Ecological risk assessment of bisphenol A in surface waters of China based on both traditional and reproductive endpoints

Lei Guo a, Zhengyan Li b,*, Pei Gao c, Hong Hu b, Mark Gibson d

a College of Environmental Science and Engineering, Ocean University of China, Qingdao 266100, PR China
b Key Laboratory of Marine Environmental Science and Ecology, Ministry of Education, Qingdao 266100, PR China
Shandong Academy of Environmental Science, Jinan 250013, PR China
d Department of Process Engineering and Applied Science, Dalhousie University, Halifax, Nova Scotia B3J 2X4, Canada

Article info
Article history:
Received 8 January 2015
Received in revised form 29 May 2015
Accepted 1 June 2015

Keywords:
Water quality criteria
Ecological risk assessment
Species sensitivity distribution
Bisphenol A
Endocrine disruptor

Abstract
Bisphenol A (BPA) occurs widely in natural waters with both traditional and reproductive toxicity to various aquatic species. The water quality criteria (WQC), however, have not been established in China, which hinders the ecological risk assessment for the pollutant. This study therefore aims to derive the water quality criteria for BPA based on both acute and chronic toxicity endpoints and to assess the ecological risk in surface waters of China. A total of 15 acute toxicity values tested with aquatic species resident in China were found in published literature, which were simulated with the species sensitivity distribution (SSD) model for the derivation of criterion maximum concentration (CMC). 18 chronic toxicity values with traditional endpoints were simulated for the derivation of traditional criterion continuous concentration (CCC) and 12 chronic toxicity values with reproductive endpoints were for reproductive CCC. Based on the derived WQC, the ecological risk of BPA in surface waters of China was assessed with risk quotient (RQ) method. The results showed that the CMC, traditional CCC and reproductive CCC were 1518 l g/L1, 2.19 l g/L1 and 0.86 l g/L1, respectively. The acute risk of BPA was negligible with RQ values much lower than 0.1. The chronic risk was however much higher with RQ values of between 0.01–3.76 and 0.03–9.57 based on traditional and reproductive CCC, respectively. The chronic RQ values on reproductive endpoints were about threefold as high as those on traditional endpoints, indicating that ecological risk assessment based on traditional effects may not guarantee the safety of aquatic biota.

1. Introduction
Bisphenol A (BPA) is a synthetic chemical primarily used to produce polycarbonate plastics and epoxy resins for various products such as reusable bottles, sports equipment, medical devices and protective coatings (Staples et al., 1998; EC, 2003). Commercial production of BPA began in the 1950s with large-scale uses for polycarbonate plastic and epoxy resins and ever since has grown worldwide with continued growth in the uses for these materials (http://www.bisphenol-a.org). In 2012, the production of BPA was approximately 6.4 million tons globally and the consumption was approximately 5 million tons, of which, China contributes 10% and 18% respectively according to the statistics by China Petroleum and Chemical Industry Federation (CPCIF, http://www.cpcia.org.cn). Despite its fast degradation in aerobic environment (Staples et al., 1998; Kang and Kondo, 2002), BPA is regularly detected in aquatic ecosystems at trace level concentrations (ng L−1 to μg L−1) throughout the world with main sources of wastewater effluent, landfill leakage and BPA-based product migration (Kang et al., 2007). Because of its ubiquitous occurrence...
in natural environment, BPA has gained wide concern from both scientific and management sectors. Numerous toxicity tests demonstrated that BPA acts as both a traditional toxicant and an endocrine disruptor. As a traditional toxicant, it causes harmful effects including carcinogenesis and teratogenesis to various species at environmentally unrealistic high dosages of mg L\(^{-1}\) (Iwamuro et al., 2003; Sone et al., 2004). Whereas as an endocrine disruptor with estrogenic and/or anti-androgenic activity (Colborn et al., 1993; Gillesby and Zacharewski, 1998), BPA affects the reproductive function at dosages of 10–1000 µg L\(^{-1}\), and even provokes molecular and biochemical biomarker response such as vitellogenin induction at 1 µg L\(^{-1}\) in vertebrates (Keiter et al., 2012). Consequently, the wide distribution of BPA in natural environment and the associated adverse effects on wildlife necessitate the assessment of ecological risks in aquatic ecosystems.

