Environ Sci Pollut Res (2015) 22:1635–1643
DOI 10.1007/s11356-014-3404-8

Global trends of research on emerging contaminants in the environment and humans: a literature assimilation

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Received: 30 January 2014 / Accepted: 29 July 2014 / Published online: 7 August 2014

Abstract Available literature data on five typical groups of emerging contaminants (EMCs), i.e., chlorinated paraffins (CPs), dechlorane plus and related compounds (DPs), hexabromocyclododecanes (HBCDs), phthalate esters, and pyrethroids, accumulated between 2003 and 2013 were assimilated. Research efforts were categorized by environmental compartments and countries, so that global trends of research on EMCs and data gaps can be identified. The number of articles on the target EMCs ranged from 126 to 1,379 between 2003 and 2013. The numbers of articles on CPs, DPs, HBCDs, and pyrethroids largely followed the sequence of biota > sediment ≥ air > water ≥ soil > human tissue, whereas the sequence for phthalate esters was water > sediment > soil > human tissue ≥ biota ≥ air. Comprehensive studies on the target EMCs in biological samples and human tissues have been conducted worldwide. However, investigations into the occurrence of the target EMCs in soil of background areas and water are still scarce. Finally, developed and moderately developed countries, such as the USA, China, Canada, Japan, and Germany, were the main contributors to the global research efforts on EMCs, suggesting that economic prosperity may be one of the main factors propelling scientific research on EMCs.

Keywords Emerging contaminant · Published article · Environmental compartment · Economic prosperity

Responsible editor: Philippe Garrigues

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Introduction

Emerging contaminants (EMCs), i.e., those of emerging concern, have been defined by the United States Environmental Protection Agency (2013) as chemicals being detected at levels in one or some environmental compartments that previously had not been discovered. In recent years, increasing concerns have focused on EMCs, such as dechlorane plus (DP), hexabromocyclododecanes (HBCDs), phthalate esters, and pyrethroids in various environmental compartments. Most of these compounds were initially used as substitutes for known toxic chemicals and have been manufactured commercially at a rate of more than 1,000 tons annually (Covaci et al. 2011; Ren et al. 2008). For example, DP was commercialized as a flame retardant to replace mirex (Xian et al. 2011), which is a persistent organic pollutant (POP) and was banned in the 1970s. In addition, HBCDs were alternative halogenated flame retardants for polybrominated diphenyl ethers (PBDEs), which were classified as POPs in 2009. Therefore, EMCs have been increasingly detected in the environment.

Due to the lack of toxicological data for EMCs before use, scientists have been working towards better understanding of their environmental occurrences and potential health risks to wildlife and humans, and produced numerous publications. Presently, consensus has been reached mostly on the bioaccumulation potential of short-chain chlorinated paraffins (CPs) in organism and persistence in the environment (Zeng et al. 2011). The detection of HBCD in polar bears from Greenland

and Svalbard in the Arctic Ocean implicated the potential for long-range atmospheric transport of HBCD (De Wit et al. 2004). In addition, toxicity tests on the amphipod (*Hyalella azteca*) exposed to runoff samples identified pyrethroids, particularly bifenthrin and cyfluthrin, as the main sources of toxicity (Weston and Lydy 2010). These findings suggested that some EMCs, such as CPs, HBCDs, and constituents of pyrethroids, may become the candidates of POPs and therefore deserve additional attention in a long run.

However, consensus has not been reached by the Persistent Organic Pollutants Review Committee on the risk profiles of some EMCs, such as short-chain CPs (Wang et al. 2013) and HBCDs (Arnot et al. 2011). One of the main reasons for the disagreement is the scarcity of monitoring data in environmental media and human for short-chain CPs (Wang et al. 2013) and inconsistency in the development of risk profiles for HBCDs (Arnot et al. 2011). Consequently, understanding of current monitoring efforts on EMCs around the world is an important step towards establishing and implementing new programs for additional data acquisitions. Although existing reviews (Bayen et al. 2006; Covaci et al. 2011; Magdouli et al. 2013; Marvin et al. 2011; Palmquist et al. 2012; Xian et al. 2011) have summarized available data about the emission sources, environmental occurrence and global levels of EMCs, a map showing monitoring efforts has yet to be drawn but would be valuable for identifying data gaps.

