Interactions of molybdenum disulfide nanosheets with wheat plants under changing environments: more than meets the eye?

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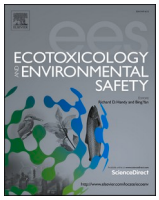

Evaluating the spatial and temporal distribution of emerging contaminants in the Pearl River Basin for regulating purposes

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ABSTRACT

Little information is available on how the types, concentrations, and distribution of chemicals have evolved over the years. The objective of the present study is therefore to review the spatial and temporal distribution profile of emerging contaminants with limited toxicology data in the pearl river basin over the years to build up the emerging contaminants database in this region for risk assessment and regulatory purposes. The result revealed that seven groups of emerging contaminants were abundant in this region, and many emerging contaminants had been detected at much higher concentrations before 2011. Specifically, antibiotics, phenolic compounds, and acidic pharmaceuticals were the most abundant emerging contaminants detected in the aquatic compartment, while phenolic compounds were of the most profound concern in soil. Flame retardants and plastics were the most frequently studied chemicals in organisms. The abundance of the field concentrations and frequencies varied considerably over the years, and currently available data can hardly be used for regulation purposes. It is suggested that watershed management should establish a regular monitoring scheme and comprehensive database to monitor the distribution of emerging contaminants considering the highly condensed population in this region. The priority monitoring list should be formed in consideration of historical abundance, potential toxic effects of emerging contaminants as well as the distribution of heavily polluting industries in the region.

1. Introduction

With increasing industrialization and urbanization, different types of new chemicals are continuously being designed, and the quantities of chemicals produced kept increasing over the years. The concept of "emerging contaminants" was adopted to demonstrate that all contaminants generated during production, construction, or other activities that are caused by human activities, which are clearly present but not yet regulated by laws, regulations, and standards, or are not well regulated, and are harmful to the living and ecological environment (Bao et al., 2015; Khan et al., 2018; Naidu et al., 2016). Currently, chemicals including pharmaceuticals, antibiotics, pesticides, illicit drugs, clothing adhesives, cleaning solvents, antimicrobial agents, cosmetics, beauty care, sunscreen, personal care products, beverages, and packaged foods and plastics can all be called emerging contaminants (ECs) (Rathi et al., 2021; Richardson and Kimura, 2017), which have been widely applied in our daily lives (Zhang et al., 2015a; Salamova et al., 2014; van der Veen and de Boer, 2012; Ashesh et al., 2022) (Wang and Kannan, 2018; Druschky et al., 2018; Liu et al., 2017; Gao et al., 2018; Li et al., 2021a; Lee et al., 2010; Roosens et al., 2007). ECs have been detected in the environment frequently as a result of their wide application in industrial processes and daily life. For instance, different antibiotics have been widely reported in freshwater, groundwater, and sewage sludge in Europe, Africa, America, and Asia – Pacific (Carvalho and Santos, 2016; Danner et al., 2019; Hanna et al., 2018). Many highly toxic pesticides have been banned for crops, but they can still be monitored at high frequencies due to their stability, resistance to environmental degradation, and bioaccumulation (Jayaraj et al., 2016; Kafaei et al., 2020). One hundred and two pesticides were detected in water, sediment, and clam samples in the Jiulong River and estuary (Wang et al., 2017a; Zheng

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et al., 2016); PCBs are still present in surface water, sediment, and suspended particulate matter along the Yangtze River (Cui et al., 2020). A recent study screened the ECs in the surface water, sediment, organisms, and groundwater of the Siverskvi Donets River Basin. The study revealed the presence of 83 types of ECs in the monitoring samples, in which bisphenol A, DEPH, and prometryn were found to potentially pose a risk to the ecosystem, whereas high concentrations of PCBs degradation products were detected in the sediment (Nikolopoulou et al., 2022).

ECs can cause adverse effects on various organisms in the environment and affect human health. For example, pharmaceuticals can affect phytoplankton and algal growth and cause reproductive defects in aquatic wildlife (Mohapatra et al., 2016). Antibiotics have been reported to inhibit the growth of green algae, duckweed, and cyanobacteria, hinder postembryonic development among amphibians, affect the reproduction system of aquatic organisms, and they may cause gastric ulceration, mucosal damage, dyspepsia, and bowel inflammation (Patel et al., 2019; Subedi et al., 2017; Karthikeyan and Meyer, 2006). Moreover, estradiol might affect the reproductive systems of fish and humans severely. This may cause congenital disabilities, abnormal sexual development, and cancer (Patel et al., 2019; Estrada-Arriaga et al., 2016). Besides, engineered nanoparticles can cause genotoxicity, DNA mutations, and impaired expression of functional proteins in different organisms, resulting in toxic damage to the organism (Song et al., 2014). Triclosan leads to genotoxicity by increasing micronucleus frequency (Binelli et al., 2009), and bisphenol A present genotoxicity by introducing chromosomal aberrations (Allard and Colaiacovo, 2010). Brominated flame retardants can cause adverse effects through different modes of action (hormone disruption, endocrine disruption, neurotoxicity, DNA damage, etc.), such as alteration of thyroid hormone homeostasis and delayed hatching in zebrafish (Xiong et al., 2019). Plastic containing plasticizers such as phthalate esters (PAEs) contribute to severe organic pollution and soil degradation (Zhang et al., 2020a). Therefore, it is essential to monitor the concentrations of ECs and evaluate the risks of ECs in order to prevent potential adverse effects of ECs on humans and the environment.

Various databases and regulations have been developed worldwide to document adverse effects of chemicals and to evaluate the risk of these chemicals. The ECOTOX knowledgebase is one of the largest ecotoxicology databases, as developed by the U.S. Environmental Protection Agency in 1981 and maintained thereafter. The database currently contains toxicity data on 12485 chemicals for various environmentally relevant species (EPA, 2022). The ECOTOX knowledge base is regularly updated by collecting the newest data from the literature (EPA, 2022). The European Union has developed various databases to track newly developed chemicals, including the tracing of all ingredients in cosmetics and the CP – DS database for identifying all relevant regulations on dangerous substances in construction products (European Union, 2022). In terms of regulation, the European Union developed a comprehensive process for the Registration, Evaluation, and Authorisation of CHemicals (REACH regulation). The chemical manufacturers and importers must identify and manage risks related to the chemicals they produce and sell in the European Union (European Union, 2022). China has also updated its regulations on environmental risk assessment and registration of chemical substances in 2019. The regular updates on the ecotoxicology database and the regulation of chemicals revealed concerns about the risk of new substances, especially these ECs that have been detected in the environment but have not yet been regulated.

Although the ecotoxicology databases are updated regularly to include ECs, it is a lack of overview on the environmental distribution of ECs in the field over time. Without known the abundance and concentration of ECs in the environment, it is impossible to carry out the risk assessment of ECs and hardly mention the regulation of ECs. It is essential to review and build up a comprehensive profile of ECs over the years for the proper risk assessment of ECs in the environment for two reasons: Firstly, the overall class of ECs contains chemicals which have different chemical properties and different fate environmental fate profiles. The historical, typically hydrophobic, ECs could for example be present in the environment for a long time. Tracking ECs profiles over the years can help understand the behavior and degradation profile of ECs better. Secondly, a comprehensive ECs profile allows evaluating the risk of mixtures of ECs in the environment and help the regulation and remediation of ECs in the regions.

The PR is the second–largest river in China when judged by flow, and the PRB has experienced rapid economic development (GDP > US$400 million) and population growth (>80 million inhabitants) (Tao et al., 2022). The environmental quality of urban rivers has deteriorated due to direct and indirect inputs from enhanced human activities. Environmental contaminants from various sources enter the PRB and eventually discharge into the South China Sea via the pearl river estuary in multiple ways (Dai et al., 2018a). Although many studies focused on investigating the fate, behavior, and pathways of different types of chemicals in this region, there is no clear overview of the abundance of ECs in this river. It is also not clear whether the types of chemicals present have evolved over the years, and if the ECs present in this river would potentially induce health and/or ecological risks. With the national and public emphasis on environmental protection and human health, it is critical to establish systematic datasets of the presence of ECs and apply a big data approach to manage and assess the distribution and potential risks of ECs.