Referring to the generic framework and guidelines of the US EPA, ecological risk assessment (ERA) is defined as a process that evaluates the likelihood of adverse ecological effects on ecosystems exposed to one or more stressors (US EPA, 1998). In this process, the first step is to derive the “predicted no-effect concentration” (PNEC), at which no harmful effects on the environment are expected. PNEC values are then compared with the “predicted environmental concentration” (PEC) to calculate the “risk quotient” (RQ = PEC/PNEC), which is used as a measurement of the ecological risk (US EPA, 1998). PEC represents the exposure of the ecosystem to a chemical, which can be achieved either by model prediction or through environmental monitoring. PNEC, also referred to as water quality criteria (WQC), represents the sensitivity of the ecosystem to this chemical. PNEC is usually obtained through two methods, the assessment factor (AF) and the statistical extrapolation based on species sensitivity distributions (SSD) (ECB, 2003). The AF approach is simple to use and is feasible when toxicity data is limited. But the PNEC estimates with this method show great uncertainty since they are solely dependent on the minimum toxicity value and a certain factor. The SSD method provides more reliable and reasonable statistics since the PNEC estimates are based on an established distribution of a full toxicity data set (Lei et al., 2012). The SSD methodology is therefore increasingly being used in ecological risk assessment procedures (Wheeler et al., 2002; Hickey et al., 2009).

WQC are the maximum concentrations of a pollutant acceptable in aquatic environments that have no obvious effects on aquatic organisms and their functions (Yang et al., 2014). The US EPA proposed two-number numerical aquatic life criteria: criterion maximum concentration (CMC) and criterion continuous concentration (CCC) (Mount et al., 1985). The CMC is associated with acute toxic effects of a pollutant on aquatic organisms experiencing short-term exposure and the CCC is related to chronic toxic effects with long-term exposure. The two-number numerical criteria reflect toxicological and practical realities more accurately than traditional one number criterion. It allows the concentration of a pollutant in a body of water above the CCC without causing an unacceptable effect in some situations (e.g. short-term exposure) and avoids “overprotection” problem. Consequently the two-number numerical criteria system is now widely adopted (Yang et al., 2012; Shuhaimi-Othman et al., 2013).

Concerning toxicological effects of pollutants, survival, development, growth and reproduction are traditional measurement endpoints at individual or population level, which can be used to derive PNEC for a traditional toxicant. BPA is however an endocrine disruptor, which can affect reproductive system at lower concentrations than non-reproductive ones. PNEC derived with traditional endpoints may therefore not be protective of the reproductive activities of biota.

This study therefore aims to build SSD models for the determination of water quality criteria for BPA based on both reproductive and traditional toxicity endpoints tested with resident Chinese species. WQC derived with non-reproductive endpoints was used to make a comparison. With the derived WQC, ecological risks of BPA in surface waters of China could be assessed.

2. Materials and methods

2.1. Toxicity data screening

The toxicity data of BPA used in this study were collected from published literature and the US EPA ECOTOX database (http://cf- pub.epa.gov/ecotox/). The quality of data was evaluated with regard to reliability, adequacy and relevance (Klimisch et al., 1997). To minimize the effects of geographic difference, only the toxicity data tested with resident Chinese species, including native species and introduced species with a wide distribution in the country, were selected in this study.

The test duration for acute toxicity data was 96 h for vertebrates and algae and 48 h for invertebrates. The toxicological endpoints were LC\(_{50}\) or EC\(_{50}\) (median lethal or effect concentration) for algal growth or animal survival. The test duration for chronic toxicity data was \(\geq 14\) days, \(\geq 7\) days and \(\geq 3\) days for vertebrates, invertebrates and algae, respectively. The toxicological endpoints were NOECs (no observed effect concentrations) for growth, development, survival or reproduction. When NOEC was not available, EC\(_{10}\) (10% effect concentration) or half of LOEC (lowest observed effect concentration) was used. If there were several NOEC values for the same species based on different endpoints, the most sensitive one was selected. The geometric mean was calculated in case of multiple data on the same species and endpoint. The chronic toxicity data were further divided into two categories: reproductive effects and non-reproductive effects. Detailed information of the toxicity data is listed in Supplementary Material S1-2.

It is notable that only endpoints of demographic importance such as survival, growth and fecundity were accepted here. These endpoints are predictive of population or ecosystem level effects (Ankley et al., 1998). For other molecular and biochemical endpoints such as vitellogenin induction, which are more sensitive with useful information on possible mechanisms of action, were excluded in this study. These endpoints do not necessarily indicate adverse effect at population or ecosystem level (Liu et al., 2005).