The present review attempted to assimilate all available literature data on five typical groups of EMCs, i.e., CPs, DP and related compounds (DPs), HBCDs, phthalate esters, and pyrethroids, accumulated from 2003 to 2013 and analyze their occurrences air, water, soil, sediment, biota, and human tissues. A worldwide comparison of monitoring efforts on the target EMCs is presented. In addition, possible correlations between economic prosperity and research output are examined.

Methodology

Topics searches spanning all areas of environmental science and technology (including environmental science, ecology, toxicology, chemistry, and public environmental occupational health) from 2003 to 2013 were conducted with Web of Science. The search results were further refined by the language of English and document types of article and review. Articles on the target EMCs were classified by individual environmental compartments, i.e., air, water, soil, sediment, biota, and human tissue. It should be noted that the results associated with indoor dust and dry and wet deposition were considered as atmospheric research, whereas sewage sludge was regarded as sediment. In addition, these articles were also categorized by country, using the first authors' address as the sole criterion. Country classification with economic prosperity was based on the FTSE Global Equity Index Series update in September 2013 (http://www.ftse.com/Indices/Country_Classification/Downloads/September_2013_Country_Classification_Update.pdf). Countries in the categories of advanced emerging and secondary emerging on the Watch List were defined as being moderately developed in the present study. All the data were processed with Microsoft Excel and SigmaPlot 10.0.

Results and discussion

Emerging contaminants in the environment

Emerging contaminants have been detected in all environmental compartments across the globe. For instance, atmospheric short-chain CPs have been found at both urban and remote areas in East Asia (China, Japan, and South Korea) (Li et al. 2012b). HBCDs have been found in polar bears of East Greenland and Svalbard of the Arctic Ocean (McKinney et al. 2011) and alpine fish from the Tibetan Plateau of China (Zhu et al. 2013b). Overall, the numbers of articles on the target EMCs ranged from 126 to 1,379 for the period of 2003–2013 (Table 1), whereas those related to the environmental compartments under investigation accounted for more than 90 % of the total searched articles. With respect to environmental compartments, the percentages of articles for CPs, DPs, HBCDs, and pyrethroids largely follow the sequence of biota > sediment ≥ air > water ≥ soil > human tissue, whereas the sequence for phthalate esters is presented by water > sediment > soil > human tissue ≥ biota ≥ air (Fig. 1).

Biota

Specifically, biota, including plants and marine and terrestrial animals, was the most investigated medium for all target EMCs except for phthalate esters (Fig. 1), with the percentages of articles as 25 %, 43 %, 48 %, and 64 % for CPs, DPs, HBCDs, and pyrethroids, respectively, among all compartments. Plants were the least studied biological species for CPs, DPs, and HBCDs, with only six articles related to tree bark and spruce needle, examined by passive atmospheric samplers (Salamova and Hites 2010). On the other hand, fish were the most popular samples, accounting for 34 % of total biological samples for CPs, DPs, HBCDs, and phthalate esters. There have been 111 articles about these four target contaminants in fish across the globe. As the main protein sources for human being, freshwater and seawater fish, such as lake trout (*Salvelinus namaycush*), rainbow trout (*Oncorhynchus mykiss*), zebrafish (*Danio rerio*), European eel (*Anguilla anguilla*), Atlantic cod (*Gadus morhua*), and polar cod (*Boreogadus saida*), have been the preferred objects for

Table 1 Total numbers of research articles on the target emerging contaminants in biota, sediment, water, soil, air, and human tissues between 2003 and 2013

