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Probabilistic ecological risk assessment for three chlorophenols in surface waters of China

Liqun Xing[1], Hongling Liu[1], John P. Giesy[1,2,3,4], Xiaowei Zhang[1], Hongxia Yu[1,*]

1. State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing University, Nanjing 210046, China. E-mail: xingliqun1821@126.com
2. Department of Veterinary Biomedical Sciences and Toxicology Centre, University of Saskatchewan, Saskatoon, Saskatchewan, Canada
3. Department of Biology & Chemistry, City University of Hong Kong, Kowloon, Hong Kong, SAR, China
4. Zoology Department, College of Science, King Saud University, P. O. Box 2455, Riyadh 11451, Saudi Arabia

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Abstract

Individual and combined assessment of risks of adverse effects to aquatic ecosystems of three chlorophenols (CPs), including 2,4-dichlorophenol (2,4-DCP), 2,4,6-trichlorophenol (2,4,6-TCP) and pentachlorophenol (PCP), were conducted. A probabilistic approach based on the concentrations of CPs in surface waters of China was used to determine the likelihood of adverse effects. The potential risk of CPs in surface waters of China was determined to be of concern, especially PCP and mixtures of CPs. The risks of adverse effects were examined as the joint probabilities of exposure and response. The joint probability for PCP was 0.271 in the worst case and 0.111 in the median case, respectively. Based on the cumulative probability, 5% of aquatic organisms included in the assessment would be affected 21.36% of the time in the worst case and 5.99% of the time in median case, respectively. For the mixtures of CPs, the joint probability were 0.171 in the worst case and 0.503 in median case, respectively and 5% of species would be affected 49.83% of the time for the worst case and 12.72% in the median case, respectively. Risks of effects of the individual CPs, 2,4-DCP and 2,4,6-TCP were deemed to be acceptable with a overlapping probability of < 0.1 with 5% of species being affected less than 4% of the time.

Key words: probabilistic risk assessment; chlorophenols; surface water; joint probability curve

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Introduction

Chlorophenols (CPs) are intermediate products of the coal and petroleum refining industries. They are used as raw materials to further synthesize pharmaceutical products, pesticides, solvents, textile additives and specialty chemicals. Therefore, a relatively large amount of CPs has the potential to reach aquatic environments. CPs are frequently found in aquatic environments (Heemken et al., 2001; Gao et al., 2008; Zhong et al., 2010). CPs have received worldwide attention due to their toxicity to aquatic life, resistance to degradation, and potential to bioaccumulate (Davì and Gnudi, 1999; Ge et al., 2007). Among the CPs, 2,4-dichlorophenol (2,4-DCP), 2,4,6-trichlorophenol (2,4,6-TCP) and pentachlorophenol (PCP) are recognized as priority pollutants of aquatic environments in the United States and China (Xia and Zhang, 1990; Zhou et al., 1990; USEPA, 1991). While concentrations of CPs have been reported for some surface waters of China (Zhang et al., 2001; Chen et al., 2005; Gao et al., 2008), there have been few studies of the risks of these compounds to aquatic organisms. CPs can exist singly or together (Davì and Gnudi, 1999; Bolz et al., 2001; Gao et al., 2008). Therefore, it is necessary to assess not only the risks of individual CPs but also of combined exposure to 2,4-DCP, 2,4,6-TCP and PCP.

Ecological risk assessment (ERA) is the process of analyzing and evaluating the possibility of adverse ecological effects caused by environmental pollutants (USEPA, 1998). In recent years, ERA has been evolving and more probabilistic approaches are being applied (Solomon et al., 2000; Chen, 2005; Wang et al., 2008, 2009; Iwai and Noller, 2010). Methods of ERA include the conventional Hazard Quotient (HQ) approach, which compares point estimate of exposure and effect, and the probabilistic approach, which determines the probability of a concentration exceeding some probability of effect. In probabilistic risk assessment (PRA), the overlapping area or joint probabilities are calculated (Solomon et al., 2000; Wang et al., 2002, 2009). The HQ method is suitable for preliminary screening-stage risk assessment, while PRA method is used to more explicitly report probabilities that can fully address the uncertainty and stochastic properties of the exposure and effects.

