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Dechlorane Plus as an emerging environmental pollutant in Asia: a review

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Abstract

Dechlorane Plus (DP) is an unregulated, highly chlorinated flame retardant. It has been manufactured from past 40 years but its presence in the environment was initially reported in 2006. Later, it has been found in various biotic and abiotic environmental matrices. However, little attention has been paid to monitor its presence in Asia. Many studies have reported the occurrence of DP in the environment of Asia, yet the data are scarce, and studies are limited to few regions. The objective of present review is to summarize the occurrence, distribution, and toxicity of this ubiquitous pollutant in various environmental matrices (biotic and abiotic). DP has also been reported in the areas with no emission sources, which proves its long-range transport. Moreover, urbanization and industrialization also affect the distribution of DP, i.e., high levels of DP have been found in urban areas relative to the rural. Tidal movement also incorporates in transport of DP across the aquatic system. Further, bioaccumulation trend of DP in various tissues is kidney > liver > muscle tissues, whereas, blood brain barrier resists its accumulation in brain tissues. Additionally, gender-based accumulation trends revealed high DP levels in females in comparison to males due to strong metabolism of males. Furthermore, methodological aspects and instrumental analysis used in previous studies have also been summarized here. However, data on biomagnification in aquatic ecosystem and bioaccumulation of DP in terrestrial food web are still scarce. Toxicity behavior of *syn*-DP and *anti*-DP is still unknown which might gain the interest for future studies.

Keywords Dechlorane Plus · Flame retardant · Emerging pollutant · Environmental occurrence · Bioaccumulation · Toxicity

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Introduction

Dechlorane Plus (CAS No. 13560-89-9) is an unregulated highly chlorinated flame retardant which was initially introduced in the market in 1960s as a replacement of mirex (Ren et al. 2018; Vorkamp et al. 2015). Its use raised in 2007 after the regulation of mirex and poly-brominated-diphenyl ethers (PBDEs) in Stockholm Convention (Talsness 2008; Betts et al. 2006; Law et al. 2006; De Wit 2002). European Commission has enlisted Dechlorane Plus (DP) as a possible alternative for 27 compounds used in electronic applications (Pakalin et al. 2007). It is preferably used owing to its less cost, low density, and high thermal and photochemical stabilities than the other brominated flame retardants (Feo et al. 2012; Tomy et al. 2007). DP is produced by the Diels–Alder condensation reaction of hexa-chloro-cyclo-pentadiene with 1, 5-cyclo octadiene in a molar ratio of 2:1 (Betts et al. 2006). Its commercial formulation consists of a mixture of two stereoisomers: *syn*-DP and *anti*-DP with the ratio of 1:3 (Zhu et al. 2007). However, dominance of *syn*-DP has been reported in environmental matrices which can be attributed to the enhanced photodegradation stability of *syn*-DP isomer or *anti*-DP to *syn*-DP isomerization during air transport (Iqbal et al. 2017; Wang et al. 2016). DP has numerous applications like in industrial polymers, plastic, resins, and nylons. As a flame retardant, it is also used for the coating of wires, cables, computer connectors, and plastic roofing materials, etc. (Qiu et al. 2007). Physio-chemical properties of DP have been described in Table 1.

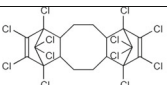
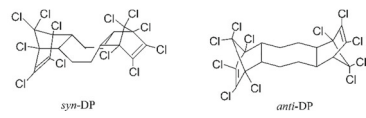
DP has been classified as a high production volume chemical by United State Environmental Protection Agency (Hansen et al. 2020) because the estimated annual production of DP is more than 450 tons (Feo et al. 2012; Howard and Muir 2010). However, China produces 300 tons of DP per year (Ren et al. 2018). Three types of DP products are available commercially (DP-25, DP-35, and DP-515). All of them have same chemical composition but differ in particle sizes (Zhu et al. 2007). DP is used worldwide; therefore, it is ubiquitous in the environment (Sverko et al. 2011). DP has been used since decades still the research on DP as a pollutant is scarce. It was first reported in the environment of Great Lakes in 2006 (Hansen et al. 2020; Hoh et al. 2006). After that, different studies have reported the existence of this global contaminant in various biotic and abiotic matrices across the globe (Ren et al. 2018) including North America (Su et al. 2015), Europe (Rjabova et al. 2016), and Asia (Ren et al. 2018; Kakimoto et al. 2015; et al. 2008). This proves its long-range transport. According to Hansen et al. (2020), the estimated DP emission in a low and high emission scenario is 0.02 and 3.2 tons per year, respectively. Furthermore, its toxicity (by dichlorination metabolism) and bioaccumulation in human body have also been presented in literature (Kim et al. 2016; Chen et al. 2015; Siddique et al. 2012). DP has also been found in some tree barks of China and Korea (Möller et al. 2010). The effects and occurrence of other flame retardants (e.g., PBDE) and similar compounds are well identified (Frederiksen et al. 2009; Costa et al. 2008; Vonderheide et al. 2008). However, limited literature is available on the environmental impacts of DP (Hoh et al. 2006). Most of the studies have been conducted in China as China is the largest producer, consumer, and exporter of flame retardants. Since 2003, Jiangsu Anpon Electrochemical Company in China is producing 454 metric tons per year (Ji et al. 2018). The available literature (Ren et al. 2018; Kakimoto et al. 2015; Möller et al. 2010; Ren et al. 2008) supports that DP is present in environment and spreading in Asia. So, there is an immense need of research in this continent to understand the occurrence, distribution, and effects of this pollutant (Ma et al. 2011).

The objective of current review is to bring attention towards the occurrence and distribution patterns of Dechlorane Plus as an emerging pollutant in Asia. Furthermore, it also aims to highlight the toxicological impacts of this flame retardant in different environmental matrices (abiotic and biotic) and discuss its movement across the different matrices involving various pathways and processes specifically in Asia.

Methodology

Data presented herein are based on literature mining of 100 documents. Relevant literature was accessed via

Table 1 Physio-chemical properties of DP

Empirical formula	C ₁₈ H ₁₂ Cl ₁₂
Chemical Structure	
Stereoisomers	
Color	White
Odor	Odorless
Thermal stability	285°C
Molar mass	653.7 g/mol
Melting point	350°C
Solubility	soluble in o-dichloro benzene very little aqueous solubility (0.044 µg L ⁻¹ to 249 µg L ⁻¹) little solubility in non-polar organic solvents like hexane (0.1%) and benzene (2%)
K _{ow} *	9.3
Degradation	Degradable in sewage sludge under aerobic conditions only.

* Octanol-water coefficient. (Oxychem, 2011)

* Octanol-water coefficient. (Oxychem 2011)

googlescholar.com, sciencedirect.com and scopus.com. This database was constructed on scopus (www.scopus.com) and kept updated by means of abstracts and current contents. Resultant data was screened using following criteria: (i) scientific documents addressing nature, properties, and production of DP were selected; (ii) data on the toxicity of DP was taken using information from the open literature; (iii) literature reporting the occurrence of DP in different matrices such as air, water, soil, sediments, crops, humans, birds, terrestrial, and aquatic biota was chosen; (iv) information regarding isomeric accumulation of *syn*-DP and *anti*-DP was favored; and (v) articles reporting DP studies conducted particularly in Asian countries were collated.

A map was constructed (see Fig. 1) which illustrates the areas where reported studies have been conducted. It is apparent from Fig. 1 that DP studies have been conducted in 13 countries of Asia and most of them are reported from South China. Furthermore, correlation was calculated using IBM SPSS 20 while multivariate statistical package MVSP was used for cluster analysis using Euclidian distance coefficient. Clustering method was nearest neighbor, and data were transformed using log 10 due to sparse nature of data. Results were obtained in the form of text dendrogram as illustrated in Fig. 2.

Occurrence and distribution of DP in various environmental matrices

Occurrence of DP in both biotic and abiotic environmental matrices has been confirmed by literature (Kakimoto et al. 2015; Möller et al. 2010). Literature reports that *anti*-DP degrades rapidly than *syn*-DP, whereas some studies says that *anti*-DP is more persistent in the environment. However, on the account of literature survey given in Table 2, it can be concluded that *anti*-DP is more persistent in environmental matrices. Li et al. (2018) compared the level of DP in various environmental matrices over the course of 10 years (2004 and 2014). They reported the elevated levels of DP in all samples. Furthermore, *anti*-DP in 70–80% samples was consistent.

Abiotic matrices

Literature reported the presence of DP in abiotic environmental matrices. DP has been found in the dust collected from house floor, AC filter, and cars in Jeddah, Saudi Arabia, with 5.5, 7.5, and 11 ng g⁻¹ of DP, respectively (Ali et al. 2016b). DP has also been reported in wetland plants and marine environment of Singapore (Wang and Kelly 2017; Zhang et al. 2015). Literature on the presence of DP in various abiotic environmental matrices of Asia has been summarized in Table 2.