	CPs	DPs	HBCDs	Phthalate esters	Pyrethroids
Total	126	135	389	615	1,379
Biota	Fish, gull egg mollusks, spruce needle, chicken.	Shark, birds, fish, tree bark, mussel, whale. zooplankton.	Fish, birds, seals, polar bears, dolphins, sharks, maize, dog, tree bark, bivalves.	Fish, bivalves, plant.	Shrimp, bugs, fish, mosquitoes.
Sediment	Switzerland: Lake Thun; South Africa; North and Baltic Sea; Spain: Barcelona; Czech Republic; China: Bohai and Yellow Sea, Liaohe River, Pearl River Delta; Lake Ontario; Canada: Lake St. Clair.	Great Lakes; Lake Ontario; China: Yellow Sea, Northeastern Chinese river, Pearl River Delta, Yangtze River Delta; Niagara River.	Netherlands: Scheldt estuary; Great Lakes; English Lakes; Sweden; Canada: Swiss Lake and Lake Winnpeg; Korea; Spain: Cinca River; Ebro River Basin; Norway: Lake Ellasjoen, Indonesia: Surabaya; China: Taihu Lake and Laizhou Bay, Dongjiang River.	USA: Anacostia River, Hackensack River, Bibao and Urdaibai Estuaries; Nigeria: Ogun River; China: Taiwan, Qiantang River, Jianghan Plain, Pearl River, Yellow River and Yongding River.	China: Pearl River Delta; USA: California; Brazil: Pantanal Wetland; Ebro River Delta.
Water	Lake Ontario and Lake Michigan	North Sea; Atlantic and Southern Ocean; China: Taihu Lake, Northeastern Chinese River.	Great Lakes; Sweden; China: Taihu Lake.	USA: West Prong Little Pigeon River; South Africa; Thailand; Cantabrian Sea; Mexico: Xochimilco Wetland; Malaysia: Selangor; France: Seine River estuary, China: Yellow River, Yangtze River, Pearl River, Weihe River and Jianghan Plain.	USA: California, Illinois; Spain: Valencia; Ebro River Delta, South Africa: Western Cape; Central Poland; Pakistan: South Punjab; India: Hyderabad Hayana and Hisar; Europe; China: West and Taihu Lake.
Soil	China: Liaohe River, Pearl River Delta and Chongming Island; South Africa	China: e-waste region, Huai'an City; Pakistan.	Sweden; China: Laizhou Bay and Yangtze River Delta.	China: Yellow River Delta, Beijing, Guangzhou and Taizhou.	Slovak; Europe; Thailand; India: Haryana; China: Shenyang, Yangtze River Delta.
Air	China: Pearl River Delta; Japan, South Korea; United Kingdom.	China; Great Lake; Canada: Ottawa and Vancouver.	USA: Amarillo/Austin; UK: Birmingham; Romania; Canada: Toronto, Vancouver; Japan: Tokyo; Czech Republic; Belgium: Flanders; China: Shenzhen, Shanghai, Beijing.	USA; Italy: Palermo; China: Taizhou, Nanjing, Shenzhen, Hong Kong, Guangzhou and Tianjing; Greece: Thessaloniki; France: Pair.	USA: California; Thailand: Bangkok metropolitan region.
Human tissue	UK	China: Laizhou Bay, e-waste region; Canada: Kingston and Sherbrooke.	Vietnam (e-waste); the Philippines; USA: Boston; Northern China; Australia; UK; Sweden; Northern Russia; Northern and Southern Norway; South Africa; Japan; Czech Republic.	USA: North Carolina and New York; Poland: Krakow; France: Pairs; China: Chongqing.	USA: New York; Northern Thailand; South Africa; Northern Poland; Japan; Indonesia: West Java; India: New Delhi; France: Bobighy; China: Jiangshu Province, Canada: Quebec.

CPs chlorinated paraffins, *DPs* dechlorane plus and related compounds, *HBCDs* hexabromocyclododecanes

assessment of human health risk through dietary exposure. Furthermore, predatory fish and fish-eating predators, e.g., dolphins, whales, shark, sea lions, and polar bears, have also been widely used to examine the bioaccumulation and biotransformation of the target contaminants through foodweb transfer.

In addition, analyses of eggs of glaucous gulls (*Larus hyperboreus*) and peregrine falcon (*Falco peregrinus*), accounting for 43 % and 59 % of all birds studies, have been more widely used for evaluating the temporal trends of DPs and HBCDs than those from other types of birds, such as guillemot (*Uria aalge*) (Lundstedt-Enkel et al. 2005) and white-