The objective of the present study was to assess the probabilities of the individual CPs including 2,4-DCP, 2,4,6-TCP and PCP, or mixtures of these compounds, by

* Corresponding author. E-mail: yuhx@nju.edu.cn

calculating the joint probability curve (JPC). The possibility and degree of current ecological effects caused by CPs in surface waters of China were evaluated and discussed.

1 Methods

1.1 Exposure and toxicity data of CPs

Concentrations of CPs in surface waters were obtained from the literature (Table 1). Two types of exposure concentrations were considered, the median concentration and the maximum concentration (worst case). If the concentration was less than the method detection limit (MDL), a surrogate value equal to half of the MDL was used (Solomon et al., 2000).

Toxicity data for three CPs were collected from the ECOTOX database (USEPA, 2011) and literature (Yin et al., 2003a, 2003b) mainly including fish, crustaceans, molluscs, algae, amphibians and so on if available. The data were selected according to the principle of water quality criteria derivation (Stephan et al., 1985). Chronic toxicity data for 2,4-DCP and 2,4,6-TCP were limited. Relative Potency (ReP) values calculated from the species sensitivity distribution based on median effect concentrations (SSD-EC_{50}) were used to extrapolate chronic toxicity and derive their species sensitivity distribution based on no observed effect concentrations (SSD-NOEC) from PCP's SSD-NOEC as described by Chèvre et al. (2006). Values used were NOECs, or if NOECs were not available, half of the maximum acceptable toxicant concentration (MATC) or two fifths of the lowest observed effect concentration (LOEC) or one fifth of the EC_{50} were used as alternative values to describe chronic toxicity (ANZECC and ARMCANZ, 2000). To provide appropriate protection for the aquatic ecosystems of China, only the aquatic species existing broadly or widely cultivated species in the freshwater of China were selected to construct the SSD (Zhong et al., 2010). Only toxicity data characterized by a specific end point were utilized. The duration of tests of effects on algae was more than 4 days, while the duration for animals was 14 days or longer. When there was more than one value for the same end point and species, the geometric mean was used (Stephan et al., 1985).

1.2 Probabilistic risk assessment (PRA) for individual CPs

The PRA method was performed based on the exposure concentration distribution (ECD) and SSD, which can provide a quantitative description of the distribution of uncertainty and variability, and give more information for environmental management (Wang et al., 2002). The method makes full use of the available data and has been used in risk assessment to describe the range of sensitivities, and rather than stating whether point estimates exceeded a threshold, to provide an estimate of the probability of exceeding a threshold (Solomon et al., 2000; Wang et al., 2002; Chen, 2005; Liu et al., 2009). Here, we used the method of reporting the probabilities of adverse effects to evaluate the risks of CPs in surface waters of China. First, we reported the probability of concentrations exceeding the value associated with the 5th percentile of species being affected.

1.3 Probabilistic risk assessment (PRA) for mixtures of CPs

In the natural environment, there is always more than one toxicant, and species are usually simultaneously exposed to various contaminants. Since CPs have a similar mode of action, the toxicity of mixtures can be determined by use of the concentration addition method (Altenburger et al., 2000) by applying relative potency factors to the concentrations of individual CPs to calculate total equivalent concentration ($C_{\text{equ,tol}}$). The $C_{\text{equ,tol}}$ was calculated by the following equation:

$$C_{\text{equ,tol}} = \sum_{i=1}^{n} C_{\text{equ},i} = \sum_{i=1}^{n} C_i \times \frac{\text{HC}_{50,\text{ref}}}{\text{HC}_{50,i}}$$

where, $C_{\text{equ,tol}}$ is the concentration of reference CP causing a response equivalent to the CPs mixtures; i is ith CP; $C_{\text{equ},i}$ is the concentration of the reference CP causing the same response as the ith CP; C_i is the environmental exposure concentration of the ith CP in surface waters of China; $\text{HC}_{50,\text{ref}}$ and $\text{HC}_{50,i}$ are the hazard concentrations for 50%