The objective of the present study is therefore to review the emergence profile of ECs in the PRB over the years to build up the ECs database in this region for the purpose of risk assessment. The research trends and the spatial and temporal distribution of ECs are analyzed in detail. Then, the pollution levels are discussed and compared to the situation in other countries and regions. The potential bottlenecks, limitations of the current study as well as advices for developing and managing the ECs database are highlighted in the discussion section, followed by a summary of the key findings in the conclusions section.

2. Methodology

2.1. Data collection

To survey ECs in the PRB, we used the Web of Science (WOS) database to retrieve relevant publications. The database was searched for studies published up to 25 October 2021. The keywords were limited to “pearl river” and “emerging contaminants” or “emerging pollutants”. Finally, 229 articles were obtained, of which 80 were duplicates, and 149 remained after removing the duplicates. The keywords of all the retrieved literature were extracted for visual keyword mapping to analyze the current research status of ECs in the PRB. Data on the concentration of all ECs mentioned in the literature were collected and extended to relevant cited literature data. Although the literature reviewed was published between 2013 and 2021, the actual concentration of ECs reported by this literature ranged from 2002 to 2021, due to other reasons such as the time gap between publication of the paper and the experimental study.

2.2. Statistical analysis

The cluster-based VOS viewer (version 1.6.18, https://www.vosviewer.com/) is used to perform the analysis. The software enables users to create, visualize, and explore scientific mappings in cluster format based on scientific network data. Using the complete count method, each co-author and co-occurrence has the same weight, regardless of the order and number of authors in the author list. The co-occurrence analysis identified items (e.g., publications, researchers, keywords, and authors of interest) based on their relevance and the number of publications in which they occurred. Publications to date about overall ECs studies in the PR were scanned in WOSCC for a detailed bibliometric analysis using the VOS viewer (version 1.6.18) because of its unique text mining feature suitable for visualizing large networks. The data

clustering techniques of this software have been explained previously (van Eck and Waltman, 2009). The co-occurrence analysis of all keywords (in the title, abstract, or keyword list) (Quino Lima et al., 2020) generated 4667 results, and 79 keywords were selected based on the threshold of 10 co-occurrences according to outputs generated by the VOS viewer WOSCC data. Developing the visualization plots after screening for irrelevant words such as "ng/L", "world", "type" etc. Based on the set threshold (minimum occurrences of common keywords were selected = 2 for this analysis) and initial screenings to eliminate the repetitive and irrelevant keywords.

The mapping and spatial analysis were performed using ArcGIS 10.6 software, and IDW interpolation was applied to recognize the overall spatial distribution of geospatial data. The geospatial maps showed the spatial distribution of contamination areas and the pollution status of the studied area.

Typical representatives of each pollutant were selected to search and choose the no observed effect concentration (NOEC) and lowest observed effect concentration (LOEC) values through EPA EXOTOX, exposure media limited to freshwater, species group selected fish and Crustaceans, the search results are shown in Supplementary material. In aquatic toxicity the maximal acceptable toxicant concentration (MATC) was often calculated. This is the geometric mean of the NOEC and the LOEC. If in the test report only the NOEC is presented, the NOEC is multiplied by $\sqrt{2}$ to get the MATC (Agency, 2008).

$$MATC = \sqrt{NOEC * LOEC} \quad (1)$$

$$MATC = \sqrt{2}NOEC(\text{Only the NOEC is presented}) \quad (2)$$

3. Results

3.1. Abundance of ECs in the PRB region

The ECs that have been reported in the PRB can be grouped into seven categories: antibiotics, flame retardants, pesticides, phenolic compounds, acidic drugs, plastics, and others. The antibiotics reported in this area include sulfonamides, lincosamides, macrolides, quinolones, chloramphenicol, tetracyclines, and lactamase inhibitors. The flame retardants detected in the PRB were organo-phosphorus flame retardants, brominated flame retardants, and chlorinated flame retardants. Pesticides found in the study region were organophosphorus pesticides and organochlorine pesticides. Triclosan (TCS), triclocarban (TCC), nonylphenol, octylphenol (OP), bisphenol A (BPA), and estrogen were often reported as phenolic compounds. Acidic pharmaceuticals reported include nonsteroidal anti-inflammatory drugs (NSAIDs), blood lipid regulators (BLRs), AEDs, etc. Plastics mainly includes microplastics, parabens, parabens metabolites (MBs), and phthalates (PAEs). While quaternary ammonium compounds (QACs), synthetic musks (SMs), and perfluoroalkyl substances (PFAS) are classified under other categories. Based on the above classification method, we conducted a detailed analysis of the collected contaminant data in the PRB.

3.2. ECs concentrations in different environmental compartments

3.2.1. Antibiotics

ECs are widely detected in water, soil, and sediment. Studies reported the concentration of ECs detected in the wastewater and organisms. Antibiotics such as macrolides, sulfonamides, and fluoroquinolones are the most detected drugs in surface water in this region. The concentration range of sulfonamides in water bodies is extensive from not detected (ND) – 3203.3 ng/L in different years, and the concentrations of tetracyclines and lactamase inhibitors are relatively low. In the aqueous phase, except for several early investigations that reported extremely high concentrations such as sulfonamides at 1152 ng/L and 1437.9 ng/L, macrolides were detected at a concentration of up to 1896 ng/L, and quinolones at a level up to 359 ng/L. On the other hand, the antibiotic monitoring results of 2020 showed concentration levels of only ND to 166 ng/L. Overall, the levels of antibiotic compounds in the PRB in southern China were lower than in northern China of the East China Sea (0.1 – 2106.1 ng/L) (Li et al., 2022a), Chaohu Lake (0 – 89,245.23 ng/L) (Zhou et al., 2022). This is most likely due to the uneven spatial distribution of antibiotic use. This has also been concluded in other studies (Zhang et al., 2015a, 2020b). In addition, the concentrations in the PRB are essentially high compared to other countries or regions, such as the Cau River in Vietnam (0.30 – 66.6 ng/L) (Ngo et al., 2021), Hanoi City (26 – 255 ng/L) (Tran et al., 2019), Persian Gulf (1.21 – 51.5 ng/L) (Kafaei et al., 2018). Worldwide, the consumption pattern of antibiotics varies from country to country. For example, Japanese formulations are dominated by macrolides and cephalosporins, followed by fluoroquinolones and penicillin (Tsutsui et al., 2018), while Vietnamese antibiotic prescriptions contain higher ratios of penicillin and cephalosporins than macrolides and fluoroquinolones (Carrique-Mas et al., 2020).

Few studies have reported antibiotic contamination in soil and sediment, and organisms. From the data collected, only one paper mentioned the concentration range of sulfonamides and quinolones in organisms from ND – 140.5 ng/L and 2.4 – 185.7 ng/L, respectively, lacking data in soil and sediment. The concentrations of antibiotics were much lower than those found in mollusks in coastal waters of the Bohai Sea (n.d. – 1575.10 ng/g) (Li et al., 2012) and slightly higher than those found in summer fish and shrimp in the Beibu Gulf of China (n.d. – 10.6 ng/g) (Zhang et al., 2018a). Compared to foreign countries, these concentrations are significantly higher than for instance in the Po delta (Italy) (up to 13.3 ng/g) (Álvarez-Muñoz et al., 2015), United States (0.3 – 8.6 ng/g) (Done and Halden, 2015), Lake Titicaca, Peru (3.6 – 8.7 ng/g) (Vilca et al., 2021), and Egypt (<LOQ – 52 ng/g) (Eissa et al., 2020).

3.2.2. Flame retardants

The concentration of flame retardants varies in different environmental media, and the content of organophosphorus flame retardants is the highest in water, up to 1790 ng/L. Soils, sediments, and organisms are more commonly polluted by brominated flame retardants, especially polybrominated diphenyl ethers (PBDE), as expected based on their environmental persistence and hydrophobicity. Flame retardants in soil and sediment were more studied. Chlorinated flame retardants were present in the highest concentration in soil and sediment, reaching 77,140 ng/g, and it did not find relevant contamination data in water. In general, the contamination concentrations in organisms are lower than in soil for organophosphorus flame retardants, chlorinated flame retardants, and brominated flame retardants, ranging from 0.2 to 11.8 ng/g, ND – 6989 ng/g, and ND – 11,000 ng/g, respectively.