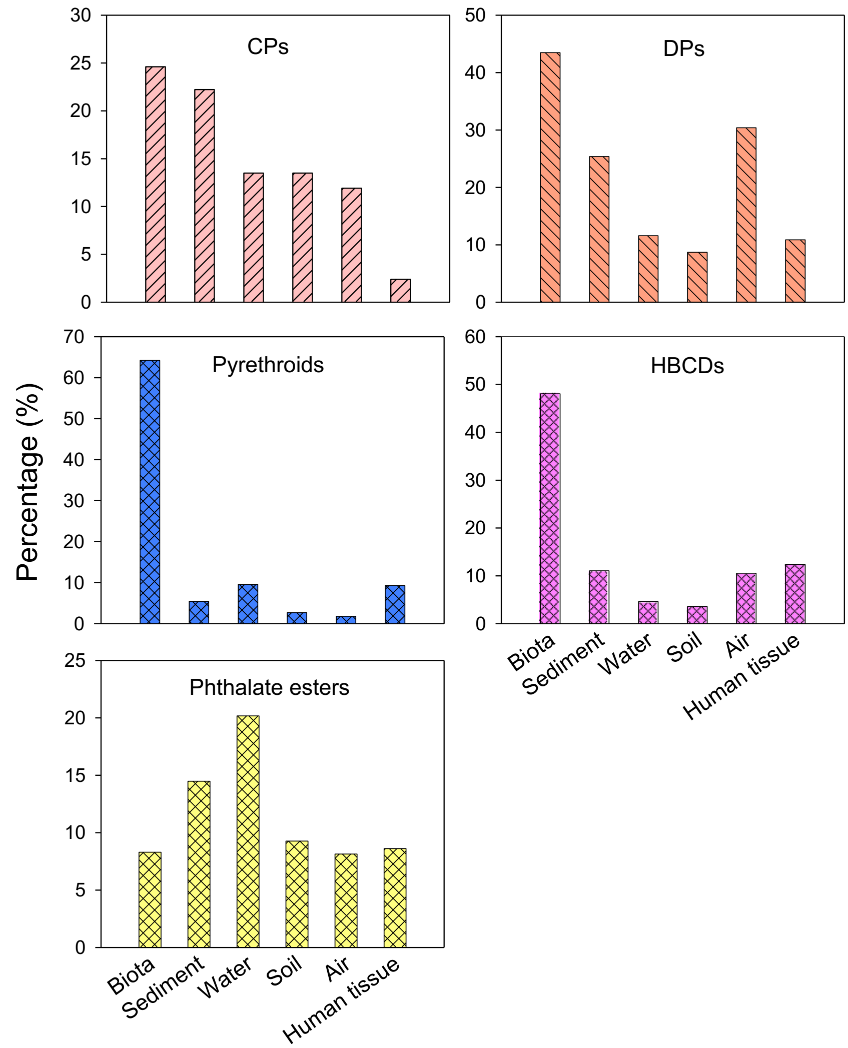

Fig. 1 Percentage of articles on the target emerging contaminants in biota, sediment, water, soil, air, and human tissues. Data were collected from Web of Science between 2003 and 2013. *CPs* chlorinated paraffins, *DPs* dechlorane plus and related compounds, *HBCDs* hexabromocyclododecanes

tailed eagle (*Haliaeetus albicilla*) (Nordlöf et al. 2010). However, few reports about pyrethroids in birds have been found in the present search. This may be due to their low detection frequency in birds, as they have short half-life (0.673–608 days) in water or soil and low bioconcentration factors (359–6,090) in prey (fish) (Ware 2002). Meanwhile, rat and chicken were also significant biological samples representative of terrestrial animals. Thirty-seven articles have been published on the toxicity and accumulation of CPs, DPs, and HBCDs in rats and chickens. To conclude, comprehensive studies on the occurrence of target EMCs in biota have been conducted worldwide; however, acute and chronic toxicity of CPs, DPs, and HBCDs in the benthic organism may merit more research in the future.

Sediment and soil

The number of articles on sediment was less than that on biota, but has gradually increased. There were 268 articles focusing on CPs, DPs, HBCDs, phthalate esters, and pyrethroids in surface sediment and sewage sludge from North and South America, Asia, and Europe (Table 1) between 2003 and 2013. It is interesting to note that most sampling sites for the target EMCs except for pyrethroids were located in China, such as the Pearl River Delta (Chen et al. 2011), the Yangtze River Delta (Zhu et al. 2013a), Yellow River (Sha et al. 2007), and Laizhou Bay (Li et al. 2012a). In these field studies, surface sediments were often collected for characterizing the levels and distributions of EMCs. However, only one sediment core, taken from Lake Ontario, has been investigated for the migration and time trends of DPs within the sediment column (Qin et al. 2007). On the other hand, there were 7 and 24 articles on HBCDs and phthalate esters, respectively, in sewage sludge from municipal treatment plants in Spain and China, while two articles were related to CPs and DPs on this issue.