Table 1 Statistical summary of CPs exposure concentrations in surface waters of China (unit: μg/L)

Watershed	Number	2,4-DCP		2,4,6-TCP		PCP	
		Median	Maximum	Median	Maximum	Median	Maximum
Songhuajiang River[a]	40	0.00055	0.2500	0.00055	0.2500	0.00055	0.0700
Liaohe River[a]	58	0.0402	0.1700	0.0300	0.0300	0.0500	0.0600
Haihe River[a]	39	0.0200	0.0400	0.0200	0.0400	0.0500	0.0700
Yellow River[a]	50	0.0200	19.9600	0.0300	28.6500	0.0500	0.0700
Yangtse River[a]	150	0.00055	0.3800	0.0007	0.0300	0.0630	0.5940
Huaihe River[a]	39	0.0202	0.2460	0.0040	0.0700	0.0600	0.3510
Pearl River[a]	150	0.00055	0.2640	0.0040	0.0700	0.0315	0.3960
Southeast drainage area rivers[a]	74	0.00055	0.0266	0.0007	0.0220	0.0018	0.0324
Northwest drainage area rivers[a]	18	0.0201	0.0550	0.0201	0.0695	0.0500	0.0600
Southwest drainage area rivers[a]	5	< 0.0011	< 0.0011	< 0.0014	< 0.0014	< 0.0011	< 0.0011
Qiantang River[b]	35	0.585	3.690	0.125	0.940	NA	NA
Taihu Lake[c]	59	0.01957[e]	0.1431	0.03538[e]	0.8404	0.012	0.012
Dongting Lake[d]	8	NA	NA	NA	NA	3.755	103.7

[a] Gao et al., 2008; [b] Chen et al., 2005; [c] Qu et al., 2004, Zhong et al., 2010; [d] Zhang et al., 2001.
[e] Mean conentration was used due to no corresponding data being available.
NA: no data available.

(where 50% of the species are affected) of the reference CP and *i*th CP, respectively, from SSD-NOEC. Here, PCP was taken as reference CPs. Then, a PRA of a mixture of CPs was performed just as for a single CP.

1.4 Statistical analysis

The Jarque-Bera and Kolmogorov-Smirnov tests were both applied to determine if values followed a normal distribution for both raw and log-transformed data from exposure concentration and toxicity data. The results (p > 0.05) indicated that after log-transformation, all data were normally distributed. Thus, all data were log-transformed before constructing SSDs and ECDs. Statistical analyses and determination of the overlapping area between SSD and ECD were performed by R version 2.10.1 (R Development Core Team, http://www.rproject.org/).

2 Results and discussion

2.1 Environmental exposure and toxicity distribution of CPs

Distributions of concentrations of CPs and the distributions of the sensitivity of individual aquatic organisms were plotted concurrently (Fig. 1). Chronic toxicity data for 2,4-DCP and 2,4,6-TCP were also considered. Concentrations in the environment and tolerances of aquatic organisms both had a range of 1000-fold between the minimum and maximum values. Concentrations of CPs also ranged widely among regions (Table 1). Uncertainty, variability and the stochastic properties of exposure and effect can be fully considered by the probabilistic approach (e.g., statistical distribution) (Wang et al., 2002).