Air

Air is one of the most often studied matrices. DP has been detected in the air of China (Yang et al. 2012), South Korea (Baek et al. 2013), Japan (Kakimoto et al. 2014), and Pakistan (Syed et al. 2013). A study conducted in Qingyuan, South China, reported that concentrations of DP in e-waste recycling site (13.1 to 1794 pg m⁻³) were two orders of magnitude higher than the reference site (0.47 to 35.7 pg m⁻³), due to the extensive use of DP in electrical applications (Chen et al. 2011). Another study described the air transport of DP through four districts of Shanghai, China. Sampling areas were 400 km away from DP manufacturing plant. The mean concentration of total particulate DP was found between 2.32 and 5.48 pg m⁻³ (Yu et al. 2011). However, Wang et al. (2010a) has reported the highest atmospheric DP concentration so far which were found to be 7737 to 26,734 pg m⁻³ near DP production plant in Jiangsu province, China. In Dalian, Northeastern city of China, the annual arithmetic mean concentration of DP in atmosphere was reported to be 3 ± 6 pg m⁻³ (Yang et al. 2012). This study also reported that the mean fraction of *syn*-isomer was 0.31 ± 0.41 ng m⁻³, which is very low than the values reported for technical mixture of DP ($f_{syn} = 0.4$) produced in China, indicating the depletion of *syn*-DP as compared to *anti*-DP due to some seasonal variations (Yang et al. 2012). Liu et al. (2018) reported the presence of DP in air of the eastern slope of Mt. Gongga on the eastern Tibetan Plateau. Levels of *syn*-DP and *anti*-DP ranged from BDL – 7.08 to BDL– 4.74 pg m⁻³, respectively. However, 11.5 pg m⁻³ DP was detected in air samples.

In Jeju, South Korea, there was no DP production source, but DP was present in different sites ranging from 9.87 to 44.8 pg day⁻¹ PAS⁻¹ (passive air sampler). The reason is that DP has been sold there for 40 years (Baek et al. 2013). This study exhibits the persistent nature of DP. One of the biggest cities of Japan, Osaka, showed total DP concentrations ranging from 7.1 to 15.4 pg m⁻³. The mean concentration was 11 pg m⁻³, which is less than DP level in Chinese urban sites (Kakimoto et al. 2014). Another study examined the atmospheric levels of DP in Northeast Asia (Japan, Korea, and China). DP was detected in all samples, and the mean total level of DP was reported between 6.7 and 0.87 pg m⁻³. Sources of DP in Japan included the use of products containing DP (Kakimoto et al. 2015). Another study reported the presence of DP in home dust samples collected from Kopawa village, Nepal. *Syn*-DP and *anti*-DP constitute 52% and 71% of total dust samples, respectively (Yadav et al. 2019).

In Pakistan, the concentrations of DP have been examined in agricultural, urban, and industrial areas along River Ravi, Punjab Province of Pakistan. As Ravi is one of the most highly polluted rivers, therefore, DP concentrations in various sites have been found within the range 1.5 to 530 pg m⁻³ with a mean value of 88 pg m⁻³ (Syed et al. 2013). Furthermore, Mahmood et al. (2015) reported that the presence of DP concentrations in the



Fig. 1 Map of Asia showing areas where studies have been conducted

air of Sialkot and Gujranwala, Pakistan, is lower than the concentrations recorded in the other areas of Pakistan and China (Syed et al. 2013; Ren et al. 2008). This can be attributed to the mixed nature of air samples which were from rural, urban, and peri-urban areas, while in previous studies, only urban areas were focused (Mahmood et al. 2015a). Similarly, the measured DP concentration in the air near e-waste recycling plant in Karachi ranged between 15.0 and 85.0 ng m^{-3} (Iqbal et al. 2017). Khan et al. (2016) compared the DP levels in indoor and outdoor dust of various areas of Pakistan. They reported that highest concentration of DP (0.02–63.1 ng g^{-1}) was found in industrial zone of Gujrat, Gujranwala, and Faisalabad, followed by rural zones of these cities (BDL–1.6 ng g^{-1}). However, 0.25–6.3 ng g^{-1} DP has been reported in dust of background areas (Kashmir, Swat, and Chitral).

Soil

Distribution of DP in soils of China was first time reported by Wang et al. (2010b). There are few DP manufacturing plants which are in Huai'an, Jiangsu Province, China, and DP (mean

5.1 ng g^{-1} dw) has been found in 21 soil samples collected from there. A concentration gradient was observed using contour map, and a notable decline in DP levels was observed while moving away from the plant. Presence of DP in the soil of South and Northeastern China has been reported in literature (Ma et al. 2011; Yu et al. 2010). Almost equal concentration (11.3 \pm 1.2 pg g^{-1} dw) of DP has been recorded in the soils of residential, business, and industrial settings (Ma et al. 2011). Another study has reported the gradient-based distribution of DP where concentration of DP decreased while moving away from an e-waste recycling plant. Additionally, high concentration of DP was detected in the soil of e-waste recycling site as compared to the industrial areas (Yu et al. 2010). Liu et al. (2018) reported the presence of DP in soil samples taken from the eastern slope of Mt. Gongga on the eastern Tibetan Plateau. Levels of *syn*-DP and *anti*-DP were found to be ranged from BDL – 34.8 to BDL – 19.0 pg g^{-1} dw, respectively. However, 48.3 pg g^{-1} dw DP was detected in soil samples.

Syed et al. (2013) reported that soils of Punjab, Pakistan, has also been contaminated with DP but the recorded levels

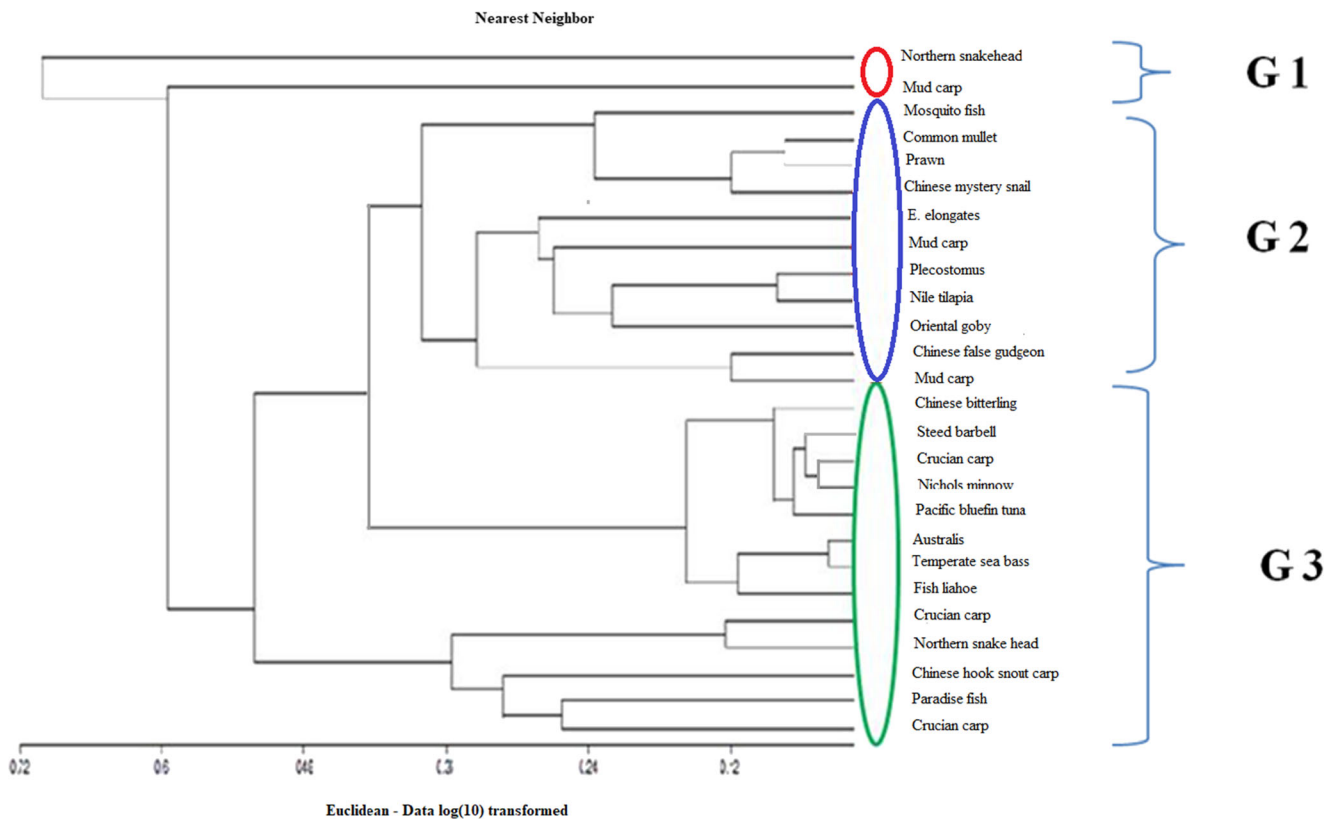


Fig. 2 Dendrogram representing specie clustering with respect to accumulation trend of *syn*-DP and *anti*-DP

were lower ($0.8 \pm 2.1 \text{ ng g}^{-1} \text{ dw}$) when compared with the soils of South China (Yu et al. 2010). This can be explained as in China, DP emission source e-waste recycling site, however, no such source was present around the sampling area of Pakistan (Syed et al. 2013). Moreover, Iqbal et al. (2017) reported the presence of DP (BDL to $26,239 \text{ ng g}^{-1}$) in soil of Karachi near e-waste recycling company. Furthermore, Mahmood et al. (2015) has reported the distribution pattern of DP as agricultural and rural areas < industrial and peri-urban areas < industrial and urban areas.