The pollution level of flame retardants in the PRB is slightly higher than in most parts of the country, like the Yellow River (35.6 – 469 ng/L) (Han et al., 2021), Qinzhou Bay (in surface seawater and sediments: range from 150 to 885 ng/L and < limit of quantification (LOQ) – 32.2 ng/g, respectively) (Zhang et al., 2021a), western Taiwan Strait (in marine sediments range from 5.26 to 34.23 ng/g) (Zeng et al., 2020). It is also at a high level from a global perspective, as compared to for instance Shinhwa Lake (concentrations in water and sediment ranging from <LOQ to 15.2 ng/L and <LOQ to 4272 ng/g, respectively) (Lee and Moon, 2021), Kuzuryu River in Japan (in water and sediment ranging from 180 to 2100 ng/L and 97–7800 ng/g, respectively) (Oh et al., 2014), western Pacific (3.02 – 48.4 ng/L) (Xiao et al., 2021). The same conclusion was reached in other studies (Choi et al., 2020). FRs in organisms are more abundant in the PRB, with concentrations ranging from ND – 11,000 ng/g, with PBDE being the highest concentration, which is much higher than in Beijing (264.7 – 1973.2 ng/g) (Hou et al., 2017), mullet fish from the Tagus estuary (up to 98.4 ng/g) (Álvarez-Muñoz et al., 2015), Korean coast (6.12 – 206 ng/g) (Choi et al., 2020), in river fish from Spain (up to 2423 ng/g) (Santín et al., 2016), in fish from three river basins of Greece, Italy, and Slovenia, with

total concentrations ranging from 14.4 to 650 ng/g (Giulivo et al., 2017), Of course, there are also some cities abroad whose concentration level is comparable to PRB. For instance, in Sweden (61 – 15,000 ng/g) (Sundkvist et al., 2010), Barents Sea, and west-coast of Spitsbergen, Norway (169.8 – 14,820 ng/g) (Remberger et al., 2009). The high pollution level in the PRB may be due to surrounding municipal e – waste disposal areas.

3.2.3. Pesticides

Among all retrieved literature, only one study reported the contamination of pesticides in water in the PRB. The average level of organophosphorus pesticides in water was slightly higher than the contamination level of organochlorine pesticides. The concentration of pesticides in water in the PRB (0.2 – 1015.4 ng/L) is far lower than the pollution level in the Taihu Lake Basin (ND – 10,600 ng/L) (Wang et al., 2021) in China, Tengi River Basin in Malaysia (1.3 – 4493.1 ng/L) (Elfikrie et al., 2020), Marun River in Iran (870 – 3229 ng/L) (Rezaei Kalantary et al., 2022) and South Georgia in the USA (90 – 10,500 ng/L) (Glinski et al., 2018), but higher than in the East China Sea (2.62 – 102.07 ng/L) (Wang et al., 2022a) and in XiangJiang (2.33 – 6.40 ng/L) (Zhang et al., 2022) in China.

Data for organophosphorus pesticides in soil and sediments and in organisms are lacking. In addition, concentrations of organochlorine pesticides in organisms are significantly higher (ND – 1700 ng/g) than in soil (0 – 60 ng/g). Among all retrieved literature, only one study reported the contamination of pesticides in water in the PRB. The average level of organophosphorus pesticides in water was slightly higher than the contamination level of organochlorine pesticides. Data for organophosphorus pesticides in soil and sediments and in organisms are lacking. In addition, concentrations of organochlorine pesticides in organisms are higher (ND – 1700 ng/g) than the concentrations reported in the three media and significantly higher than in soil (0 – 60 ng/g). Although the pesticides in the soil in the PRB have a large span, the average concentration of pesticides is only 3.6 ng/g, which is similar to the level of Taihu Lake and the Bohai Sea (0.24 – 5.67 ng/g) (Hu et al., 2009), Xiangjiang (1.52–21.2) ng/g (Zhang et al., 2022) in China, and is significantly lower than in other foreign countries like the Nile River area in Egypt (18 – 180 ng/g) (Eissa et al., 2022), the central coasts of Vietnam (9.72–3730 ng/g) (Tham et al., 2019), the Ebro River Delta (up to 18×10^4 ng/L) (Barbieri et al., 2021). Pesticides in organisms in the PRB are mainly organochlorine pesticides, with concentrations ranging from ND – 1700 ng/g, which is much higher than the concentration in Huanghai (<0.014 – 5.3 ng/g) (Li et al., 2021a) and Lake Chaohu (ND – 32.6 ng/g) (Liu et al., 2016), and slightly lower than for biota in the Qiantang River (28 – 43,495 ng/g) (Zhou et al., 2008), Internationally, a slightly lower concentration level is detected in the PRB than in Peninsular Malaysia (0.5 – 2880 ng/g) (Santhi et al., 2012), Greenland (ND – 2000 ng/g) (Vorkamp et al., 2004), and the Danube Delta, Romania (ND – 4977 ng/g) (Covaci et al., 2006).

3.2.4. Phenolic compounds

Phenolic compounds, in addition to antibiotics, are the most studied ECs in the PRB. The pollution of nonylphenol and octylphenol in water is the most serious. The concentrations are 1.0–33,231 ng/L and 28.1 – 8890 ng/L, respectively. Bisphenol A (BPA) is predominantly present in soils and sediments, with a concentration of up to 10,800 ng/g, whereas the concentration of other pollutants ranges from hundreds to thousands of ng/g. There are few studies on phenolic compounds in organisms, and there is only a small amount of data on BPA at a concentration of ND – 807.4 ng/g.

Compared with other regions, concentrations of phenolic compounds in water in the PRB before 2011 (up to 33,231 ng/L) were significantly higher than the concentrations detected in other regions in China and abroad. However, after 2011 the concentrations of phenolic compounds in the PRB water decreased significantly (drop to 370 ng/L), and were lower than in Taihu Lake basin (150 – 2750 ng/L) (Wang et al., 2020), Lhasa River basin (ND – 433 ng/L in surface water and 0.044 – 8.7 ng/g in sediment) (Liu et al., 2020), and the Bengal bay in India (40 – 4460 ng/L) (Chakraborty et al., 2021). But the concentration in the soil remained high (up to 10,800 ng/g), which is much higher than in soils surrounding the Xi River in Shenyang (2720 ng/g) (Li et al., 2016a) and the Beitang Drainage River in Tianjin (ND – 299 ng/g) (Zhong et al., 2018). Apparently, concentrations of phenolic compounds are much lower abroad, like in the United States (4 – 14 ng/g) (Agency, U.S.E.P.) and in Spain (0.7 – 44.5 ng/g) (Sánchez-Brunete et al., 2009). The concentration of BPA in organisms at PRB ranges from ND – 807.4 ng/g, which is in the middle of the range worldwide. The concentration is much higher than detected for Taihu Lake (1.69 – 35.2 ng/g) (Wang et al., 2017b) and the Persian Gulf (ND – 23.85 ng/g) (Akhbarizadeh et al., 2020), and at similar levels as in the Southern Baltic (<2.0 – 836.1 ng/g) (Bodziach et al., 2021), Sacca di Goro, Italy (<0.05 – 534.1 ng/g) (Casatta et al., 2015) and Peninsular Malaysia (LOQ – 729 ng/g) (Santhi et al., 2012). Obviously, there are some areas with extremely high levels of pollution at home and abroad, such as the East China Sea (<LOQ – 19,890.5 ng/g) (Gu et al., 2016) and North American Pacific Coast estuaries (136 – 11,200 ng/g) (Diehl et al., 2012).