Median concentrations did not exceed the minimum tolerance, but maximum concentrations were greater than the tolerances of some of the most sensitive species (Fig. 1). The median and maximum exposure concentrations of 2,4-DCP were 0.585 and 19.96 μg/L (Table 1), respectively, while the most sensitive species had a no observed adverse effect level (NOAEL) of 68.58 μg/L, which was greater than any of the concentrations in surface waters. The median and maximum exposure concentrations of 2,4,6-TCP were 0.125 and 28.65 μg/L (Table 1), respectively, while the most sensitive species toxicity data available was 14.61 μg/L, less than the maximum value of the worst exposure case. The median and maximum exposure concentrations of PCP were 3.755 and 103.7 μg/L (Table 1), respectively, while the most sensitive species toxicity data available was 2.16 μg/L, less than the maximum exposure level values, indicating possible risk. Moreover, PCP had a larger toxicity range than the other CPs, which may be due to its characteristics and tolerance of species to these characteristics. PCP has been banned since the 1980s, but it still exists widely in the environment and poses a possible threat to the ecosystem, which indicates that there may be some natural sources or sources from transformation of other chemicals.

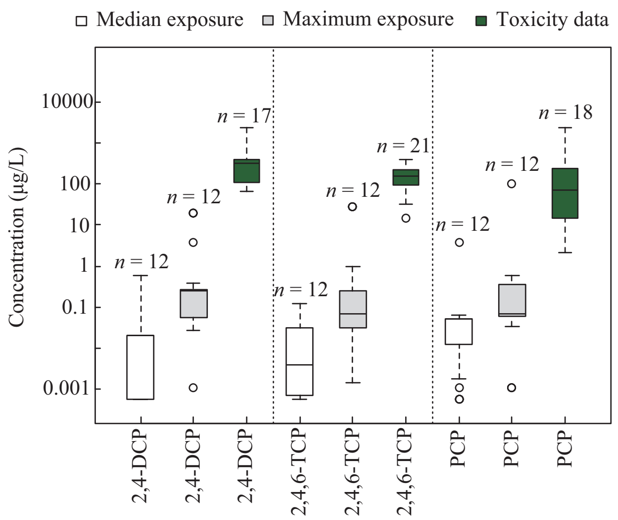

Fig. 1 Boxplot of exposure concentrations and toxicity data distribution of CPs. *n* is the number of samples or aquatic species.

2.2 Probabilistic risk assessment (PRA) of individual CPs

Probabilistic risk assessment of the three individual CPs was performed by calculating the joint probabilities of exceeding a concentration in surface waters and the probability of affecting species (Fig. 2). From Fig. 2, it was very easy and clear to visually evaluate the degree of risk expected (Solomon et al., 2000).

The ECDs of median exposure concentrations ($ECDs_{median}$) of 2,4-DCP and 2,4,6-TCP had small overlapping areas (0.010 for 2,4-DCP, 0.0030 for 2,4,6-TCP) with their own SSD (Fig. 2, dark green area), while the ECDs of maximum exposure concentrations ($ECDs_{max}$) of 2,4-DCP and 2,4,6-TCP had a larger overlapping area (0.077 for 2,4-DCP, 0.080 for 2,4,6-TCP) with their own SSD (Fig. 2, dark green + light gray area). The nearer the JPC is to the axes, the lower the probability of adverse effects (Solomon et al., 2000; Wang et al., 2002). It can been seen that the JPCs based on median exposure concentrations (JPC_{median}) of 2,4-DCP and 2,4,6-TCP are very close to the axes (Fig. 2), indicating a small probability of adverse effects. The risks based on the JPC_{median} of 2,4-DCP and 2,4,6-TCP were negligible (probability for 5% of species affected < 0.5% for the worst case), while the probabilities for 5% of species affected for 2,4-DCP and 2,4,6-TCP in the worst case (JPC_{max}) was 3.48% and 3.74%, respectively (Fig. 2).

However, The ECDs of both median and maximum exposure concentrations of PCP had an overlapping area with their own SSD of 0.111 and 0.271, respectively (Fig. 2), and the probabilities for 5% of species affected from the JPCs were 5.99% and 21.36%, respectively (Fig. 2). Thus, risk of PCP should be considered, and it presents a potential threat to aquatic species.