Water

There are very few studies which support the presence of DP in water (Mahmood et al. 2015). As DP is hydrophobic, its reported concentrations in water are negligible. A study reported the presence DP in both dissolved and suspended form in water of Dongjiang River catchment, Pearl River Delta, South China (a highly industrialized area). The concentration of dissolved DP ranged between 1.2 and 3.3 pg L^{-1} , whereas, particulate DP has been reported in the range of 0.24 to 0.78 ng L^{-1} (He et al. 2014a). Furthermore, DP has also been found in Daling River and Harbin, China, with the mean value of 0.40 ng L^{-1} and 0.2 pg L^{-1} , respectively (Wang et al. 2012; Ma et al. 2011).

Mahmood et al. (2015a) has examined DP pollution in two tributaries of Chenab river, Pakistan, and reported that the concentration of DP ($1.03 \pm 1.48 \text{ ng L}^{-1}$) were higher as compared to the previously reported levels in various rivers of China (Ma et al. 2011; Qi et al. 2010).

Sediments

Several studies have stated the presence of DP in water sediments (Fang et al. 2014; Syed et al. 2013; Sun et al. 2012; Wang et al. 2012). A study conducted in Songhua River, China, reported the elevated levels of DP in sediments collected from downstream points (242 and $115 \text{ pg g}^{-1} \text{ dw}$) as compared to the upstream ($14 \text{ pg g}^{-1} \text{ dw}$) due to the addition of industrial effluents and wastewater from wastewater treatment plant (Ma et al. 2011). Moreover, spatial distribution and vertical profile of DP have also been reported in a study conducted in Pearl River Delta, South China. Results revealed the source-based increase in concentrations that ranged from 0.08 to $19.4 \text{ ng g}^{-1} \text{ dw}$ with a mean value of $2.9 \text{ ng g}^{-1} \text{ dw}$. The reason for such high concentration ($19.4 \text{ ng g}^{-1} \text{ dw}$) was the presence of electronic manufacturing plant nearby while the lowest concentration was detected in the areas far away from industrial zone. Further, vertical profile analysis of DP revealed the elevated level of this pollutant in upstream section as compared to the lower. This was attributed to the recent

Table 2 Levels of DP and its isomers in various abiotic matrices

Sample matrix	Sampling location	<i>syn</i> - DP	<i>anti</i> -DP	Σ DP	Reference
Air (pg m ⁻³)	North Eastern China	100	600	700	Wang et al. 2012
	Qingyuan, South China	–	–	903.55	Chen et al. 2011
	Jiading District, China	1.58	3.90	5.48	Yu et al. 2011
	Zhabei District, China	1.31	3.77	5.08	
	Huangpu District, China	0.651	1.67	2.321	
	Pudong District, China	1.30	2.77	4.07	
	Dalian, Northeastern China	–	–	3	Yang et al. 2012
	Bihar, India	4.42	2.06	6.46	Yadav et al. 2020
	Kathmandu, Nepal	2.17	1.69	3.86	Yadav et al. 2017
	Pokhara, Nepal	1.29	0.81	2.10	
	Birgunj, Nepal	2.36	2.36	4.72	
	Biratnagar, Nepal	1.48	0.93	2.41	
	Beijing, China	0.41	0.83	1.24	Kakimoto et al. 2015
	Busan, Korea	0.26	0.62	0.88	
	Sapporo, Japan	1.49	4.36	5.85	
	Sagamihara, Japan	0.47	1.04	1.51	
	Kanazawa, Japan	0.73	2.26	3	
	Kitakyushu, Japan	1.80	2.68	4.48	
	Eastern Tibetan Plateau	2.07	1.98	4.05	Liu et al. 2018
	Osaka, Japan	2.92	8.2	11.12	Kakimoto et al. 2014
Punjab, Pakistan	75	13	88	Syed et al. 2013	
Karachi, Pakistan	41,000	8500	49,500	Iqbal et al. 2017	
Air (pg day ⁻¹ PAS ⁻¹)	Khovd, Mongolia	0.3	0.7	1	Baek et al. 2013
	Ulaanbaatar, Mongolia	0.19	0.36	0.55	
	Beijing, China	0.28	0.69	0.97	
	Yanji, China	0.16	0.39	0.5	
	Seoul, South Korea	5.30	39.5	44.8	
	Busan, South Korea	1.79	8.08	9.87	
	Pohang, South Korea	1.30	8.68	9.98	
	Jeju, South Korea	2.45	11	13.45	
Soil (ng g ⁻¹ dw)	South china, Guiyu	0.51	1.7	2.21	Yu et al. 2010
	Harbin City, Northeastern China	1.55	2.68	4.23	Ma et al. 2011
	Huai'an city, China	140.32	455.0	595.32	Wang et al. 2010b
	e-waste facility, Bui Dau, Vietnam	17	48	65	Someya et al. 2016
	Open burning place, Bui Dau, Vietnam	12	40	52	
	Paddy fields, Bui Dau, Vietnam	0.33	1.1	1.43	
	Pakistan, Punjab	0.6	0.2	0.8	Syed et al. 2013
	Karachi, Pakistan	10,697	21,620	32,317	Iqbal et al. 2017
Water (pg L ⁻¹)	North Eastern China, Daling River	120	290	410	Wang et al. 2012
	Southeastern China, Pearl Delta River	4.3	2.2	6.5	He et al. 2014a
	Punjab, Pakistan	730	300	103	Mahmood et al. 2015
	Jurong Island, Singapore	2.4	1.8	4.2	Zhang et al. 2015
	East Coast, Singapore	2.4	8.6	11	
	Pasir Ris, Singapore	2.3	1.3	3.3	
Sediments (ng g ⁻¹ dw)	Pearl Delta River, Southern China,	1.55	4.55	6.1	He et al. 2014a
	Daling River, North Eastern China	0.35	0.96	1.31	Wang et al. 2012
	Huai'an City, China	140.19	455.64	595.83	Wang et al. 2010b
	Qiantang River, China	–	–	0.29	Sun et al. 2013

Table 2 (continued)

Sample matrix	Sampling location	<i>syn</i> - DP	<i>anti</i> -DP	Σ DP	Reference
	Yellow Sea North China	0.0237	0.111	0.135	Zhao et al. 2011
	Hainan Island, China	–	–	1.064	(Qiu et al. 2019)
	River near e-waste facility, Bui Dau, Vietnam	1.8	4.8	6.6	Someya et al. 2016
	Pohang, Korea	–	–	0.027	Fang et al. 2014
	Jurong Island, Singapore	0.005	0.01	0.015	Zhang et al. 2015
	East Coast, Singapore	0.006	0.004	0.010	
	Pasir Ris, Singapore	0.011	0.023	0.034	
	Busan, Korea	–	–	0.072	Fang et al. 2014
	Punjab, Pakistan	1.5	0.4	1.9	Syed et al. 2013
	Mehmood Booti Drain, Lahore, Pakistan	–	–	0.02	Ali et al. 2016a
	Punjab, Pakistan	1.31	0.55	1.86	Mahmood et al. 2015

PAS, passive air sampler, *dw*, dry weight

incorporation of DP in the sediments (He et al. 2014a). DP has also been reported in the mangrove sediments of Hainan Island, China, with concentration ranged from 86 to 3522 pg g^{-1} *dw* (Qiu et al. 2019).

In Korea, two industrial bays were examined for DP in the cities of Pohang and Busan bay. Pohang has the biggest steel manufacturing complex in the world and Busan is surrounded by various industries. The average value of DP in Pohang was 270 pg g^{-1} *dw*, whereas, 72 pg g^{-1} *dw* was the mean value of DP detected in Busan. Elevated levels of DP in Pohang can be associated with the presence of steel manufacturing complex in the city (Fang et al. 2014). However, presence of DP in Busan can be attributed to the occurrence of fine sediment particles (Fang et al. 2014) as they have more adsorption capacity than coarser particles due to their large surface area (Gunawardana et al. 2014). Moreover, DP has also been reported in sediments of Daling river (Wang et al. 2012), Qiantang river (Sun et al. 2012), and Yellow sea (Zhao et al. 2011) of China. Literature supports that urbanization and industrialization are the major sources of DP pollution in sediments of urban areas (He et al. 2014a; Ma et al. 2011). Contrarily, restricted movement of water in dams has been reported as a main cause of DP pollution in sediments as tides play important role in distribution of DP (Sun et al. 2013). DP levels found in sediments of Asian countries are higher than Svalbard Fjord, Arctic (6.0 pg g^{-1} *dw*) (Ma et al. 2015).

DP has also been found in sediments of Ravi River, Pakistan (Syed et al. 2013). However, the measured concentrations (0.3 to 4.7 ng g^{-1} *dw*) were below the levels detected in sediments of Pearl River Delta (China) which exist near electronic manufacturing plant (He et al. 2014a). Comparable results have also been reported in the sediments of a Nullah near the industrial areas of Sialkot, Pakistan (Mahmood et al. 2015).