3.2.5. Acidic pharmaceuticals

The main monitoring data of acidic pharmaceuticals are generated in water bodies. Among several major pharmaceuticals categories, NSAIDs had the highest concentration in water (ND – 15,571 ng/L), the average concentrations reached one to two hundred ng/L, which is much higher than the concentrations reported in the Beiyun River of Beijing (7.8 – 170 ng/L) (Dai et al., 2015), and the Huangpu River (ND – 75.5 ng/L) (Wu et al., 2015) in China. In comparison, the concentration of AEDs in the PRB (0.2 – 43 ng/L) is slightly lower than the concentrations detected in Turkey (53 – 2090 ng/L) (Korkmaz et al., 2022), Brazil (up to 3500 ng/L) (Pivetta et al., 2020), European and North American waters (ND – 380 ng/L) (Peng et al., 2008a), the United Kingdom (up to 1008 ng/L) (Kasprzyk-Hordern et al., 2008).

There is only one research that mentioned it in soil and sediment (ND – 50.4), and no relevant research data was found in living organisms. Overall, the concentration of Acidic pharmaceuticals in the PRB is lower compared with foreign countries, and in recent years, the pollution situation has shown a trend of gradual reduction.

3.2.6. Plastics

Microplastics in the PRB show an extremely high level with concentrations of 379 – 19,860 items/m^3. The distribution of phthalates in the PRB ranged from a few thousand to tens of thousands of ng/L which is the most widely used plasticizers. The PAEs concentration in water was as high as 28,100 ng/L, and the water of the PRB was also slightly polluted by parabens and MBs. Which is similar with Tunisia (106,000 – 321,000 ng/L) (Jebara et al., 2021), and much high than the Bohai Sea and the Yellow Sea (453 – 5108 ng/L, and 597 – 4304 ng/L, respectively) (Zhang et al., 2018b), the West Pacific (3.02 – 48.4 ng/L) (Xiao et al., 2021), the Rhône River (France) (97 – 541 ng/L) (Schmidt et al., 2020), and in the Besos river, Spain.(<LOQ – 7957 ng/L) (Bolívar-Subirats et al., 2021).

The detected concentrations of plasticizers in soil and sediment in the PRB (108 – 38,500 ng/g) are much higher than in the East China Sea (1649.5 – 8451.5 ng/g) (Zhao et al., 2020), the Bohai Sea (1240 – 15, 800 ng/g) (Zhang et al., 2018b), the Yellow Sea (310 – 62,150 g/g) (Li et al., 2014a), as well as in other global regions such as coastal Kuwait (2150 – 15,720 ng/g) (Saeed et al., 2017), the Bay of Bengal in India (430 – 7630 ng/g) (Chakraborty et al., 2021) and the Canada-Port of False Creek (4 – 2100 ng/g) (Mackintosh et al., 2006). The abundance of microplastics in the sediments of PRB was about 80 – 9597 items/kg, which was much higher than the abundance of 1765 items/kg reported for the Yellow Sea (Wang et al., 2019), 60 – 240 items/kg in the East China Sea (Zhang et al., 2019), and 96.7 – 333.3 items/kg in the Bohai Sea (Wu et al., 2019). Compared with countries outside of China, the

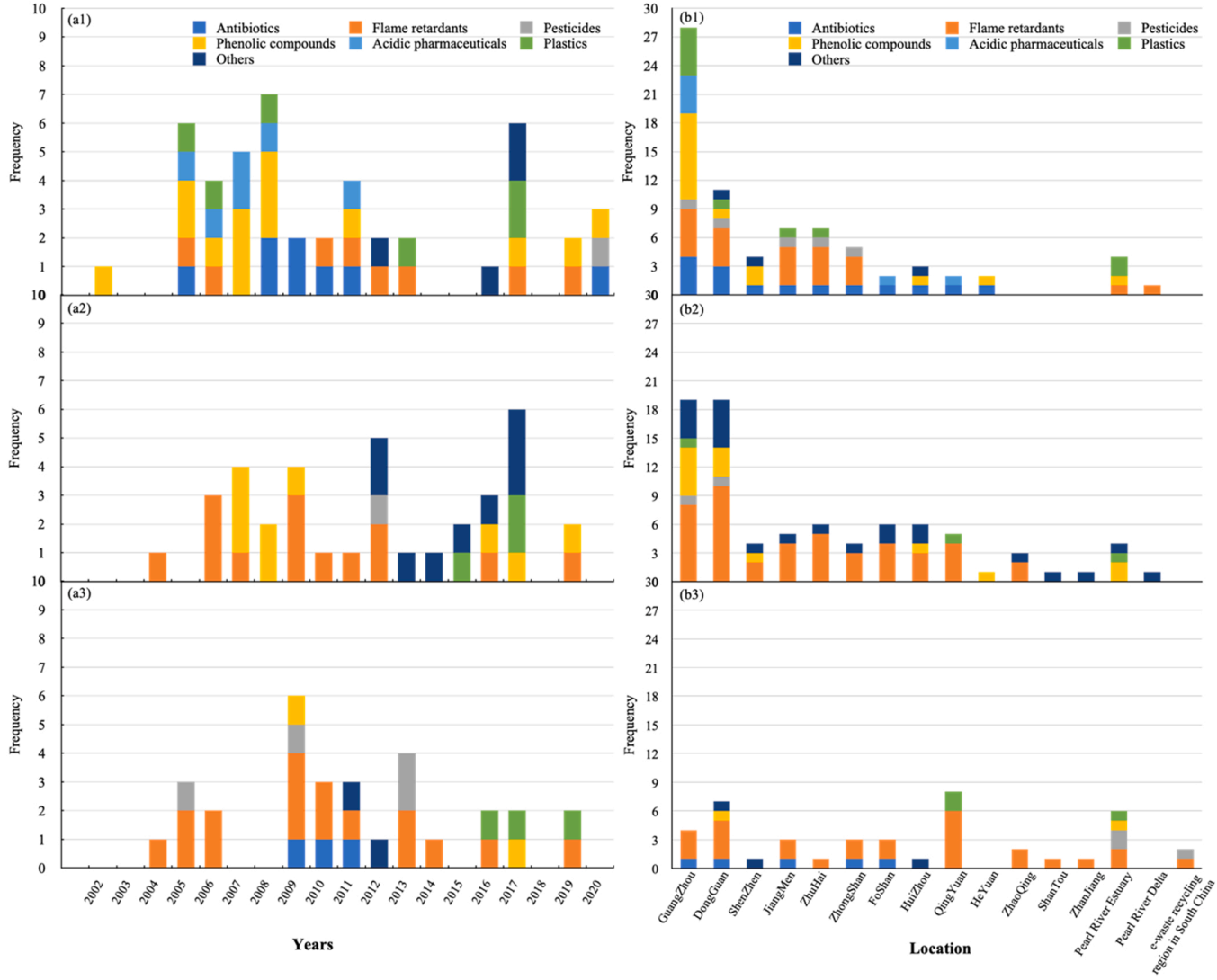

Fig. 1. Comparison of the frequency of ECs in the PRB (a) time trends, (b) in different area (1,2 and 3 means in water, soil and sediment, and organisms, respectively).

reported concentrations are close to the levels of the Mediterranean Sea with an abundance of 33 – 7989 items/kg (Constant et al., 2019) and much lower than in Denmark with 9.5×10^5 items/kg (Olesen et al., 2019), whilst the characteristics of the microplastics in water bodies are similar across these waters.

The concentration of plasticizers in organisms in PRB (4.72 – 8194 ng/g) belongs to the low – moderate level in the world as a whole. This can be illustrated for example by data on marine organisms from Hangzhou Bay, China (64–2840 ng/g) (Hu et al., 2020), on fish in Tunisian (3340 – 9700 ng/g) (Jebara et al., 2021), and on freshwater fish and marine fish in Hongkong (1660 – 3140 ng/g, 1570 – 7000 ng/g, respectively) (Cheng et al., 2013).

3.2.7. *Others*

There are many different types of pollutants in soil and sediment, include QACs, SMs, and PFAS. Among these chemicals, QACs were present at the highest concentration and they induce the most severe pollution, with a concentration range of 10 – 10,917 ng/g, which is similar to Tokyo Bay, Japan (230–11,000 ng/g) (Dai et al., 2018a), much lower than 100–110000 ng/g in Hudson River Basin, U.S (Li and Brownawell, 2010)., but higher than 4.4–2700 ng/g in Riverine, Austria (Martínez-Carballo et al., 2007).