2.3 Probabilistic risk assessment (PRA) of mixtures of CPs

PCP was used as a reference CP and equivalent concentrations of 2,4-DCP and 2,4,6-TCP were calculated based on their relative potencies. Thus the PRA of mixtures of CPs could be determined. The results of this analysis are

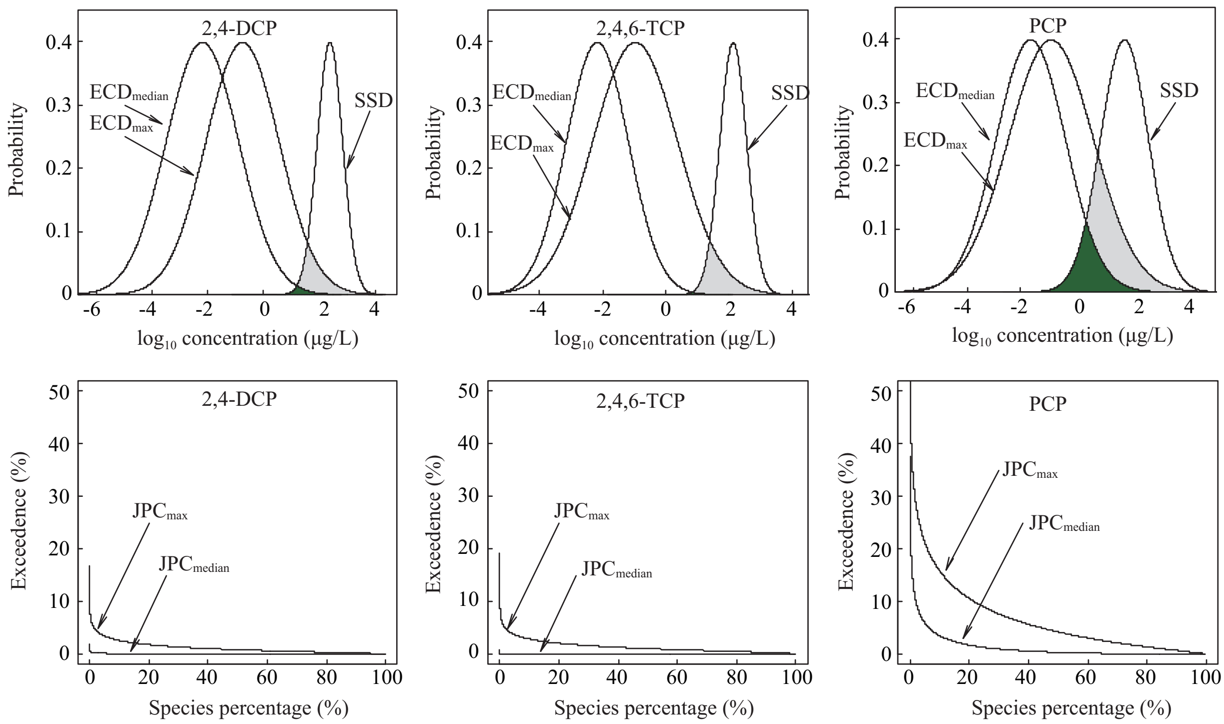

Fig. 2 Probability density curves and joint probability curves for the exposure concentrations and toxicity data of three individual CPs in surface waters in China. ECD_{median} and ECD_{max} were ECD of median and maximum exposure concentrations, respectively, and JPC_{median} and JPC_{max} were JPC of median and maximum exposure concentrations, respectively.