Biotic matrices

Existence of DP in biotic environmental matrices has been reported in literature (see Table 3). Wang et al. (2013b) reported the highest DP concentration (67,500 ng g^{-1} *lw*) in biotic samples of the world. DP has also been reported in the mangroves of Hainan Island, China. The average concentrations of DP in leaves, branches, roots, and fruits of the mangrove were reported to be 294, 181, 108, and 165 pg g^{-1} *dw*, respectively. These values are less than DP levels reported in tree barks of developed areas (Qiu et al. 2019). Presence of DP in mangroves can be associated with the atmospheric sedimentation. Almost similar levels of DP has been reported in the barks of rural *S. matsudana* and *S. babylonica* in inner Mongolia (Wulateqianqi and Dengkou), Ningxia Province (Guyuan, Yinchuan, Yongning, Qingtongxia, Zhongning, and Zhongwei), and Gansu Province (Jingtai, Baiyin, Pingchuan, and Lanzhou), China (He et al. 2014b). Another study reported the accumulation of DP in marine microalgae. DP reduces the light utilization capacity of *Ulva pertusa* as a notable decline in chlorophyll fluorescence parameters has been reported with increasing DP levels (Gong et al. 2018).

Aquatic biota

Bioaccumulation of DP in fishes was first time reported by Hoh et al. (2006). After that, presence of DP in fish samples collected from various locations of China (He et al. 2014a; Wang et al. 2012; Zhang et al. 2011a; Zhang et al. 2011b), Japan (Kakimoto et al. 2012), and South Korea (Kang et al. 2010) has also been described.

Wu et al. (2009) reported the presence of DP in various aquatic species; Chinese mystery snail (*Cipangopaludina chinensis*), mud carp (*Cirrhinus molitorella*), crucian carp (*Carassius carassius*), prawn (*Dendrobranchiata*), water

snake (*Nerodia*), and northern snakehead (*Channa argus*) were collected from a reservoir near an e-waste recycling plant in south China. The concentration of *syn*-DP and *anti*-DP in mud carp was 496 ± 310 and 1212 ± 842 ng g⁻¹ lw, respectively. However, the mud carp collected from reference site (with no DP emission source) had accumulated 1.35 ± 1.35 and 7.41 ± 7.32 ng g⁻¹ lw *syn*-DP and *anti*-DP, respectively (Wu et al. 2009). Furthermore, southwestern part of Northeast China has one of the most polluted rivers in China, i.e., Liaohe River. The commercial fishes were collected from this old industrial area and DP levels were examined in muscle tissues. The reported DP concentrations were ranged from below detection limit (BDL) to 470 pg g⁻¹ lw (Ren et al. 2013).

Similarly, King fishers and their prey fishes (*Macropodus opercularis*, *Gambusia affinis*, *Opsariichthys bidens*, *Abbottina rivularis*, *Nicholsicypris normalis*, and *Chinese bitterling*) were collected from Dinghu Mountain, which is an agricultural zone, and highly polluted Pearl River Delta, South China. DP concentration was examined in whole body of bird and fishes. It was reported that level of DP in prey fishes of Pearl River Delta were two orders of magnitude higher than the reference site. Furthermore, the recorded DP levels in King fishers (*Alcedinidae*) collected from Pearl River Delta and Dinghu Mountain were 58 ng g⁻¹ lw and 3.9 ng g⁻¹ lw, respectively (Mo et al. 2013).

In another study from e-waste recycling site at South Longtang Town China, two species, northern snakehead (*Channa argus*) and Crucian carp (*Carassius carassius*) were studied for maternal transfer and sex-related accumulation of DP. The concentrations of hepatic DP for northern snakehead and Crucian carp were 260–1920 ng g⁻¹ lw and 340 to 1670 ng g⁻¹ lw, respectively. These concentrations were much higher than the DP concentrations in Dinghu Mountain, Pearl River Delta. In eggs, DP concentrations were found in the range of 4.6–310 ng g⁻¹ lw. Results exhibited that higher levels of DP were present in males as compared to females. This shows maternal transfer of DP from female fish to egg (Wu et al. 2013). Another study was conducted to analyze the concentration of DP in various fish species. Common mullet (*Mugil cephalus*), Oriental goby (*Acanthogobius flavimanus*), Steed barbell (*Hemibarbus labeo*), temperate seabass, and crucian carp (*Channa argus*) were collected from rivers near the industrial areas of South Korea. There is no manufacturing plant in study area, yet DP was found in fish muscles (Kang et al. 2010). However, these levels were much lower than the concentrations reported in fishes of China (Ren et al. 2013; Wu et al. 2013).

Terrestrial biota

Inadequate data are available related to the occurrence of this widespread pollutant in terrestrial organisms. In Pearl River Delta, South China, concentrations of DP and its

accumulation trend were studied in muscles and liver tissues of light-vented bulbul (*Pycnonotus sinensis*), long-tailed shrike (*Lanius schach*), and Oriental magpie-robin (*Copsychus saularis*). High accumulation of DP was recorded in the liver as compared to the muscles. The concentrations were reported as 7.0 to 1300 ng g⁻¹ lw in the liver and 3.9 to 930 ng g⁻¹ lw in muscles. The accumulation was higher in urban areas than rural areas. The mean concentrations were reported as 300 ng g⁻¹ lw and 73 ng g⁻¹ lw in urban and rural areas, respectively (Sun et al. 2012). Another study reported the presence of DP in 5 bird species (*Ardeola bacchus*, *Amurornis phoenicurus*, *Gallirallus striatus*, *Porzana fusca*, and *Gallinago gallinago*) collected from Qingyuan county, a largest e-waste recycling plant of Pearl River Delta, China. The trend of DP accumulation in birds was kidney > liver > muscles (Zhang et al. 2010). Similar results have been reported by other studies in which DP showed high affinity with liver tissues than muscle, serum, gonad, and adipose tissues (Zeng et al. 2014; Peng et al. 2012). In Beijing, Northern China, muscles and liver tissues of six terrestrial raptors were examined, and recorded DP concentrations ranged between 9.5 and 810 ng g⁻¹ lw, respectively (Chen et al. 2013). In another study, accumulation pattern of DP was studied in people of an e-waste recycling industry and residential area. Comparison revealed that intake by workers was higher (dermal route 37-fold and inhalation fourfold) than the residents. Furthermore, children showed same response as of adult residents (Wang et al. 2013a).

In Pakistan, Mahmood et al. (2015) reported the presence of DP in wheat (*Triticum*) and rice (*Oryza sativa*), ranged from 0.49 to 0.90 ng g⁻¹ and 0.00 to 12.5 ng g⁻¹, respectively. However, the concentration of DP in food crops of China was reported to be 0.45–16.7 ng g⁻¹. The reason is in the former study; samples were collected from agricultural area near the DP source, whereas, in the latter study, plants were taken from an e-waste site (Chen et al. 2011). Another study reported the average DP concentrations of 1038 ng g⁻¹ ww and 877 ng g⁻¹ ww in vegetables and grains, respectively, collected from agricultural land in Huai'an, Jiangsu province, China (Wang et al. 2013a).

Foods

DP has been reported in foods, a major source of human exposure (Kakimoto et al. 2012, 2014; Malak et al. 2019). Kakimoto et al. (2012) first time reported the presence of DP in market fish of Japan. After that, a market basket study conducted in Osaka, Japan, revealed the presence of DP in certain foods, i.e., sugar and confectionary, legumes and their products, fish, shellfish, and their products, and meat and eggs, with levels of 3.3, 2.8, 1.9, and 1.5 pg g⁻¹ ww, respectively (Kakimoto et al. 2014). Since there is no DP manufacturing plant in Japan, these studies show that sea