SM contamination levels in PRB sediments (1.89 – 27.1 ng/g) were slightly higher than in the Northeast Songhua River (<0.5 – 17.5 ng/g) (Lu et al., 2015), the Hai River (1.5 – 32.3 ng/g) (Hu et al., 2011), Germany (<0.5 – 90 ng/g) (Kronimus et al., 2004), Lake Erie and Lake Ontario (3.2 – 16.0 ng/g; 0.96 ng/g) (Peck et al., 2006), but much lower than in the U.S. Upper Hudson River (72.8 – 544 ng/g) (Reiner and Kannan, 2011).

PFAS concentration levels in aqueous in the PRB ranged from 0.14 to 346.72 ng/L, which is at a similar level as other regions in China and abroad, like Tianjin (6.64–119.86 ng/L) (Li et al., 2022b), Hai River (1.74–172 ng/L) (Li et al., 2020), Taihu (205.6 ng/L and 171.9 ng/L in 2018 and 2019, respectively) (An et al., 2021), New York State, USA (27.6–94.2 ng/L) (Zhang et al., 2021b), Alabama (ND-237 ng/L) (Viticoski et al., 2022) and France (0.11–198 ng/L) (Schmidt et al., 2019). Concentration in soil and sediment (0.024–12.23 ng/g) is slightly higher than 1.49–2.66 ng/g in Dalian Bay, China (Ding et al., 2018) and 0.33–2.78 ng/g in Bohai Sea, China (Chen et al., 2016), while much lower than in Nevada, USA (ND-345.7 ng/g) (Bai and Son, 2021). PFAS concentrations detected in PRB organisms ranged from 0.4 to 1500 ng/g, which is slightly higher than that in Spain (63.8–938 ng/g) (Pignotti et al., 2017) and Czech (0.15–877 ng/g) (Hloušková et al., 2013).

To summarize, it was found that the concentrations of ECs in surface water were generally in the range from dozens to hundreds of ng/L. In

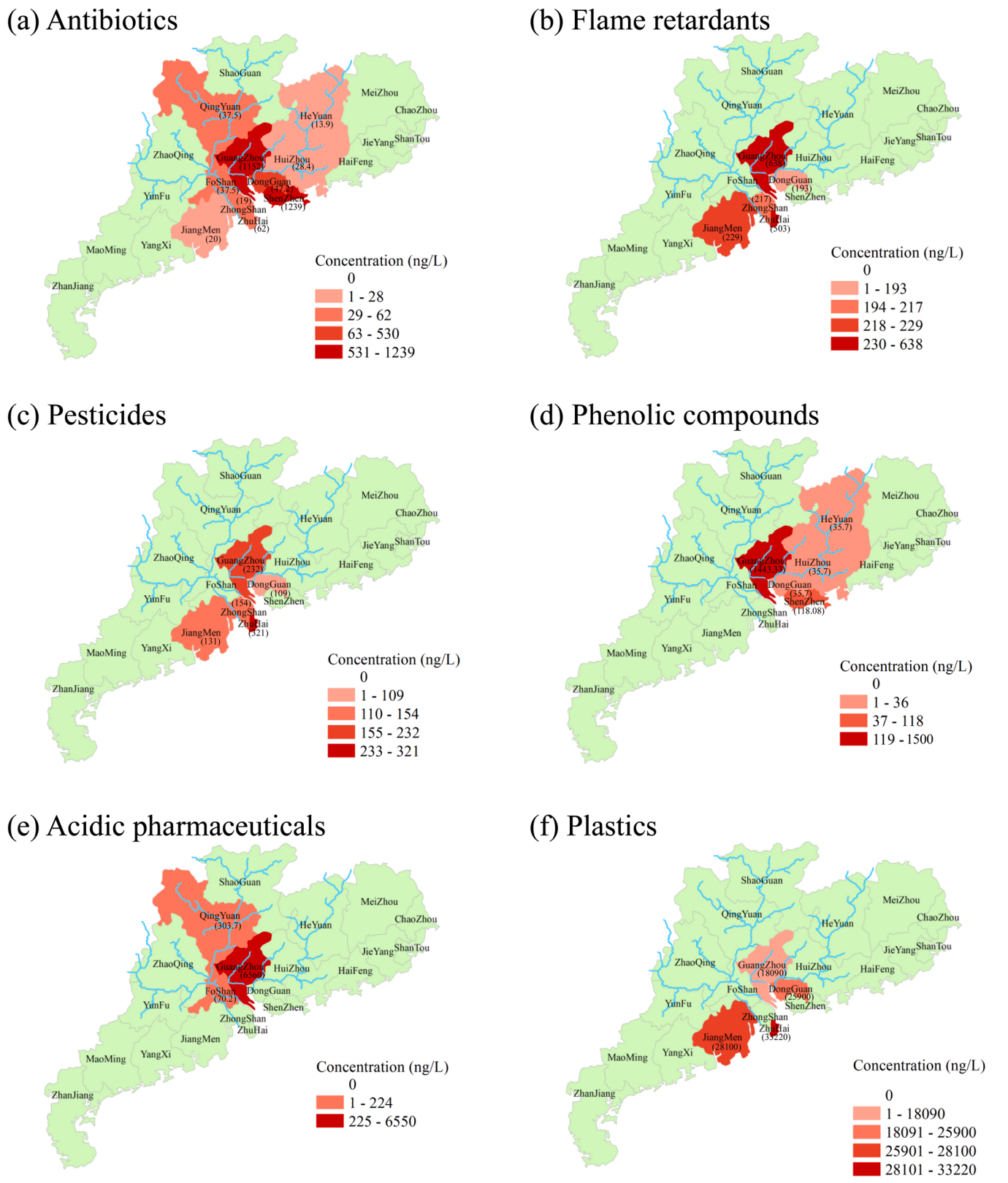

Fig. 2. Detected average concentrations of ECs (a) antibiotics, (b) flame retardants, (c) pesticides, (d) phenolic compounds, (e) acidic pharmaceuticals, (f) plastics in the PRB.

some heavily polluted areas, these concentrations even reached μg/L levels. In general, the highest concentrations of pollutants in water were phenolic compounds detected before 2011, followed by acidic pharmaceuticals, and mainly plastics after 2011. In soil and sediment, the concentration of flame retardants was significantly higher than the concentration of any other pollutant before 2011, while after 2011 mainly plastics was detected, followed by phenolic compounds and QACs. The lowest number of monitoring data were available for biota,

Table 1
Detected concentrations of antibiotics in different environmental compartments and organisms, presented as average and range.

ECs	Concentration / Abundance			Sampling year	References
	Water (ng/L)	Soil and sediment (ng/g)	Biota (ng/g)		
Sulfonamides	1152			2005	(Xu et al., 2007)
	6.16 – 3203.3			2008	(Yang et al., 2011)
	25.3 – 2027.6			2009	(Yang et al., 2011; Zhang et al., 2012b)
	11.7 – 327.6			2010	(Chen et al., 2013b)
	9 – 79			2011	(Yang et al., 2013)
			ND – 140.5	2009 – 2011	(He et al., 2016)
	ND – 82			2020	(Zhang et al., 2020b)
Lincosamides	0.12 – 15			2020	(Zhang et al., 2020c)
Macrolids	636			2005	(Xu et al., 2007)
	8.07 – 179			2008	(Yang et al., 2011)
	2.5 – 3580			2009	(Ruijie et al., 2012; Yang et al., 2011)
	13.6 – 1239			2010	(Chen et al., 2013b)
	0.2 – 11			2011	(Yang et al., 2013)
	1.4 – 166			2020	(Zhang et al., 2020c)
Quinolones	359			2005	(Xu et al., 2007)
	1.8 – 124			2009	(Yang et al., 2011)
	ND – 90.9			2010	(Chen et al., 2013b)
	15 – 304			2011	(Yang et al., 2013)
			2.4 – 185.7	2009 – 2011	(He et al., 2016)
Chloram phenicol	ND – 26.7			2010	(Chen et al., 2013b)
	187			2005	(Xu et al., 2007)
Tetracyclines	ND – 16.4 * *			2010	(Chen et al., 2013b)
Lactamase inhibitors	ND – 18.8			2010	(Chen et al., 2013b)

* * means that the environmental monitoring concentration is slightly greater than the MATC value.

and from the available information it could be concluded that primarily flame retardants were present before 2011, whilst mainly plastics was detected after 2011. From the perspective of the time dimension, the extent of pollution in the early stages of monitoring is significantly higher than in recent years. This shows that the pollution status of the PRB has improved.

