total yearly precipitation (*typ*) ranging −6 °C–21 °C and 245 mm–2129 mm, respectively; distance from the major urbanized areas (assumed here as proxy of major primary sources locations) ranging between a few tenths to several hundred km.

The monitoring plan was defined taking into consideration the need of sampling in all major accessible mountain areas of China, achieving homogeneous geographical distribution, and collecting multiple samples along slopes at locations differing at least 200 m in elevation.

2.2. Sampling of soil

Three small trenches located at about 5 m distance from each other were excavated at the depth of 30 cm in each individual altitude location. Vegetation litter was carefully removed and a preliminary classification of the soil layers was performed in-situ based on colour and structure of the material present in each horizon. Samples from the O- (organic) and A- (mineral) horizons were collected separately from each trench using a metal spoon, folded in aluminium foil, placed in polyethylene zip-bags, cooled and transported to the laboratory where their mass, water content, bulk density and total organic carbon (TOC) content were determined. The samples were then freeze dried and stored at −20 °C for maximum 3 months before chemical analysis began.

2.3. Chemical analysis

Dry samples were sieved in order to remove stones and aggregates larger than 2 mm. The samples from each trench were homogeneously mixed to create aggregated samples of the O-horizon and A-horizon reflecting average conditions of the sampling location at the selected altitude. Sample extraction and preparation, instrumental analysis and adopted quality assurance and control methods were consistent with those described in a previous study (Huang et al., 2013). Details on analytical methods are also reported in the Supplementary information (SI) available on line.

2.4. Geographic information

A set of spatially explicit datasets were used in this study to feed the exploratory analysis performed in order to assess physical and anthropogenic influences on contaminant distribution. Geo-referenced datasets of elevation, *ymt*, *typ* (0.5′ resolved) and human population counts (2.5′ resolved) were obtained from WorldClim (Jurado et al., 2004; WORLDCLIM) and CIESIN (CIESIN; Gioia et al., 2006). Geographic information was elaborated using ArcGIS Desktop software. Multiannual averaged *ymt* of individual sites was calculated using monthly averages of time series (1950–2000). The information on human population distribution was used as

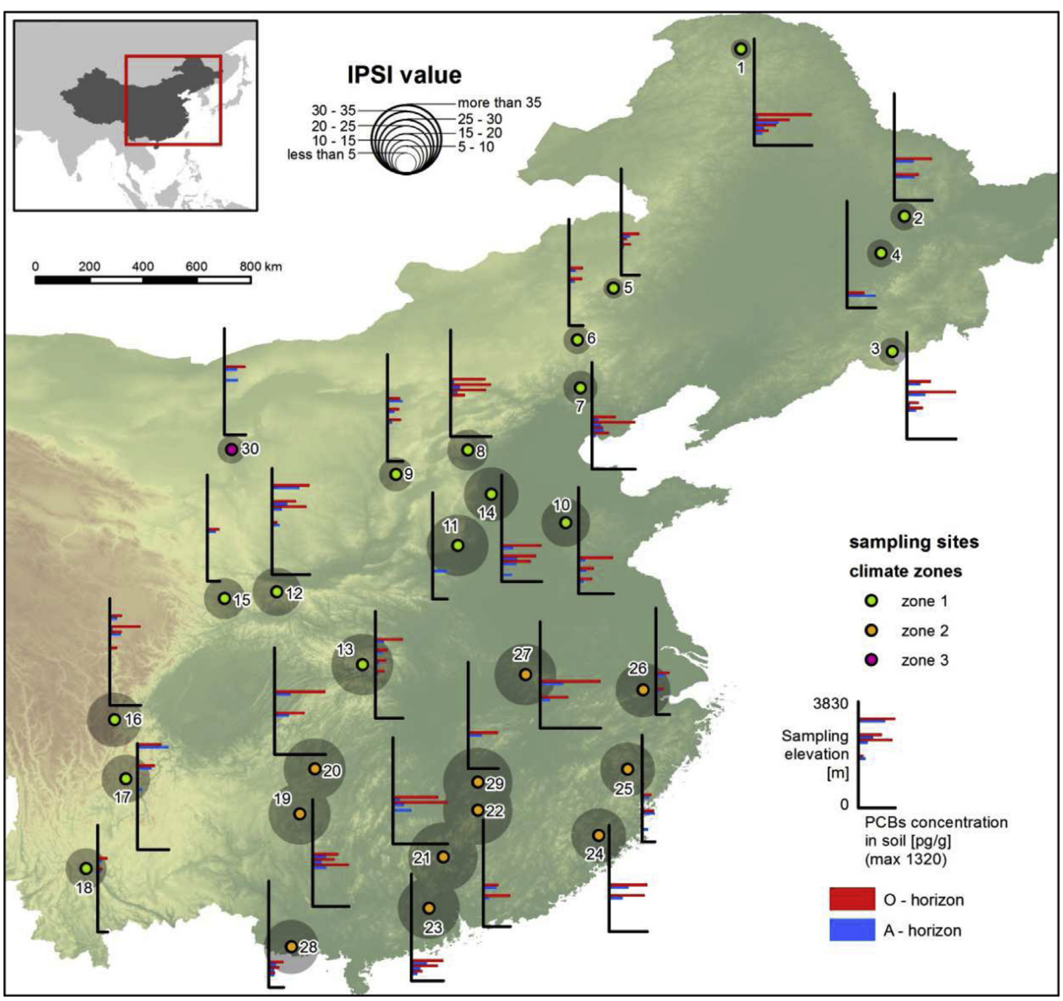

Fig. 1. Altitudinal and regional distribution of PCBs in organic (red bars) and mineral (blue bars) soil horizons, and the index of potential source influence (IPSI) (defined in Section 2.6). Different climate zones are represented by points with different colors: green: Climate zone 1: humid continental climate; orange: Climate zone 2: humid subtropical climate; purple: Climate zone 3: semiarid continental climate. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)