Application of contaminated sewage sludge to agricultural or other lands may cause leaching of embedded EMCs into soil. However, only 137 articles on the target EMCs in soil have been published. Furthermore, 15 % of the articles were related to method development in laboratory. The remaining studies on CPs, DPs, and HBCDs in soil were conducted mainly around electronic waste recycling areas (Gao et al. 2011) or production facilities (Wang et al. 2010b), which are the point sources of the target EMCs. In addition, four articles reported phthalate esters in urban soil, and two articles described the occurrence of phthalate esters in agricultural and arable soils throughout China. Equally, only two articles assessed pyrethroids insecticide in soil from China, whereas three articles provided maps of soil contamination by pyrethroids in Slovak, India, and Thailand. Overall, investigations of the target contaminants in soil from background areas have been limited.

Water

Investigations of the occurrence of CPs and HBCDs in water have been limited to a few sites of lakes. For example, concentrations of CPs were determined only in Lake Ontario and Lake Michigan (Houde et al. 2008). Similarly, HBCDs have also been detected in lakes only, e.g., nine English Lakes (Harrad et al. 2009), Great Lakes of North America (Hites 2006), and Taihu Lake of China (Xu et al. 2013). On the other hand, DPs have been found in seawater of North Sea (Möller et al. 2012), the Atlantic and Southern Ocean (Xie et al. 2011), along the Atlantic transect from Arctic to Antarctica (Möller et al. 2010). Opposite to limited monitoring data of CPs, DPs, and HBCDs in lakes and oceans, numerous measurements of phthalate esters and pyrethroids in rivers, lakes, and estuaries have been conducted. There were 124 and 112 articles related to method development and determination of phthalate esters and pyrethroids in water, respectively, contributing to 20 % and 10 % of the total studies. Among these data, 11 articles presented the concentrations of phthalate esters in Chinese rivers or lakes, whereas 16 articles reported the occurrence of pyrethroids in storm water, river water, or seawater of California, one of the most developed agricultural regions in the USA (Spurlock and Lee 2008). In addition, 11 articles described the occurrences of pyrethroids in rivers, estuaries, and lakes of Spain, Poland, Pakistan, South Africa, India, and China. Overall, although field measurements of dissolved CPs are relatively scarce, the number of analytical protocols for determination of aqueous short-chain CPs in water has been on the rise (Geiß et al. 2011, 2012). However, there is no related report of DPs, HBCDs, phthalate esters, and pyrethroids in water.

Air and human exposure

The present literature search yields 172 articles on air concentrations of the target EMCs around the world. Phthalate esters were the most detected contaminants with 50 published articles. Specifically, 28 articles were related to the occurrence of all the target contaminants except for pyrethroids in ambient air of urban and remote areas, suggesting that CPs, DPs, HBCDs, and phthalates esters may have the potential of long-range atmospheric transport. In addition, two studies seemingly developed or utilized passive air samplers for monitoring atmospheric HBCDs and CPs, which may signal the beginning of long-term or large-scale air monitoring of EMCs. On the other hand, indoor air contamination by EMCs has been a source of increasing concerns. For example, 50 % of the articles on the occurrence of atmospheric HBCDs and phthalate esters were related to indoor air quality. For pyrethroids, its concentrations have been determined in indoor dust from homes located in California of the USA (Hwang et al. 2008; Quirós-Alcalá et al. 2011; Trunelle et al. 2013) and Bangkok metropolitan regions of Thailand (Pentamwa and Oanh 2008).

Intake of contaminated indoor air or dust is an important pathway of human exposure to EMCs. Levels of emerging contaminants in human tissue have been used to assess human health risk. There have been 245 articles related to all five target EMCs in human tissues between 2003 and 2013. Human tissues under investigation include breast milk, hair, urine, saliva, serum, plasma, blood, and adipose tissue. Different EMCs have been studied in various human tissues. For example, seven articles were related to DPs in serum from residents of Laizhou Bay of China (He et al. 2013) or e-waste recycling areas (Ben et al. 2013), whereas there were five articles on DPs in breast milk. In contrast, 54 % of the articles on HBCDs in human tissues focused on their levels or temporal trends in human milk. For phthalate esters and pyrethroids, urine was the most widely used human tissue; 13 and 18 articles have been published on the urinary levels of the parent compounds and their metabolites. Nevertheless, only one paper was found, describing short- and median-chain CPs in 25 human milk fat samples from urban London and rural Lancaster areas of the UK (Thomas et al. 2006). Overall, the number of studies on CPs, DPs, HBCDs, phthalate esters, and pyrethroids in human tissues has been on a gradually increasing trend in recent years.