At the 2009 Toxicology and Risk Assessment Conference in Cincinnati, Ohio, a session entitled "Emerging Contaminants" was held. Information on programmes and technologies related to emerging pollutants was shared. This may be one of the reasons why people beginning to take ECs seriously (Murnyak et al., 2011). A technique for dealing with ECs and reducing their consequences was developed according to European Commission (Commission, 2011). In the case of China, the Ministry of Ecology and Environmental Protection issued the Measures for the Environmental Management of New Chemical Substances in 2009, which provides environmental management of new chemical substances engaged in research, production, import and processing in China. In the same year, the National Implementation Plan for Persistent Organic Pollutants (POPs) Surveys was updated. In 2010, the technical policy for pollution prevention and control in livestock and poultry breeding proposed to strengthen the treatment of environmental pollutants such as heavy metals, antibiotics and growth hormone contained in livestock and poultry breeding wastewater.

3.3. Occurrences, spatial and temporal distribution of ECs in the PRB

The frequency of reports of ECs in the literature varies significantly over time, as shown in Fig. 1(a). The frequency of detection of ECs varies over the years. The highest reporting frequency of ECs in the environment and in organisms was observed at 2009 and 2017, 12 and 14 times, respectively. Few data on concentrations of ECs in the environment were available for 2002, 2004, 2014, 2015, and 2020. The literature reviewed did not report any ECs concentrations in 2003 and 2018.

In general, the reported ECs concentrations are more plentiful in water compared to other media, mainly concentrated during 2005 – 2008 and 2017, while the concentrations reported in soil and sediment have better time continuity and more stable study frequencies. The research of contamination data in organisms is the least compared to water, soil and sediment. There are many vacant research data for many years which result in poor data continue.

Fig. 1(b) shows the frequency of detection of ECs in water, soil and sediment and organisms in various urban areas and regions. All pollution data are obtained within Guangdong Province as upstream of the PRB there is a lack of pollution studies. It can be seen that Guangzhou (GZ) and Dongguan (DG) studied the most prominent and most significant number of pollutant species, with 36 and 31 species, respectively, with the study in the DG region focusing on the inlet river network of the tributaries of Dongjiang River. Shantou (ST), Zhanjiang (ZJ), and Zhaoqing (ZQ) have fewer types of ECs, 1, 1, and 3, respectively, and the contaminants found in the studies of other cities are basically in the range of 8 – 16 chemicals.

To analyze the spatial distribution trends of ECs in the PRB, the municipal average concentration distribution maps of significant ECs concentrations in the PRB (within Guangdong Province) were drawn (Fig. 2), and Fig. 2a–f show the spatial distribution of antibiotics, flame retardants, pesticides, phenolic compounds, acidic pharmaceuticals and flame retardants in the PRB, respectively. From Fig. 2, it can be seen that the ECs are mainly distributed in GZ, Shenzhen (SZ), DG, Foshan (FS), Zhongshan (ZS), Zhuhai (ZH), Qingyuan (QY), Heyuan (HY) and Jiangmen (JM). At the same time, other cities in the basin, such as Shaoguan (SG), ZQ, and Yunfu (YF), lack relevant research data. From Fig. 2a it can be seen that the concentration of antibiotics in SZ, GZ, and DG are much higher than in other regions, 1239 ng/L, 1152 ng/L, and 529.9 ng/L, respectively, while the detected concentrations in HY and JM are only 20 ng/L and 13.9 ng/L. Figs. 2b and 2c indicate that the monitoring of flame retardants and pesticides is mostly concentrated in GZ, ZH, JM, ZS, and DG, where the concentration of flame retardants in the water of GZ and ZH is as high as 638 ng/L and 503 ng/L respectively. The distribution of pesticides in each region is relatively uniform, with concentrations between 109 ng/L and 321 ng/L. Fig. 2d shows the distribution of phenolic compounds. It can be seen from this figure that the concentration of phenolic compounds in GZ is significantly higher than in other regions. In fact, the concentration in GZ (33,231 ng/L) is almost 930 times higher than in DG, HY and Huizhou (HZ) (35.7 ng/L). The pollution data for acidic pharmaceuticals in the region are less available, as shown in Fig. 2e, only in three cities, GZ, QY, and FS, and again, as far as acidic pharmaceuticals are concerned, the concentration measured in GZ (6550 ng/L) is significantly higher than the concentration in the other two cities (223.5 ng/L). Fig. 2f shows that plastics are also relatively evenly distributed across regions, but the overall concentration is high, with the lowest concentration in the GZ area reaching 18,090 ng/

Table 2
Detected concentrations of flame retardants (FRs) in different environmental compartments and organisms, presented as average and range.

ECs	Concentration / Abundance			Sampling year	References
	Water (ng/L)	Soil and sediment (ng/g)	Organism (ng/g)		
Organo – phosphate (OPFRs)	1.1 – 3.1			2011 – 2012	(Wang et al., 2014)
		58 – 322		2012	(Pintado-Herrera et al., 2017)
	14.9 – 1790			2013	(Li et al., 2021b; Lai et al., 2019)
			0.2 – 13	2016	(Yin-E et al., 2019)
	134 – 442	7.1 – 143		2017	(Pintado-Herrera et al., 2017; Shi et al., 2020)
	21.2 – 91	25.2 – 492	5.2 – 11.8	2019	(Chen et al., 2020; Mai, 2021)
Chlorinated flame retardants			1.4 – 9.3	2005	(Sun et al., 2015a)
		77140	255 – 6989	2006	(Ying et al., 2011)
		ND – 45.0		2009 – 2010	(A et al., 2013)
			ND – 37	2013	(Sun et al., 2015a, 2015b
		0 – 71.6		2016	(Li et al., 2021b)
		20.0 – 3420		2011	(Wu et al., 2020)
Polybrominated diphenyl ether (PBDE)		1.0 – 7434.7		2004	(Mai et al., 2005)
			0.2 – 8.2	2005	(Sun et al., 2015a)
	2.7 – 19.1			2005 – 2006	(Guan et al., 2007)
		20.7 – 1130000	52.4 – 10100	2006	(Ying et al., 2011; Shi et al., 2009; Zhang et al., 2009)
		24.0 – 64672.0		2006 – 2007	(Shi et al., 2009)
		64.5 – 212	8.1 – 620	2009	(He et al., 2012; Zhang et al., 2010b)
		3.7 – 2517		2009 – 2010	(A et al., 2013)
	0.1 – 0.2		35 – 820	2010	(He et al., 2012)
			35 – 11000	2009 – 2011	(Sun et al., 2012)
		1.3 – 206		2012	(Zhang et al., 2015b)
			0.1 – 59	2013	(Sun et al., 2015a, 2015b
Pentabromotoluene (PBT)			ND	2005	(Sun et al., 2015a)
			ND – 6.8	2009 – 2011	(Sun et al., 2012)
			ND – 5.1	2013	(Sun et al., 2015a, 2015b
Polybrominated biphenyls (PBB)			ND – 6800	2009 – 2011	(Sun et al., 2012)
Hexabromobenzene (HBB)			0.3 – 240	2009	(Zhang et al., 2010b)
Decabromodiphenyl ethane (DBDPE)		19 – 430		2006	(Zhang et al., 2009)
		2.5 – 1995		2006 – 2007	(Shi et al., 2009)
		37 – 110		2009	(He et al., 2012)
		ND – 1728		2009 – 2010	(A et al., 2013)
			ND – 230	2010	(He et al., 2012)
			ND – 130	2009 – 2011	(Sun et al., 2012)
		0.4 – 34.9		2012	(Zhang et al., 2015b)
			ND – 15	2013	(Sun et al., 2015b)
1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE)		0 – 21.9		2006 – 2007	(Shi et al., 2009)
		ND – 73.4		2009 – 2010	(A et al., 2013)
		ND – 0.8		2012	(Zhang et al., 2015b)
Tetrabromobisphenol A (TBBPA)		2.3 – 230		2006	(Zhang et al., 2009)
		ND – 8946		2006 – 2007	(Shi et al., 2009)
		ND – 82.3		2009	(He et al., 2013)
Hexabromocyclododecanes (HBCDs)			ND – 0.2	2004 – 2005	(Meng et al., 2012)
		ND – 82.3	ND – 832	2009	(He et al., 2013)
			5.9 – 518	2014	(Luo et al., 2018)