Table 3 Levels of DP and its isomers in various biotic matrices

Matrices	Location	Species	syn-DP	anti-DP	∑ DP	References		
Aquatic biota (ng g ⁻¹ lw)	An e-waste recycling site, South China	Crucian carp (<i>Carassius carassius</i>)	134	142	276	Wu et al. 2009		
		Mud carp (<i>Cirrhinus molitorella</i>)	496	1210	1706			
		Northern snakehead (<i>Channa argus</i>)	224	30	254			
		Chinese mystery snail (<i>Cipangopaludina chinensis</i>)	3.17	17.06	20.23			
		Prawn (<i>Macrobrachium nipponense</i>)	64.13	126.2	190.33			
	Reference site, South China	Mud carp (<i>Cirrhinus molitorella</i>)	1.35	7.41	8.76	Ren et al. 2013		
		Fish Liaohu (<i>Microphysogobio liaohensis</i>)	0.082	0.141	0.223			
	Pearl River Delta, South China	Paradise fish (<i>Macropodus opercularis</i>)	77	110	187	Mo et al. 2013		
		Mosquito fish (<i>Gambusia affinis</i>)	32	47	79			
		Chinese hook snout carp (<i>Opsariichthys bidens</i>)	122	290	412			
		Chinese false gudgeon (<i>Abbottina rivularis</i>)	2	6.4	8.4			
		Nichols minnow (<i>Nicholsicypris normalis</i>)	0.35	1.4	1.75			
	South China Longtang Town	Northern snake head (<i>Channa argus</i>)	274.1	257	531.1	Wu et al. 2013		
		Crucian carp (<i>Channa argus</i>)	297	338	635			
		Common mullet (<i>Mugil cephalus</i>)	15.9	37.1	53			
	Ulsan–Onsan, South Korea	Oriental goby (<i>Acanthogobius flavimanus</i>)	7.2	11.62	18.82	Kang et al. 2010		
		Steed barbell (<i>Hemibarbus labeo</i>)	0.56	1.4	1.96			
		Temperate sea bass (<i>Dicentrarchus labrax</i>)	0.25	0.56	0.81			
		Crucian carp (<i>Channa argus</i>)	0.42	1.19	1.61			
		Mud carp (<i>Cirrhinus molitorella</i>)	2.2	16	18.2			
Dongguan, Southern China	Nile tilapia (<i>Oreochromis niloticus</i>)	5.9	19.5	25.4	He et al. 2014a			
	Plecostomus (<i>Hypostomus plecostomus</i>)	4.9	17.6	22.5				
	Fish australis (<i>Acanthopagrus Australia</i>)	0.18	0.46	0.64				
	E. Elongates (<i>Equulites elongatus</i>)	14	15	29				
Daling river, North Eastern China	E. Elongates (<i>Equulites elongatus</i>)	52.7	103.6	156.3	Wang et al. 2012			
	Pacific bluefin tuna (<i>Thunnus orientalis</i>)	0.54	1.04	1.58				
Aquatic biota (pg g ⁻¹ ww)	Nagasaki, Japan				Kakimoto et al. 2012			
Mangroves (pg g ⁻¹ dw)	Hainan Island, China	Leaves	–	–	294	(Qiu et al. 2019)		
		Branches	–	–	181			
		Roots	–	–	108			
		Fruits	–	–	165			
Human (ng g ⁻¹ lw)	Longtang Town, Qingyuan, China	Serum of e-waste workers	578	1640	2218	Yan et al. 2012		
		Serum of control	36.3	55	91.3			
	Guoyu, China	Serum of e-waste workers	16.2	22.5	38.7	Ren et al. 2009		
		Serum of control	4.3	7.5	11.8			
	Haojiang, China	Serum of e-waste workers	77	120	197	Chen et al. 2015		
		Serum of control						
	e-waste recycling site, Qingyuan, China	Wenling, China	Breast milk (had been involved in e-waste recycling)	10.4	27.4	37.8	Ben et al. 2013	
			Serum (had been involved in e-waste recycling)	25.4	46	71.4		
			Breast milk (control)	0.6	2	2.6		
			Serum (control)	1.3	3.5	4.8		
			Blood (worker having direct contact with DP)	286	471	757		Zhang et al. 2013
			Blood (workers without direct contact with DP)	143	207	350		
DP manufacturing plant, East China								

Table 3 (continued)

Matrices	Location	Species	syn- DP	anti- DP	∑ DP	References	
Human (ng g ⁻¹ dw)	3 km away DP manufacturing plant, East China	Blood (control)	106	207	313		
	DP manufacturing plant, East China	Hair (worker having direct contact with DP)	279	158	437		
		Hair (workers without direct contact with DP)	102	158	260		
	3 km away DP manufacturing plant, East China	Hair (control)	28.5	53.3	81.8		
	e-waste recycling site, Qingyuan, China	Hair	23	24	47	Chen et al. 2015	
Terrestrial biota (ng g ⁻¹ lw)	Sun Yat-sen University, Guangzhou, China	Hair (male)	n.d.	0.18	0.18	Qiao et al. 2018	
		Hair (female)	0.15	0.11	0.26		
	Pearl River Delta, South China Beijing, China	Kingfisher (<i>Alcedinidae</i>)	19	38	57	Mo et al. 2013	
		Common kestrel (<i>Falco tinnunculus</i>)	160	650	810	Chen et al. 2013	
		Eurasian sparrowhawk (<i>Accipiter nisus</i>)	21	80	101		
		Japanese sparrowhawk (<i>Accipiter gularis</i>)	21	65	86		
		Little owl (<i>Athene noctua</i>)	44	200	244		
Scops owl (<i>Otus</i>)	3	40	43				
Long-eared owl (<i>Asio otus</i>)	2.5	7	9.5				
Tree barks (ng g ⁻¹ lw)	Dhulikhel, Nepal	–	–	–	1.37	Salamova and Hites 2013	
	Bukit Kototabang, Indonesia	–	–	–	46.1		
Tree barks (pg g ⁻¹ dw)	Inner Mongolia Ningxia Province, China Gansu Province, China	<i>S. matsudana</i> and <i>S. babylonica</i>	–	–	–	23	He et al. 2014b
			–	–	–	62	
Crops (ng g ⁻¹)	Pakistan	Wheat (<i>Triticum</i>)	–	–	–	0.90	Mahmood et al. 2015
		Rice (<i>Oryza sativa</i>)	–	–	–	12.5	
	China	Eucalyptus (<i>Eucalyptus globulus</i>)	–	–	–	16.7	Chen et al. 2011
		Pine needles (<i>Pinus</i>)	–	–	–	51.9	
Vegetables (ng g ⁻¹ ww)	Huai'an, China	Green onion (<i>Allium fistulosum</i>)	–	–	–	2720	Wang et al. 2013a
		Pak choi cabbage (<i>Brassica rapa</i> subsp. <i>chinensis</i>)	–	–	–	305	
		Lettuce (<i>Lactuca sativa</i>)	–	–	–	679	
		Garlic chive (<i>Allium tuberosum</i>)	–	–	–	683	
		Field mustard (<i>Brassica rapa</i>)	–	–	–	802	
			–	–	–	–	
Grains (ng g ⁻¹ ww)	Huai'an, China	Common wheat (<i>Triticum aestivum</i>)	–	–	–	1370	Wang et al. 2013a
		Maize (<i>Zea mays</i>)	–	–	–	763	
		Soybean (<i>Glycine max</i>)	–	–	–	498	
Food (pg g ⁻¹ ww)	Beirut, Lebanon	Meat and poultry	1.8	8.1	9.9	Malak et al. 2019	
		Fish	4.2	3.9	8.1		
		Egg	3.1	5.8	8.9		
		Milk and dairy products	2	1.1	3.1		
		Fats and oils	25	28	53		
	Osaka, Japan	Fish	3.2	7	10.2	Kakimoto et al. 2012	
	Osaka, Japan	Sugar and confectionary	1	2.3	3.3	Kakimoto et al. 2014	
		Legumes and their products	0.9	1.9	2.8		
		Fish, shellfish, and their products	1	0.9	1.9		
		Meat and eggs	0.6	0.9	1.5		

lw lipid weight, ww wet weight, dw dry weight

water around east Asia is highly contaminated with DP due to the presence of DP source and recycling of electronic waste happening in China. Another study reported the presence of DP and related compounds in fish, egg, meat and poultry, milk and dairy products, and fats and oil samples collected from Beirut, Lebanon. Concentration of DP ranged from 1.7 to 52.8 $\mu\text{g g}^{-1}$ ww Lebanese foods (Malak et al. 2019). Although these concentrations are too low to cause health effects in humans, still further research is needed to mark a safe level for DP exposure.

Humans

Humans are exposed to DP through various sources. Major exposure routes are direct contact with DP, indoor dust, and food. Based on food habits, dietary exposure of DP to Lebanese adults is estimated to be 5.61 ng day^{-1} (Malak et al. 2019). In workers of e-waste site in Karachi Pakistan, the accidentally inhaled and ingested DP concentrations via soil are reported to be 5.42 and 1.9 $\text{ng kg}^{-1} \text{day}^{-1}$, respectively. The ingestion exposure is reported to be lower than inhaled; however, the ingestion values of DP were higher than the other flame retardants (Iqbal et al. 2017). Presence of DP in hair, serum, blood, breast milk, and hair of human has been reported in literature (Chen et al. 2015; Ben et al. 2013; Zhang et al. 2013). A study was conducted in Dhaka, Bangladesh, where T-shirts and wristbands were used to access DP exposure to the workers. Results revealed that wristband (0.44–208 $\text{ng dm}^2 \text{h}^{-1}$) accumulated more DP than T-shirts (0.07–71.9 $\text{ng dm}^2 \text{h}^{-1}$). So, clothes can also be a source of DP exposure to humans (Wang et al. 2020). Ren et al. (2009) collected and analyzed human serum samples from two different groups: workers of an e-waste recycling plant and control group (50 km away from this site). Results depicted a clear difference among the mean concentrations of DP between both groups. DP level in workers was recorded to be 38.7 $\text{ng g}^{-1} \text{lw}$, nevertheless, 11.8 $\text{ng g}^{-1} \text{lw}$ DP was found in the serum of control group. In Longtang Town, South China, concentrations of DP were measured in the workers of an e-waste recycling plant and residents of urban area. The concentrations determined were very high in the workers, up to 2218 $\text{ng g}^{-1} \text{lw}$, as compared to the residents of urban area with range 2.7–91 $\text{ng g}^{-1} \text{lw}$ having mean value of 4.6 $\text{ng g}^{-1} \text{lw}$. Furthermore, in females, there was a correlation between DP accumulation and their ages. Higher DP concentrations were recorded in females as compared to males due to high metabolic potential in males (Yan et al. 2012). Furthermore, they also reported that presence of DP was high in females aged above 50; however, no such relation of age has been found in men. Chen et al. (2015) reported that DP concentration was

tenfold higher in female hair as compared to the male workers. Similar results have been reported by Qiao et al. (2018). The levels of DP (0.26 $\text{ng g}^{-1} \text{dw}$) was higher in female students of Sun Yat-sen University, Guangzhou, China, as compared to male students (0.18 $\text{ng g}^{-1} \text{dw}$). This could be associated with the high exposure duration of DP to female hair than male. This gender-related variation shows that metabolism of DP varies in male and female which needs further investigation. Ben et al. (2013) reported that no stereoselective accumulation occur during transfer of DP from blood to milk. Zhang et al. (2013) reported the positive correlation between the presence of DP in blood and hair, whereas, Chen et al. (2015) stated that positive correlation exists in DP concentrations in hair and serum.