Worldwide distribution of research on emerging contaminants

To assess the regional research trends, world distribution of articles by country was assessed. The countries, responsible for 58–92 % of the total articles for all target EMCs, are mainly located in Asia, North America, and Europe (Fig. 2). Specifically, China's research groups have contributed the most to the studies on DPs and phthalate esters (Fig. 2). In particular, the number of articles on DPs produced in China amounted to 43 % of the total. It is interesting to note that the number of articles on DPs published annually in China increased from 1 in 2008 to 24 in 2013, after Wang et al. (2010a) pointed out that there were few data of DPs reported from China before 2010. On the other hand, there were 261 articles

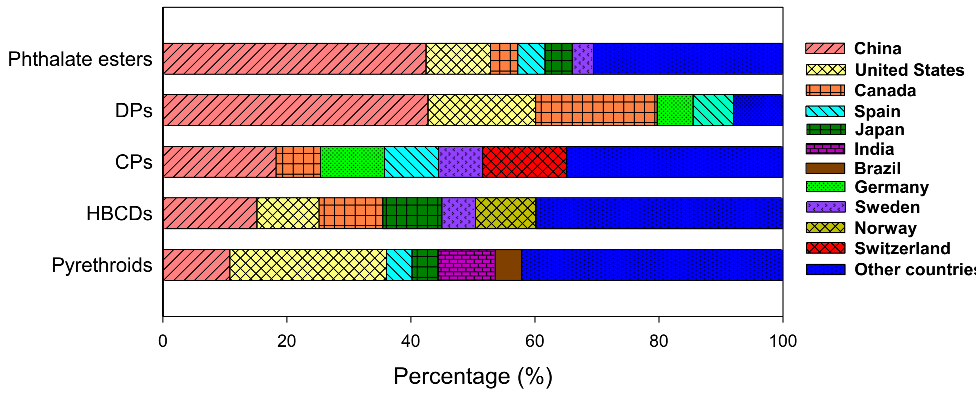

Fig. 2 Global distribution of articles on the target emerging contaminants. Data were collected from Web of Science between 2003 and 2013 and the first authors' addresses are used to assign countries. *CPs* chlorinated paraffins, *DPs* dechlorane plus and related compounds, *HBCDs* hexabromocyclododecanes

on phthalate esters from China, accounting for 42 % of the total studies. This may be related to the large amount of plastic products manufactured in China, e.g., 5.6 million tons in 2013 (National Bureau of Statistics of China 2013), which has caused alarming concerns about white pollution. Similarly, 348 articles on pyrethroids in various environmental compartments were produced from the USA, with 10 % focused on the toxicity of pyrethroids to benthic organisms. In addition, articles on CPs or HBCDs, alternative halogenated flame retardant for PBDEs, were mainly contributed from China and Switzerland or Canada, respectively.

The global distribution of research on the target EMCs by country, using biota as a representative compartment, is displayed in Fig. 3. Apparently, China and the USA have