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3.4. ECs with potential risk by comparison with MATC

By comparing the actual environmental data collected by the literature with MATC (Table S4), it is found that although there are prosperous data on antibiotics in water, they are generally in the safe concentration range, and only the maximum concentration of Tetracyclines detected in 2010 exceeds the MATC value, which may be moderately risky. Flame retardants have fewer data on contamination in the water, which is a low-risk range as far as the available data is concerned. The concentration of Organochlorine pesticides in water in 2000 was much greater than the MATC, indicating that they had a very high risk to organisms in the early years, and became medium risk in 2020. Phenolic compounds and acidic pharmaceuticals are among the pollutants with an overall high risk, which may be due to their older pollution data, mostly concentrated between 2005 and 2011, especially NP, BPA, NSAIDs and BLRs. BPA detected in 2019 presents a moderate risk in water. PFAS in aquatic environments also presents a high risk to organisms, mainly due to their extremely low environmental limits, where only traces of PFAS can cause toxicity to various organisms. In addition, the concentration of TCC and OP is not a risk for the time being, but if it is not restricted and controlled by related chemicals, which is very likely to have a harmful impact on the ecological environment in the future.

4. Discussion

4.1. The abundance of ECs coupling with the industrial development in the region

As one of the most important manufacturing bases in China, the PRB is known for its electronics and related industries and as such is often reported as a high BFR polluted watershed (Chen et al., 2013a). PCBs and PBDEs are ubiquitous because of the increasing manufacturing industry (Zhang et al., 2010a). On the contrary, it has also been shown that Dachlorane (a type of flame retardant) in PRB sediments is mainly from urban rather than from industrial activities (Li et al., 2021b). In addition, the PRB is considered one of the most developed agricultural regions in China (Chen et al., 2019a). Pesticides are widely used to control pests and improve agrarian production (Barbieri et al., 2021). There is much research reporting high levels of microplastic pollution in aquaculture water of fish ponds in the PRB (Ma et al., 2020). It is commonly

Table 3
Detected concentrations of pesticides and phenolic compounds in different environmental compartments and organisms, presented as average and range.

ECs	Concentration / Abundance			Sampling year	References
	Water (ng/L)	Soil and sediment (ng/g)	Organism (ng/g)		
Pesticides					
Organochlorine pesticides	102.6 – 1015.4 * *[a]			2000	(Zhang et al., 2020c)
			5.0 – 171.2	2005	(Sun et al., 2015a)
			ND – 850	2009	(Zhang et al., 2010b)
		0 – 60.0		2012	(Pintado-Herrera et al., 2017)
			6.7 – 1700	2013	(Sun et al., 2015a, 2015b
	1.8 – 170 **			2020	(Zhang et al., 2020c)
Organophosphorus pesticide	0.2 – 172			2020	(Zhang et al., 2020c)
Phenolic compounds					
Triclosan (TCS)	31.6			2005	(Wu et al., 2007)
	35 – 1023			2005 – 2006	(Peng et al., 2008b)
	0.6 – 347			2007	(Zhao et al., 2009)
	7.7 – 217.5			2008	(Yu et al., 2011)
	<LOQ – 478	<LOQ – 1329		2007 – 2008	(Zhao et al., 2013a, 2010a
		LOQ – 656		2008 – 2009	(Zhao et al., 2013b)
		0.5 – 41.1		2016	(Peng et al., 2016)
	1.5 – 5.6	0.7 – 17.5		2019	(Chen et al., 2020)
Trichlorocarban (TCC)	<LOQ – 338[a]	<LOQ – 2633		2007 – 2008	(Zhao et al., 2013a, 2010a
	4.9 – 155.1			2008	(Yu et al., 2011)
		LOQ – 2723		2008 – 2009	(Zhao et al., 2013b)
		ND – 332		2016	(Peng et al., 2016)
Nonylphenol (NP)	36 – 33231 * *[a]			2005 – 2006	(Peng et al., 2008b)
	1.0 – 2470	144.0 – 293.0		2007	(Zhao et al., 2009; Peng et al., 2007)
	234 – 437			2011	(Weihai et al., 2014)
Octyl phenol (OP)	28.1 – 8890[a]			2007	(Zhao et al., 2009)
Bisphenol A (BPA)	6 – 881 * *[a]			2005 – 2006	(Peng et al., 2008b)
	2.2 – 1030 * *[a]	2.0 – 4.3		2007	(Zhao et al., 2009; Peng et al., 2007)
			ND – 66	2009	(He et al., 2013)
	228 – 625 * *[a]			2011	(Weihai et al., 2014)
		0.7 – 217.7		2016	(Peng et al., 2016)
	44.9 – 370 * *[a]	416 – 10800	1.4 – 807.4	2017	(Zhao et al., 2019)
	8.5 – 37.3 * *	14.0 – 86.3		2019	(Chen et al., 2020)
Estrogen	3.0 – 300			2002	(Lee et al., 2006)
	ND – 72			2005 – 2006	(Peng et al., 2008b)
	ND – 82.5			2007	(Zhao et al., 2009)
	33.3 – 1671.5			2008	(Yu et al., 2011)
	ND – 14			2011	(Weihai et al., 2014)
	ND – 138.9			2020	(Tang et al., 2021)

* * means that the environmental monitoring concentration is slightly higher than the MATC value
* ** means that the environmental monitoring concentration is much higher than the MATC value
[a] means that the environmental monitoring concentration is slightly lower than the MATC value

believed that mariculture wastewater is a typical non – point source of antibiotic contamination in natural water (Wang et al., 2022b), which would be one of the reasons for the observed high level of antibiotic contamination of PRB. In conclusion, environmental pollution cannot be ignored with the development of industry, agriculture and urbanization in the PRB.

4.2. The potential risk of mixture toxicity in the region

Co – exposure of ECs and other environmental contaminants (e.g., heavy metals) may lead to different mixture effects such as additive, synergistic, antagonistic or even other complex reactions including altered toxicokinetic/toxicodynamic that vary depending on the properties of individual components, environmental exposure conditions, and biological systems. A combination of sulfadiazine and methoxybenzyl aminopyrimidine will for instance lead to oxidative stress and hepatotoxicity in gilthead sea bream Sparus aurata L (Varó et al., 2013). Other findings suggest that fluoroquinolone combined with tetracycline leads to development toxicity, cardiotoxicity, immunotoxicity and disordered locomotion behavior (neurotoxicity) in antagonistic action (Li et al., 2016b; Zhang et al., 2016). All binary mixtures of ampicillin, amoxicillin, cephalothin, ciprofloxacin, gentamycin and vancomycin are reported to exert synergistic effects to algae growth (Magdaleno et al., 2015). Secondly, the toxicity of antibiotics can be altered by traditional pollutants such as heavy metals (Zhang et al., 2012a).

Actually, many research scholars have assessed contaminants individually without considering the possibility of combined or synergistic effects. Multiple contaminants are however always coexisting in the real environment and the toxicity of contaminants is inevitably influenced by various environmental conditions such as temperature and pH, and in addition a large number of factors may affect the toxicity of contaminant mixtures, which makes the risk assessment of mixed contaminants a complex task. There is still a long way to go to fully understand the toxicological effects of mixtures of ECs in the environment, and therefore mixture toxicology should receive more attention in future studies.

4.3. Call for continuous effort in monitoring ECs concentration

Severe contamination of ECs can cause irreversible damage to the entire ecosystem, and in order to better manage ECs in the environment, their origin, their interaction with environmental properties, and their transport behavior must be understood. The most common method of investigation to determine such phenomena is through in situ monitoring or surveillance programs. The observed spatial patterns and relationships with environmental factors further enhances the overall understanding of the distribution and behavior of these contaminants in urban watersheds. This can in turn allow for specific countermeasures and directions for subsequent ecological environmental protection efforts, as well as the assessment of the actual situation of the ecological environment. This assists decision makers in developing effective

Table 4
Detected concentrations of acidic pharmaceuticals, plasticizers, and others in different environmental compartments and organisms, presented as average (median) and range.