Wang et al. (2013a) calculated the hazard index (HI) for e-waste recycling workers. HI was calculated based upon the total estimated exposure doses and reference doses. Results revealed that calculated HI was three orders of magnitude lower than 1. Similarly, the measured hazard quotients (HQs) for oral, derma, and inhalation exposures were significantly low (Li et al. 2015; Wang et al. 2013a). Based on indoor dust, risk to human health is reported to be insignificant in Nepalese population as well (Yadav et al. 2017). Yadav et al. (2020) reported that inhalation exposure can cause more risk than dietary exposure. Similarly, HQs calculated for indoor and outdoor dust in various regions of Pakistan has also been reported < 1 (Khan et al. 2016). Although the HI and HQ values for DP are very low, but it has been detected in human bodies. Further studies are required for estimation of adverse effects and development of regulations.

Methodological and analytical aspects

The sample collection, extraction, cleanup, and equipment used for analysis have been presented in Table 4. Similar equipment and processes are used for analysis of DP as that for measurement of other lipophilic and volatile organic compounds, e.g. polychlorinated biphenyls (PCBs). Literature reported different method detection limits (MDI) for DP and its isomers; therefore, the lowest MDI has been stated in Table 4.

Sample collection

Abiotic matrices

Method of sample collection for the analysis of DP varies with the type of environments from where they are being collected (see Table 4).

Table 4 Experimental methods for analysis of DP in different biotic and abiotic matrices

Matrix	Sample collection/pre treatment	Extraction	Cleanup	Instrumental analysis	Detection limits	References
Air	High-volume air sampler equipped with a glass-fiber filter (GFF). Polyurethane foam passive air samplers (PUF-PAS). PUF	Ultrasonically dichloromethane, centrifuge and dissolved with in hexane Soxhlet-extracted for 24 h with DCM	Sulfuric acid added to silica gel column. Hexane used as an eluent Copper granules to remove sulfur. Eluted with 50 mL of DCM/hexane (1:1) Silica gel column after a hexane pre-rinse and eluted with hexane/DCM mixture (1:1, v/v)	GC/MS using a gas chromatograph, coupled with a high-resolution mass spectrometer GCECNI-MS applied with a DB5-MS capillary column Thermo Trace gas chromatograph coupled with a Thermo Polaris Q mass spectrometer (GC/MS) GC coupled with mass spectrometer (GC/MS) with MS capillary column	0.01 pg m ⁻³ * N.D. 0.0017 ng m ⁻³ *	Kakimoto et al. 2014 Syed et al. 2013 Wang et al. 2012
	PUF (15 cm diameter, volume, 256 cm ³). High-volume air sampler, glass fiber filters (GFF). Glass fiber filters (GFFs) Polyurethane foam (PUF) High volume air sampler.	Extracted with DCM and acetone: hexane (1:1 v/v) for 24 h Soxhlet extracted with a mixture of hexane and acetone (1:1) for 48 h	Cleaning column containing anhydrous sodium sulfate and silica gel, eluted with mixture of DCM and HEX(1:1 v/v) A silica/alumina column, eluted with mixture of hexane and dichloromethane (1:1)	GC coupled with mass spectrometer detector, (GC/MSD) with MS capillary column gas chromatograph coupled with a mass spectrometer (GC MS) using electron capture negative ionization (ECNI)	45 m ³ ** 0.15 pgm ⁻³ **	Ma et al. 2011 Chen et al. 2011
Sediments	Extracted using stainless spades and then sealed in polyethylene (PE) bags. Froze at -20 °C. Collected by hand trowel, kept in polyethylene bags, frozen dried, and sieved.	Extracted in a Soxhlet apparatus for 72 h using DCM. Soxhlet-extracted for 24 h with DCM	A modified column packed anhydrous sodium sulfate, sulfuric acid silica neutral silica gel, and neutral alumina from the top to bottom was used for clean-up. Copper granules to remove sulfur. Alumina/silica column packed, from the bottom to top. Eluted with 50 mL of DCM/hexane (1:1)	GC equipped with fused silica capillary, Column using methane as the reagent gas GCECNI-MS applied with a DB5-MS capillary column	0.4 pg g ⁻¹ ** N.D.	Zhao et al. 2011 Syed et al. 2013
Soil	Collected using stainless steel grab sampler (top 0–10 cm) freezer at -20 °C and sieved. A DIK-115B soil sample was used and stored at -20 °C in a refrigerator. Samples taken using hand trowel, kept in polyethylene	Soxhlet extracted with dichloromethane (DCM) for 48 h Extracted with dichloromethane using a PSE (pressurized solvent extraction) system Soxhlet-extracted for 24 h with DCM	Silica/alumina/Florisil column with anhydrous sodium sulfate, neutral Florisil, neutral activated silica gel, neutral alumina, and anhydrous sodium sulfate. The Dechlorane mixture was eluted hexane: dichloromethane (7:3) Samples were loaded on an alumina column with anhydrous Na ₂ SO ₄ and activated copper powder.	GC coupled with mass selective detector (GC/MSD) in electron capture negative ionization (ECNI) mode using methane as the moderating gas. Gas chromatography mass spectrometry (GC/MS) GCECNI-MS applied with a DB5-MS capillary column	0.009 ng g ⁻¹ dw* 2.8 pgg ⁻¹ dw** N.D.	Sun et al. 2013 Wang et al. 2010b Syed et al. 2013

Table 4 (continued)

Matrix	Sample collection/pre treatment	Extraction	Cleanup	Instrumental analysis	Detection limits	References
	bags frozen dried and sieved.		Copper granules to remove sulfur. Alumina/silica column packed, from the bottom to top. Eluted with 50 mL of			
	Samples collected with stainless steel scoop that had been prewashed by acetone.	Samples were extracted for 24 h with 100 mL mix solvents (hexane/acetone, 1:1 v/v) in Soxhlet apparatus.	Extracts were filtered through a funnel filled with anhydrous sodium sulfate and then rotary-evaporated. The extracts were passed through silica gel column.	GC/MSD equipped with MS capillary column in selected ion monitoring (SIM) mode.	0.9 pg m ⁻³ dw**	Ma et al. 2011
Water	Brown glass bottles used to collect water samples. Bacterial decay stopped by adding dichloromethane. Stored at 4 °C in dark.	Liquid-liquid extracted, DCM	Silica column. Hexane/DCM (1:1)	Agilent 6890 GC coupled with Agilent 5973 N mass spectrometer detector (GC/MSD)	40 pg L ⁻¹ **	Ma et al. 2011
	Water collected in acetone-rinsed brown glass bottles mixed with dichloromethane (DCM) for storage at 4 °C.	Extracted with 100 mL DCM in a separator funnel with agitation followed by a 1-h settling time	40 mL of hexane/DCM mixture (1:1, v/v).	Thermo Trace gas chromatograph coupled with a Thermo Polaris Q mass spectrometer (GC/MS)	0.027 ng L ⁻¹ *	Wang et al. 2012
Aquatic biota	Muscle tissue separated from fish samples homogenized, freeze dried, and stored in Teflon-lined capped glass jar.	Dichloromethane using a Dionex ASE 200 accelerated solvent extraction (ASE) used for extraction	Gel permeation chromatography. Multi-layer silica gel column.	Gas chromatography-high resolution mass spectrometry (GC-HRMS). gas chromatography coupled with a JEOL 800D mass spectrometry with the electron impact (EI) mode	N.D.	Kang et al. 2010
Terrestrial biota	Pectoral muscles and the liver were removed from each bird and stored at -20 °C	Muscles and liver sample homogenized and Soxhlet extracted for 48 h with acetone and hexane	Purified by gel permeation chromatography and eluted with dichloromethane/hexane (v/v = 1:1).	(GC/MS) using electron capture negative ionization (ECNI) in the selective ion monitoring mode	N.D.	Sun et al. 2012
	Captured by plastic bird netting and stretched mesh bottom set gillnet.	48-h Soxhlet extracted using acetone/hexane mixture (1/1, v/v).	Samples purified using gel permeation chromatography (GPC) and multilayer silica column.	Gas chromatograph coupled with a single quadrupole mass spectrometer in electron capture negative ionization (ECNI) mode	0.01 ng g ⁻¹ lw**	Mo et al. 2013
Humans	Within anticoagulant free tube 8–10 ml venous blood sample collected. Serum was frozen and stored at -80 °C	Serum was denatured using 1.5 ml of 6 M HCl. Extracted with 10 ml hexane/methyl-tert-butyl ether (MTBE) (1:1, v/v)	Multi-layer silica/alumina column hexane/dichloromethane (1:1, v/v) 35 ml	Gas chromatograph coupled with a mass spectrometer with electron capture negative ionization	N.D.	Yan et al. 2012