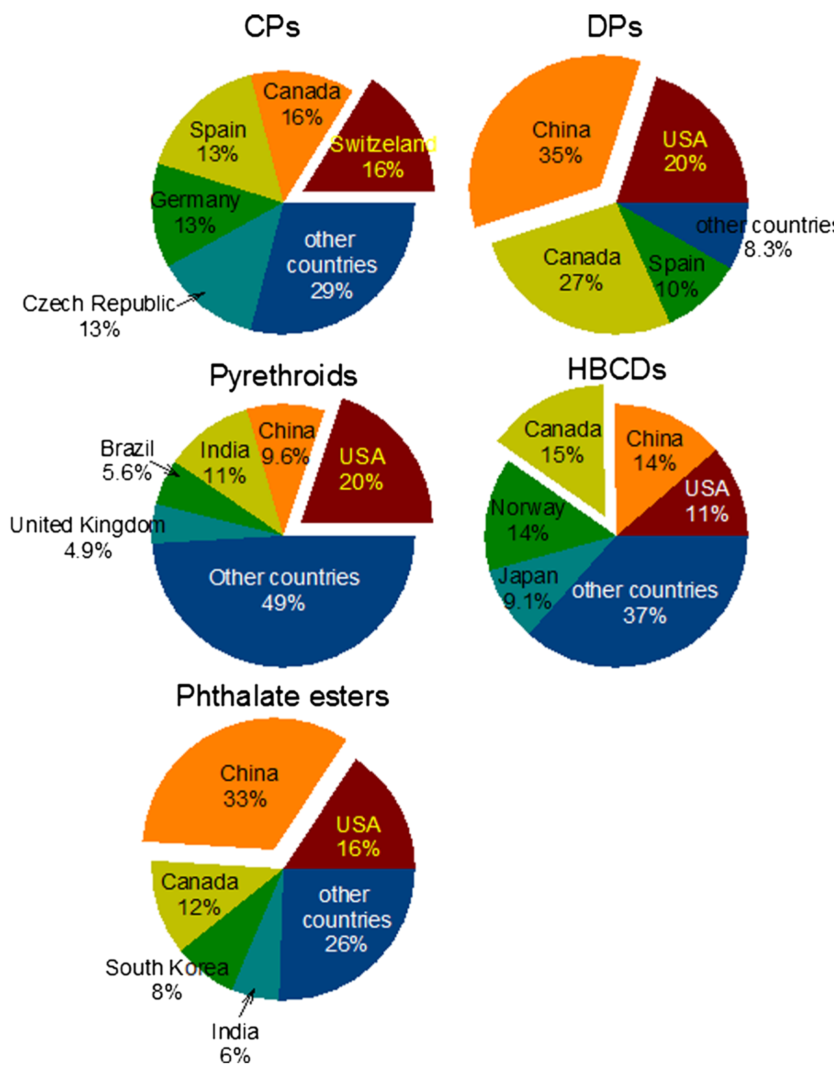

Fig. 3 Global distribution of articles on the target emerging contaminants in biota. Data were collected from Web of Science between 2003 and 2013 and the first authors' addresses are used to assign countries. *CPs* chlorinated paraffins, *DPs* dechlorane plus and related compounds, *HBCDs* hexabromocyclododecanes

played a major role in the studies of DPs, phthalate esters, and pyrethroids. Furthermore, Canada published 15–16 % of the articles on HBCDs and CPs. On the other hand, the UK, South Korea, and Czech Republic, which were responsible for less than 3 % of the total articles (Fig. 2), contributed to 4.9 %, 8 %, and 13 % of the articles on pyrethroids, phthalate esters, and CPs in biota, respectively (Fig. 3). Specifically, research groups in Canada, China, and Switzerland have focused on monitoring the occurrence of CPs and DPs in biota samples, contributing to more than 80 % of the total articles. In addition, 27 % of the articles on HBCDs in biota published by China were concentrated in bioaccumulation of HBCDs in zebrafish (*D. rerio*) and its embryo toxicity tests. Similarly, researchers in the USA have paid more attention to toxicity tests of pyrethroids in aquatic environment, which accounted for approximately 21 % of the articles related to toxicity analysis. These results indicate that specific research groups in different countries may have been interested in a specific group of target contaminants in a certain environmental compartment, based on their expertise areas and/or countries' main sources of contamination. To sum up, the main contributors to the studies of CPs, DPs, HBCDs, phthalate esters, and pyrethroids were largely research groups from developed and moderately developed countries, such as the USA, Canada, and China. More efforts should be directed towards widespread surveys of EMCs in undeveloped regions so that ecological and human health risk assessments can be conducted globally.

Correlation between economic prosperity and research efforts

Economic prosperity may be one of the key factors dictating the distribution of research efforts discussed above. To test this hypothesis, gross domestic product (GDP), an integrated national economic index, in 12 countries for the period of 2003–2013 (International Monetary Fund 2014) was correlated with the numbers of articles produced (Fig. 4a). As shown, the USA with the highest GDP contributed much more articles than all other countries except for China. Although the total GDP in China was less than that for the USA, the number of articles on the target EMCs from China was slightly more than that from the USA. This is reflective of China's increasingly important role in the global scientific community, particularly in leading the efforts to deal with EMCs. One of the main driving forces may be ascribed to the fact that China has produced and used the largest amounts of these target EMCs in the world (Fiedler 2010; IHS Chemical 2013; Li et al. 2014; Xian et al. 2011). Figure 4b shows that the number of articles on the target EMCs produced annually in China increased proportionally with increasing GDP from 2003 to 2013. This good correlation may partly be resulted from China's rising