2.2. The construction of species sensitivity distribution models

The species sensitivity distribution (SSD) is widely used in the development of WQC (Anzec, 2000). This methodology assumes that the acceptable effect level (sensitivity) of different species in an ecosystem follows a probability function (Dowse et al., 2013). The acceptable effect level for all biological species can be estimated based on the assumption that a limited number of tested species is a random sample of the whole biological system (Van der Hooven, 2004). The SSD model is generally constructed by fitting cumulative probability distribution of a full set of toxicity data. The hazardous concentration (HC\(_{p}\)), the concentration expected to be hazardous for \(p\%\) of all species in an ecosystem, can be obtained by cutting off the \(p\)th percentile of the distribution (Kooijman, 1987; Wheeler et al., 2002). Currently, HC\(_{p}\) (hazardous concentration for \(p\%\) of species) is commonly used to calculate PNEC by being divided with an appropriate assessment factor (AF, usually 1–5). The choice of AF depends on further uncertainties identified, including gap between laboratory and field system, number of species tested and model goodness of fit.

Although many distributions have been used to generate SSD (Aldenberg and Jaworska, 2000; Hose and Van den Brink, 2004), the log-normal distribution recommended by TGD of EU is most
commonly used because of the available methodology for in depth analyses of various uncertainties. In this study, log-normal distribution was adopted for fitting the toxicity data, including acute toxicity data, chronic traditional toxicity data, chronic reproductive toxicity data and chronic non-reproductive toxicity data. The data was fitted with the software ETX 2.0 (exploited by RIVM) (Van Vlaardingen et al., 2004). The model goodness of fit was evaluated by the Anderson-Darling test. To calculate the PNEC values, the AF was set as 2 for acute and chronic traditional toxicity data and 3 for chronic reproductive and chronic non-reproductive toxicity data.

2.3. Characterization of the ecological risk

The ecological risk of BPA was assessed on the basis of risk quotient (RQ = PEC/PNEC). Due to a shortage of detailed geographic distribution data for the usage and discharge amount of BPA in China, the measured environmental concentration (MEC) which can be obtained by actual field measurements (monitoring data) was used to represent the exposure condition. The maximum concentration in an area was used to calculate the acute risk quotient, whereas the median concentration was for chronic risk quotient. Considering possible interaction with other endocrine disruptors, a risk quotient of >0.3 was regarded as high risk (Warren-Hicks and Cardwell, 1996).

3. Results and discussion

3.1. Toxicity data collected

The acute toxicity data of BPA covered 15 species and 13 families (Supplementary Material S1). The LC50 values ranged from 2240 µg L-1 for mollusc species Marisa cornuarietis to 17,930 µg L-1 for fish species Xiphophorus helleri with a median of 7753 µg L-1. The chronic traditional toxicity data covered 18 species and 14 families (Supplementary Material S2). The NOEC values ranged from 1.75 µg L-1 for fish Salmo trutta to 3460 µg L-1 for another fish Oncorhynchus mykiss with a median of 128.5 µg L-1. The chronic reproductive toxicity data covered 8 invertebrate and 4 fish species. The NOEC values ranged from 1.75 µg L-1 for fish S. trutta to 1800 µg L-1 for rotifer Brachionus calyciflorus with a median of 153.5 µg L-1. The chronic non-reproductive toxicity data covered 4 invertebrate, 5 fish and one algal species. The NOEC values ranged from 10 µg L-1 for amphibian Rana temporaria to 3460 µg L-1 for fish O. mykiss with a median of 173.5 µg L-1. The reproductive endpoints were therefore generally more sensitive than non-reproductive ones.

3.2. Water quality criteria for BPA

Based on the toxicity data (Supplementary Material S1-2), the SSD models were constructed and the simulated curves were shown in Figs. 1 and 2. The HC5 value for acute toxicity endpoints was calculated as 3036 µg L-1. The HC5 for chronic toxicity endpoints was calculated as 4.38 µg L-1, 2.59 µg L-1 and 7.74 µg L-1 for traditional, reproductive and non-reproductive endpoints respectively (Supplementary Material S3). Based on the HC5 values, the PNEC could be derived, which can be recommended as WQC for BPA to protect the aquatic species. The CMC is derived as 1518 µg L-1 whereas the CCC is 2.19 µg L-1 and 0.86 µg L-1 for traditional and reproductive endpoints, respectively.