N.D. not defined, *instrument detection limit (IDL), **method detection limit (MDI)

- For air sampling, both active and passive samplers can be used for the measurements of DP in both gaseous and particulate phase. For active sampling, high volume air samplers are used with glass fiber filter. However, for passive sampling, mostly for particulate phase, polyurethane foam passive air samplers are used (Kakimoto et al. 2014; Syed et al. 2013; Wang et al. 2012; Ma et al. 2011; Chen et al. 2011).
- Soil samples are collected using hand trowel and stainless steel scoop. These samples must be kept in polyethylene bags, frozen, dried, and sieved before use (Syed et al. 2013; Ma et al. 2011; Wang et al. 2010b).
- Amber glass bottles are used for water sampling to avoid the effect of light. Further, di-chloromethane is used to prevent bacterial decomposition. Samples are stored in dark conditions at 4 °C (Ma et al. 2011).
- For sediment sampling, various techniques, i.e., stainless steel spades, hand trowel, stainless steel grab samplers, are employed and sealed in polyethylene bags. Samples are stored at –20 °C (Syed et al. 2013; Sun et al. 2013; Zhao et al. 2011).

Biotic matrices

Biotic matrices are difficult to handle; therefore, various methods are adopted for their collection (see Table 4). Muscle tissues and liver are extracted and homogenized from individual species to get a common pool sample. Samples are frozen and dried to use for analysis (Ren et al. 2013; Mo et al. 2013; Kang et al. 2010). However, in case of humans, venous blood is collected in anticoagulant free tubes; serum is separated and stored at –80 °C (Yan et al. 2012). While, in case of birds, muscles and liver are separated and homogenized for further analysis (Sun et al. 2012).

Extraction of samples

Abiotic matrices

In case of air samples, extraction is carried out ultrasonically with dichloromethane. Soxhlet extraction is also used involving different solvents like acetone hexane (1:1 v/v) for 24 h and in some studies for 48 h. Soxhlet extraction with DCM has also been used in some studies (Kakimoto et al. 2014; Syed et al. 2013; Wang et al. 2012; Ma et al. 2011; Chen et al. 2011). For soil samples, pressurized solvent extraction system is used. Soxhlet extraction with DCM and hexane/acetone has also been implied (Syed et al. 2013; Ma et al. 2011; Wang et al. 2010b). In case of water samples, liquid-liquid extraction with DCM and Soxhlet extraction with DCM was

employed (Ma et al. 2011). For sediment samples, Soxhlet extraction was done with DCM for both 48 and 72 h (Syed et al. 2013; Sun et al. 2013; Zhao et al. 2011).

Biotic matrices

Biological samples can be extracted using Soxhlet extraction with hexane and acetone (1:1 v/v) for 48 and 72 h in different studies for fishes (Ren et al. 2013; Mo et al. 2013; Kang et al. 2010), and mist nets are used for the collection of avifauna (Zhang et al. 2011). For humans, extractions are carried out using hexane and methyl-tert-butyl ether (Yan et al. 2012).

Cleanup of samples

Abiotic matrices

For cleanup of abiotic samples, alumina and silica gel columns are used, which are re-eluted with di-chloro-methane or hexane. Moreover, copper granules are also used to remove sulfur (Kakimoto et al. 2014; Syed et al. 2013; Ma et al. 2011; Zhao et al. 2011; Wang et al. 2010b).

Biotic matrices

Multilayer silica-alumina column chromatography or gel permeation chromatography are employed for the purification of aquatic biota (Ren et al. 2013; Mo et al. 2013; Kang et al. 2010). For avifauna, gel permeation chromatography, eluted with dichloromethane and hexane, is used (Sun et al. 2012). In case of human samples, multi-layer silica-alumina column are used with solvents like hexane and di-chloro-methane (Yan et al. 2012).

Instrumental analysis

Abiotic matrices

Various instruments can be used for the analysis of DP in different matrices. For air samples, gas chromatography coupled with mass spectrometry (GC-MS), gas chromatography coupled with electron capture negative ion mass spectrometry (GC/ECNI-MS), and thermo trace gas chromatography and gas chromatography with mass spectrometer detector (GC/MSD) are used (Kakimoto et al. 2014; Syed et al. 2013; Wang et al. 2012; Chen et al. 2011).

For sediment samples, analysis is performed using gas chromatography (GC) with fused silica capillary column whereas methane is used as reagent gas. However, GC/ECNI-MS is also appropriate for sediment samples (Syed et al. 2013; Sun et al. 2013; Zhao et al. 2011). For soil and

water samples, gas chromatography with mass spectrometer detector (GC/MSD) is useful (Syed et al. 2013; Ma et al. 2011; Wang et al. 2010b).

Biotic matrices

For fishes, GC with selected ion monitoring mode is used. Gas chromatography coupled with a single quadrupole mass spectrometer (GC-MS) in electron capture negative ionization (ECNI) mode is also used (Ren et al. 2013; Mo et al. 2013; Kang et al. 2010). This equipment is also suitable for the analysis of samples collected from humans and other terrestrial biota (Yan et al. 2012).

Toxicity summary of DP

Toxicity data on DP are limited. Most of the data are presented in Oxychem manual (Dou et al. 2015; Crump et al. 2011; Wang et al. 2013a). DP caused no ocular irritation in Albino rabbit and no skin sensitization in Guinea pig. Moreover, no mutagenicity was observed as a result of standard Ames assay, mouse lymphoma forward mutation assay (both with and without activation), and Ames assay of urine from rats treated with DP (Oxychem 2011). Moreover, it produced no adverse effects in male Sprague Dawley rats after continuous exposure (0, 1, 10, and 100 mg kg⁻¹ day⁻¹) for 90 days (Li et al. 2013). Similarly, no observed effect level of DP in Sprague Dawley rats (6 to 8 weeks old) is reported to be 5000 mg kg⁻¹ day⁻¹ (Brock et al. 2010). DP induced oxidative damage and changes in acetylcholinesterase (AChE) activities, stress response, calcium binding, neurological dysfunction and signal transduction, and altered gene expression in earthworm after 14 days exposure at 0.1, 1, 10, and 50 mg kg⁻¹ concentrations (Zhang et al. 2014). Toxicity of DP increases with increased exposure time (Yang et al. 2016). Furthermore, genotoxicity of DP is species-specific as it caused DNA damage in protozoa (Dou et al. 2015) while no genotoxicity was observed in rats at same concentration (Wu et al. 2012). Kang et al. (2016) reported that environmentally relevant levels (0 to 3 µg g⁻¹ fish ww) of DP have potential to disrupt thyroid and sex hormones in zebrafish.

Wang et al. (2013b) has performed human health risk assessment among the most polluted areas of DP near e-waste recycling site in Huai'an in Jiangsu province of China. Reference doses (oral, dermal, and inhalation routes) were selected based upon literature available in US EPA HPV reports. Results revealed that hazard quotient was three orders of magnitude lower than 1. They concluded that occupational exposures were safe; however, bioaccumulation and metabolites of DP can induce

toxicity (Wang et al. 2013a). Toxicity summary of DP has been presented in Table 5.

Bioaccumulation and isomeric accumulation trends

Various studies have reported that DP has more affinity towards the liver (Zeng et al. 2014; Peng et al. 2012; Zhang et al. 2010), which can be associated to its lipophilic nature. Literature stated that disposition of DP in the liver can also be caused by sequestration DP isomers by hepatic proteins, though such proteins have not been identified yet (Zeng et al. 2014; Li et al. 2013a; Peng et al. 2012). Fraction of *syn*-DP or *anti*-DP are usually calculated to check the bioaccumulation, fate, and movement of DP in the environment (Wu et al. 2009). The accumulation of both isomers differs in organisms depending upon different factors, i.e., sources, bioavailability of isomer, inter species difference, and biotransformation of both isomers (Tomy et al. 2007). Studies revealed that accumulation and variation of different isomers in aquatic and terrestrial species differ with trophic level and bio degradation ability (Xian et al. 2011). The larger is the trophic level, the lesser is the *anti*-DP level. Higher organisms have more effective metabolism to degrade *anti*-DP. The concentration of DP *syn*-isomer was reported high in higher organisms and it is more lipophilic (Wu et al. 2009). In a study conducted in South China, DP and its analogs (dechlorinated products *anti* Cl₁₁ DP) were studied in the workers of an e-waste recycling and residents of city. Genders were compared too. Accumulation of DP varied with the age of females; however, this trend was not observed in case of males. Additionally, *anti*-DP and *anti* Cl₁₁ DP were found higher in females. The lower DP levels in males exhibited that they have better metabolism than females (Yan et al. 2012). Moreover, high levels of DP have been reported in the livers of quails as compared to the muscle tissues. Additionally, *syn*-DP was found higher in high-dose groups while concentration of *anti*-DP reduced with increasing dose. These results revealed that dose-based pharmacokinetic of DP exists in quails (Li et al. 2013a). Another study reported that children have same DP accumulation trend as of adults (Wang et al. 2013b). Furthermore, the accumulation of DP was also studied in wild frogs from an e-waste site in Southeast China. Brain tissue, the liver, and muscles of frogs were analyzed. It was observed that *anti*-DP was accumulated more in muscles as compared to the liver, whereas, *syn*-DP was found higher in liver tissues. Further, the blood brain barrier was working to reduce the amount of DP entering in brain tissues (Li et al. 2014).