ECs	Concentration / Abundance			Sampling year	References
	Water (ng/L)	Soil and sediment (ng/g)	Organism (ng/g)		
Acidic pharmaceuticals					
Nonsteroidal anti-inflammatory drugs	9 – 6560 * **			2005 – 2006	(Peng et al., 2008b)
(NSAIDs)	ND – 777.4 * **			2007	(Zhao et al., 2009)
	148.6 – 15,571 * **			2007 – 2008	(Zhao et al., 2010b)
	27 – 339 * **			2011	(Yang et al., 2013)
Blood lipid regulators (BLRs)	ND – 248 * **			2005 – 2006	(Peng et al., 2008b)
	ND – 17.4 * *			2007	(Zhao et al., 2009)
	7.3 – 37 * *			2007 – 2008	(Zhao et al., 2010b)
Antiepileptic drugs (AEDs)	17.9 – 43 * *			2007 – 2008	(Zhao et al., 2010b)
	0.2 – 10			2011	(Yang et al., 2013)
Other acidic pharmaceuticals	ND – 824.9			2007	(Zhao et al., 2009)
	ND – 107			2011	(Yang et al., 2013)
		ND – 50.4		2016	(Peng et al., 2016)
Plastics					
Microplastics		178 – 544 items/m^3		2015	(Wang et al., 2017c)
	379 – 19,860 items/m^3	80 – 9597 items/kg		2017	(Lin et al., 2018; Yan et al., 2019)
Parabens	ND – 4204			2005 – 2006	(Peng et al., 2008b)
	2.4 – 180.5			2008	(Yu et al., 2011)
		108 – 292	4.7 – 19.6	2017	(Zhao et al., 2019)
Paraben metabolites (MBs)	57.3 – 522.5	4900 – 38,500	292.2 – 8194.5	2017	(Zhao et al., 2019)
Legacy phthalates (LPs)			132 – 1286	2016	(Yin-E et al., 2019)
			1402.2 – 2209.3	2019	(Mai, 2021)
Alternative plasticizers (Aps)			19 – 59	2016	(Yin-E et al., 2019)
Phthalate esters (PAEs)	500 – 28,100			2013	(Li)
Others					
Quaternary ammonium compounds (QACs)		10 – 10,917		2012	(Pintado-Herrera et al., 2017)
		64 – 260		2013	(Dai et al., 2018b)
		49 – 7800		2014	(Li et al., 2014b)
Synthetic musks (SMs)		1.89 – 27.1		2017	(Huang et al., 2016)
Perfluoroalkyl substances			0.4–1500	2011–2012	(Pan et al., 2014a)
(PFAS)	0.14–346.72 * **	0.10–12.23		2012	(Pan et al., 2014b)
		0.024–0.181		2017	(Chen et al., 2019b)

* * means that the environmental monitoring concentration is slightly higher than the MATC value
* ** means that the environmental monitoring concentration is much higher than the MATC value

strategies to mitigate the potential impacts of these chemicals on the environment and on human health. At the same time, the monitoring of environmental organic pollutants can also provide powerful data support for scientific research and can provide data guarantee for technology research and development for relevant scientific research departments, thus helping China's ecological environmental protection to continue to develop and improve the technology level.

The research method applied in this paper was to search the relevant literature by keywords and collect data from the literature. The literature search was extended by intensive reading, and included only experimental data collected in the WOS database. Therefore, some omissions of pollution data cannot be excluded. This may lead to bias in the assessment. In addition, the lack of relevant pollution data for some years and the apparent unevenness of contaminant contamination data in water, soil, and organisms are detrimental to the systematic assessment of the pollution situation in the regions. A regular monitoring program can better guide the understanding of ECs present in the environmental compartments, and help to understand the transport of ECs and the potential risk of ECs in the environment and in humans. Therefore, it is imperative for continuous and systematic monitoring of the occurrence and detection of ECs in PRBs to develop a better assessment process. The effort will provide proper data to support the decision-making departments and guide the regulation of ECs in the long term.

4.4. Current detection methods and instruments

According to the review, the most commonly used detection method for ECs is a combination of chromatographic techniques and mass spectrometry. It mainly combines the separation of chromatography and the qualitative ability of mass spectrometry, and chromatography can be divided into gas chromatography (GC-MS) and liquid chromatography-chromatography (LC-MS) (McClellan, Halden, 2010; Gabet-Giraud et al., 2010).

GC is the mainstream technology for separating volatile and semi-volatile organic pollutants in various environmental samples, with the advantages of high separation efficiency, good sensitivity and large peak capacity. In GC-MS technology, EI sources can be used to analyze a variety of compounds, but due to the relatively unstable chemical structure of some ECs, soft ionization techniques such as electron capture negative chemical ionization (ECNI) are generally recommended for the analysis of compounds such as DBDPE (Badea et al., 2020). In this study, it was found that many types of ECs use GC-MS detection technology, in particular FR (Li et al., 2021b; Lai et al., 2019; He et al., 2012; Ying et al., 2011; Mai et al., 2005) , as well as TCS (Wu et al., 2007) , phenolic compounds (Zhao et al., 2009), and so on.

For some ECs with large molecular weight, low volatilization, or poor thermal stability, LC technology can be used for separation. Since 2004, UHPLC has become a commonly used separation technology. This technique enables fast separation of a wide range of analytes with higher resolution by using small inner diameter (<2 mm) columns (Rodriguez-Aller et al., 2013), It also provides higher sensitivity and better separation for the analysis of new contaminants in complex samples. ESI technology has the advantages of wide application and high sensitivity, and can be used for the analysis of high molecular weight, volatile and thermally unstable compounds, and is one of the most widely used ionization techniques for the analysis of ECs using LC-MS technology. Most antibiotics are LC-MS technology (Xu et al., 2007; Zhang et al., 2020c; Ruijie et al., 2012; Yang et al., 2011) , Some studies on TCS (Zhao et al., 2013a, 2010a、QAC (Pintado-Herrera et al., 2017; Dai

et al., 2018b) also apply this technique.

However, this approach is not suitable for real-time and on-site monitoring applications because it requires expensive instrumentation, specialized operators, and complex and lengthy sample preparation. In addition to chromatography-based techniques, other detection methods, such as optical and electrochemical techniques, have been extensively explored (Ryu et al., 2021). Inconsistencies and irregularities in measurement methods also led to differences in researchers' findings. It is necessary to establish a standardized sampling, analytical and testing method as soon as possible, which is important to ensure the accuracy of the research results and to develop effective standards for managing emission systems. Tables 1–4.

5. Conclusions

This study analyzed the distribution characteristics of the ECs in time and space. Few pollution data related to ECs in recent years but concentrated in the years 2005 – 2011 indicates a lack of continuous monitoring studies. Analysis of the pollution data mentioned in the literature reveals that some pollutants had extremely high concentrations in the early years and decreased significantly in recent years. We speculate that there are two reasons for this: with the attention paid to environmental safety in recent years, the use of some polluting chemicals has been reduced. On the other hand, the difference between sampling locations and monitoring methods may also be the reason. In addition, we found that people pay extra attention to different kinds of pollutants in various media. Increasing attention is paid to antibiotics, phenolic compounds, and acidic pharmaceuticals in water, and the primary studies in living organisms deal with flame retardants and plasticizers. In soil, on the other hand, phenolic compounds receive most attention. These differences are likely to be related to the difference in characteristics of the different pollutants and their migration and transformation pathways.

This study provides a baseline database for determining future pollutant trends and the impact of pollutant management strategies. It is recommended that local governments or watershed management departments establish a priority list of ECs that are widely distributed in the region by considering the environmental and health risks of ECs. Comprehensively and continuously monitoring the concentrations of those ECs in the region will help to assess the health and environmental risks of these ECs and will provide data to support source tracing and to fulfill regulation purposes in the region.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2023.114918.

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