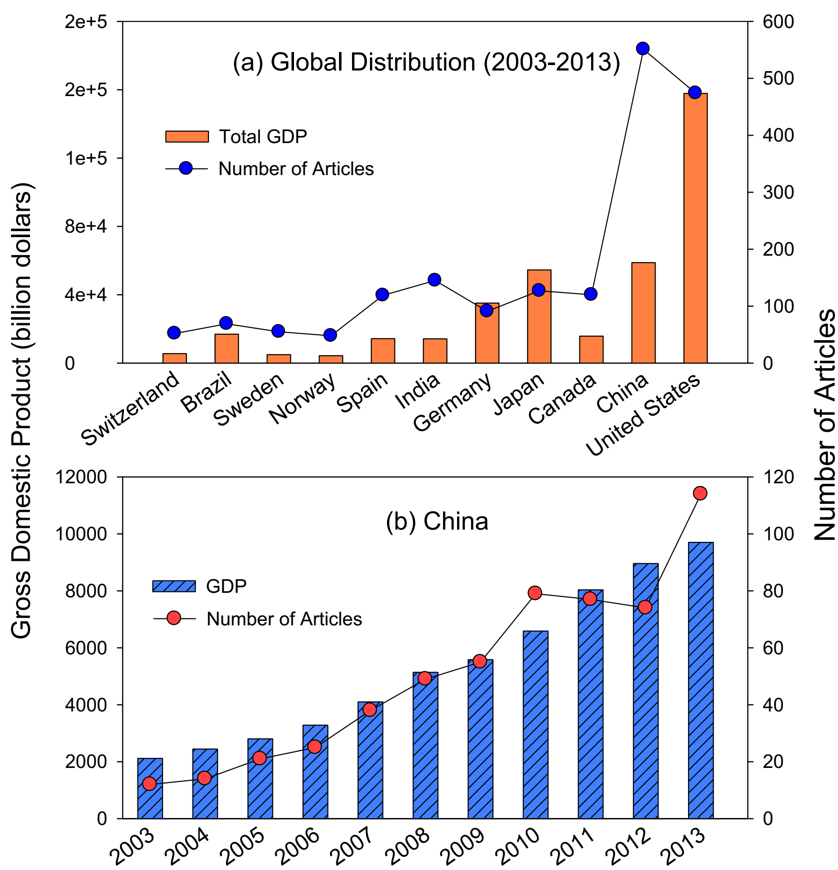

Fig. 4 Global trends of **a** correlation between gross domestic product (GDP) and the number of articles on the target emerging contaminants (chlorinated paraffins, dechlorane plus and related compounds, hexabromocyclododecanes, phthalate esters, and pyrethroids) for the time period of 2003–2013 and **b** annual GDP and the number of articles in China from 2003 to 2013. Data were collected from Web of Science and the first authors' addresses are used to assign countries. GDP data were acquired from World Economic Outlook Database (update on January 2014) of International Monetary Fund

spending on scientific research, which carried an average annual growth rate of 16 % during the same period of time (National Bureau of Statistics of China 2013). Moreover, approximately equal numbers of articles were produced from Canada (120) and Japan (127), although Japan's GDP was more than three times than that of Canada. One explanation for the large research output from Canada than Japan may be its unique geographic advantage, e.g., close to the Great Lakes and Arctic regions, where there were 28 articles related to EMCs in environmental samples. Clearly, economic prosperity is highly related to the research efforts on EMCs, with some influences from individual nations' priority on specific groups of contaminants and/or sampling locations. Overall, the environmental occurrence of EMCs has become a global concern, with the USA, China, Canada, Japan, and Germany seemingly leading the monitoring efforts.

Conclusions

From the above analyses of the global trends of research on the target EMCs, we have arrived at several observations/recommendations. First, comprehensive studies on the target EMCs in biological and human tissue samples have been conducted worldwide, whereas there have been limited investigations on soil of background areas, which should be enhanced in future monitoring. Second, international standard methods for determination of DPs, HBCDs, phthalates esters, and pyrethroids are desirable for water quality assessment. On the other hand, improved passive air sampling techniques may provide the opportunity for monitoring of atmospheric EMCs across the globe. Finally, the studies of the target EMCs have been largely conducted by research groups from developed and moderately developed countries, such as the USA, Canada, and China. Apparently, economic prosperity has been the driving force for research efforts on EMCs.

Acknowledgments This work was financially supported by the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences (GIGCAS 135 project Y234081001) and the Natural National Science Foundation of China (Nos. 41121063 and 41329002). This is contribution No. IS-1938 from GIGCAS.

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