The difference in SSD models between reproductive and non-reproductive endpoints was shown in Fig. 2. As expected, the SSD curve of the reproductive endpoints shifted to the left of that of the non-reproductive ones. The resulting HC5 value for reproductive endpoints was only about one third of that for non-reproductive endpoints, indicating the high sensitivity of reproductive effects.

The traditional CCC derived in this study was 2.19 µg L-1, which was about three times of the reproductive CCC. This traditional CCC could not provide sufficient protection for species such as S. trutta and M. cornuarietis because the NOEC value of these species for reproductive activities (1.75 µg L-1 and 2.10 µg L-1, respectively) were lower than this CCC. Similar phenomenon was also observed for other endocrine disruptors. For example, the PNEC of EE2 derived with reproductive effects was about 100 times lower than that with traditional effects (Caldwell et al., 2008).
To date, there have been several reports on PNEC derivation and risk assessment for BPA in the world. In the European Union (EC, 2008), a PNEC of $1.5 \, \mu g \, L^{-1}$ was derived based on European species, which is proximate to the corresponding CCC of $2.19 \, \mu g \, L^{-1}$ in this study. The geographic distribution of species, therefore, does not seem a significant factor for PNEC derivation. The selection of toxicological endpoints was however a critical factor (Dobbs et al., 1994). Crain et al. derived a PNEC of $0.03 \, \mu g \, L^{-1}$ based on chronic physiological endpoints including biochemical biomarker response (Crain et al., 2007). This value was much lower than that in this study or in the EU report. There however exist disputes concerning whether the sensitive sub-individual toxicity data should be included in the derivation of PNEC. The direct link between these response and ecosystem health is sometimes unclear (Lin et al., 2005; Oehlmann et al., 2008).

### 3.3. Ecological risk assessment of BPA in surface waters of China

BPA was widely reported in surface waters of China (Supplementary Material S4). The concentrations ranged from non-detected to $27.4 \, \mu g \, L^{-1}$, which were comparable to those reported in other countries (Huang et al., 2012).

The acute RQ value ranged from $10^{-4}$ to $10^{-2}$ in surface waters of China, indicating negligible acute risk to aquatic organisms in short-term exposure condition. For chronic risk, the RQ value regarding traditional endpoints (RQt) ranged from 0.01 to 3.76 with an average of 0.56 and the RQ value regarding reproductive endpoints (RQr) ranged from 0.03 to 9.57 with an average of 1.42 (Fig. 3). High risk with an RQ value of >0.3 was recorded in 42% of the reported area. The maximum RQ values were recorded in two inland rivers: Weihe River (tributary of the Yellow River, RQt = 3.8, RQr = 9.6) and Neijiang River (tributary of the Yangtze River, RQt = 1.4, RQr = 3.7). These rivers received superfluous discharge of industrial effluents and landfill leakages and consequently the resident species showed higher percentage of females and smaller egg size (Sun, 2008; Guo et al., 2013). The minimum RQ values occurred in urban residential riverine waters with limited industrial discharges. This distribution pattern indicates the major source of BPA originating from industrial sectors, which is consistent with previous studies (Fürhacker et al., 2000).

### 4. Conclusions

Due to the wide usage and its associated adverse effects on various species, BPA has aroused much environmental concern. The lack of water quality criteria for BPA in China hinders the risk assessment for the pollutant. This study adopted the SSD methodology to derive water quality criteria for BPA. Both acute and chronic toxicity data on reproductive and traditional endpoints for BPA tested with resident Chinese species were screened and the SSD models were established. Based on model simulation, the CMC, traditional CCC and reproductive CCC were derived as $1518 \, \mu g \, L^{-1}$, $2.19 \, \mu g \, L^{-1}$ and $0.86 \, \mu g \, L^{-1}$, respectively. With the derived criteria, the ecological risk in surface waters of China was assessed with RQ methodology. High risk was recorded in 42% of the studied area. The chronic RQ values on reproductive
endpoints are about threefold as large as those on traditional endpoints. For endocrine disruptors such as BPA, the ecological risk assessment based on reproductive endpoints therefore better protects the safety of aquatic species.

Conflict of Interest

The authors declare that there is no conflict of interest.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (No. 41476090) and the Chinese Major Science and Technology Program for Water Pollution Control and Treatment (No. 2013ZX07202-007).

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.chemosphere.2015.06.001.

References