Table 5 Toxicity summary of DP

Toxicity study	Exposure routes	Test species	Results	References
Acute toxicity	Oral	Albino rat (<i>Rattus norvegicus</i>)	LD ₅₀ = 25,000 mg kg ⁻¹	(Oxychem 2011)
	Dermal	Albino rabbit (<i>Rattus norvegicus</i>)	LD ₅₀ = 8000 mg kg ⁻¹	(Oxychem 2011)
	Inhalation	Rat (<i>Rattus</i>)	LC ₅₀ = 2.25 mg L ⁻¹	(Oxychem 2011)
	Oral (28 days)	Sprague Dawley rats (<i>Rattus norvegicus</i>)	No effects were observed on clinical pathology, fertility indices, fetal development	(Brock et al. 2010)
	Dermal (28 days)	Female rabbits (<i>Oryctolagus cuniculus</i>)	Decline in the liver and ovary weight at 2000 mg kg ⁻¹ bw	(USEPA HPV 2008)
	Bioassay	Luminous bacteria	Luminosities remained 100% (at 369 µg L ⁻¹)	(Dou et al. 2015)
	Micronucleus	<i>Vicia faba</i>	No mutagenicity was observed (at 1500 µg L ⁻¹)	(Dou et al. 2015)
	Comet assay	Protozoa (<i>Tetrahymena thermophila</i>)	DNA damage was observed at concentration ≥ 300 µg L ⁻¹ .	(Dou et al. 2015)
	Soil with DP (14 days)	Earthworms (<i>Eisenia fetida</i>)	Oxidative damage, neurotoxicity, altered gene expression	(Zhang et al. 2014)
	Soil with DP (28 days)	Earthworms (<i>Eisenia fetida</i>)	Change in SOD and CAT activities, altered GSH levels, increased level of tDNA, and increased tail length even at 0.1 mg kg ⁻¹ concentration.	(Yang et al. 2016)
Sub chronic toxicity	Oral (90 days)	Albino rat (<i>Rattus norvegicus</i>)	At 1, 3, and 10% dietary levels, no difference between test and control animals was observed except slight increase in liver weight at 10% dietary level and not below. No mortalities were observed. Behavioral, growth and food intake patterns remained unaffected. Clinical blood chemistry, hematology, urinalysis, gross pathology, and histopathology at all feed levels were same as control.	(Oxychem 2011)
	Dermal (28 days)	Albino rabbit (<i>Rattus norvegicus</i>)	At 500 and 2000 mg/kg exposure, only weight of gonads in females was decreased while lower liver weight was observed at exposure level of 2000 mg/kg. However, body weight, hematology, blood chemistry, urinalysis, and pathology remained unaffected.	(Oxychem 2011)
	Dust inhalation (28 days)	Albino Rat (<i>Rattus norvegicus</i>)	At 640 and 1540 mg/cm exposure level, liver weight, lungs weight as well as lung to body weight ratios were higher than normal. Reversible histopathological changes in lung tissues were also observed. However, body weight, blood chemistry, clinical chemistry, and urinalysis, and gross pathological examination remained unaffected.	(Oxychem 2011)
	Oral (90 days)	Sprague Dawley rats (<i>Rattus norvegicus</i>)	No adverse effects were observed on the liver and body weight, endocrine levels, and enzyme activity	(Li et al. 2013)
Aquatic toxicity	–	Bluegill Sunfish (<i>Lepomis macrochirus</i>)	After 96 h, no mortalities were observed at concentration level up to 100 ppm	(Oxychem 2011)

Correlation

Correlation was calculated between two isomers (*syn*-DP and *anti*-DP) and species based upon the results of previously reported studies presented in Table 3. Both isomers showed positive correlation at the significance level of 5% probability (Table 6). Correlation is justified based on same nature of both isomers, i.e., lipophilicity. Although, *syn*-DP has high rate of deposition in fatty acid tissues in comparison with *anti*-DP; however, a steady

increase in concentration of latter isomers has been observed in case of exposure to the former isomer. However, no significant correlation has been observed between species and isomers ($p > 0.05$).

Cluster analysis

Cluster analysis was applied on the information of DP found in aquatic biota (presented in Table 3), and its results have been revealed in Fig. 2. In this dendrogram, species have been

Table 6 Spearman correlation between *syn*-DP, *anti*-DP, and species (*n* = 26)

		Species	<i>Syn</i> -DP	<i>Anti</i> -DP
Species	Correlation coefficient	1	-0.358	-0.346
	Sig. (2-tailed)		0.073	0.084
<i>Syn</i> -DP	Correlation coefficient		1	<i>0.962</i>
	Sig. (2-tailed)			0.000
<i>Anti</i> -DP	Correlation coefficient			1

Italic value represents significantly positive correlation

divided into three main groups based upon the levels of *syn*-DP and *anti*-DP isomers. Accumulation of *syn*-DP and *anti*-DP is preferential (Table 3), but the reason is still unknown. Green color showed highest similarities among species of G-3 while accumulation trend in G-2 (blue) was found in between G-1 and G-3. G-1 showed highest variability as compared to G-2 and G-3, hence shown by red color (see Fig. 2). Because accumulation trend of both isomers varies between both species and within the species as well which proves that accumulation trend of both isomers is species independent. Conversely, both G-2 and G-3 clusters have almost similar accumulation trend, i.e., levels of *anti*-DP are higher than *syn*-DP, and the increment in the accumulation of latter enhances the level of former isomer (Table 6). However, the levels accumulated by the species of G-3 cluster varied from the levels found in the species of G-2. The largest group has been formed by G3 which represents species having quite similar accumulation of both isomers. Within G-3, australis (*A. Australia*) and temperate sea bream (*D. labrax*) has shown highest resemblance as the isomeric levels found in both species were highly similar (shown in Table 3). Afterwards, fish liaohoe (*M. liaohensis*) presented the highest similarity with australis and temperate sea bream than other species; however, height of the cluster revealed that the difference between values was slightly high. Additionally, paradise fish (*M. opercularis*) has the highest similarity with crustacean carp followed by Chinese hook snout carp (*O. bidens*), crucian carp (*C. carassius*), and northern snakehead (*C. argus*). There is a sub-group in G-3, from fish liaohoe to Chinese bitterling (*R. ocellatus*); these species have found to have high similarities than the whole G-3 cluster (Fig. 2). Similarly, in G-2 cluster, the highest resemblance of accumulation trend has been found between common mullet (*M. cephalus*) and kingfisher (*Alcedinidae*) followed by Chinese mystery snail (*Cipangopaludina chinensis*). However, the lowest similarity has been found between mosquito fish (*G. affinis*) and fish qingyuan (*A. pseudoharengus*) (Fig. 2). From the abovementioned information, it appears that accumulation trend of *syn*-DP and *anti*-DP is considered species-independent as proved by correlation co-efficient (Table 6).

Conclusions and recommendations

- In the perspective of above discussion, it can be concluded that Dechlorane Plus is an emerging global pollutant because of its presence in aquatic, terrestrial, and avifauna as well as in humans.
- Occurrence of this pollutant in different environmental matrices revealed that it is ubiquitous, and both humans and environment are susceptible to it. However, very limited studies have been conducted in Asia to understand the distribution, monitoring, bioaccumulation, and toxicity of DP and its analogs.
- The studies already conducted in Asia reveals its presence in the continent, but these studies are restricted to some specific areas. There is a dire need of more researches which explains the occurrence of DP and its analogs in other countries.
- Concentration trends of DP are inversely proportional to the distance from its emission sources. It can also be found in regions with no DP source which presents its long-range transport. It is needed to develop some rules and policies before it imposes a negative externality to the environment and affect humans.
- Moreover, data on biomagnification of DP in aquatic ecosystem are scarce, and information about degradation products of DP in terrestrial food web is also limited.
- Additionally, medium-specific half-lives of DP and its analogs are yet to be determined.
- Although HQs values calculated for DP are found below 1, however, toxicity mechanism must be evaluated since its presence in human blood, serum, hair, breast milk, and adipose tissues is detected.
- Toxicity mechanism of DP is still not clear. Nonetheless, few studies have been conducted on animals to check its effects; however, for humans, there is still uncertainty.
- Accumulation of *syn*-DP and *anti*-DP is preferential, but the reason is still unknown. No literature is available on toxicity behavior of *syn*-DP and *anti*-DP.
- Acute and chronic toxicity studies should also be conducted in the future for DP and its analogs.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflicts of interest.

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