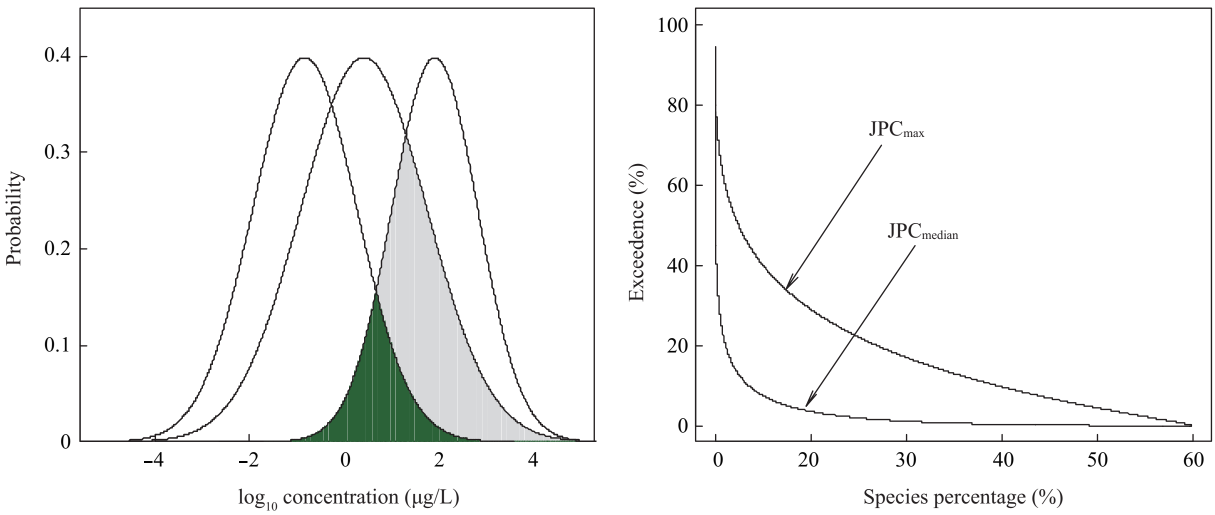

Fig. 3 Probability distributions and joint probability curves for the exposure concentrations and toxicity data for the mixtures of CPs in surface waters of China.

given in Fig. 3. The overlapping areas of the mixtures of CPs were 0.171 (Fig. 3, dark green area) and 0.503 (Fig. 3, dark green+light gray area) in the median and maximum exposure concentration cases, respectively; the probabilities for 5% of species to be affected from JPC_{median} and JPC_{max} were 12.72% and 49.83%, respectively. This result indicates that the ecological risks caused by CPs were worth concern, especially for PCP, which contributes most of the total risk.

Only three of the potential CPs were considered. Thus, additional CPs and other compounds that co-exist in the real aquatic ecosystem could affect the responses of organisms (Davì and Gnudi, 1999; Zhong et al., 2010), and thus the risk for the aquatic ecosystem might be underestimated. These results could provide useful information on the relative risk of CPs in surface waters of China and are of benefit to environmental management for related departments.

2.4 Uncertainty analysis

Uncertainty in ecological risk assessment is inevitable no matter how advanced the methods used are. The uncertainty is associated with variability in the current limited exposure and species effect data, risk models, and characteristics of actual waters, and lack of knowledge (Chen, 2005; Wang et al., 2009). However, a relatively high-level model could be used to decrease the uncertainty as much as possible in the present study.

In particular, the spatial and temporal scales of CPs exposure data in the surface waters of China and the toxicity data used to develop SSDs are important sources of uncertainty. Except for reported data on detected CPs concentrations, there is no additional CPs concentrations data available, and the temporal scale of existing data is not consecutive. The spatial scale mainly includes the different sampling sites, and other sites may have very different concentrations due to social and geographical reasons, and also living species may differ for a different spatial scale. Additionally, application of ReP values (Chèvre et al., 2006) and concentration addition (Altenburger et al., 2000) contributes to some degree of uncertainty. The ecosystem has an intrinsic self-purification capacity and the real ecosystem is more complex than the experimental conditions in the laboratory, including availability of food, water composition, light, temperature, etc., and species have the capacity for acclimation and evolution when exposed to low concentrations. Moreover, the NOEC utilized in this study is based on the experiment design and is derived through hypothesis testing, while the true NOEC should be between the reported NOEC and LOEC. Thereby, the toxicity data itself contain some uncertainty. Future work should be done to obtain more CPs data in various spatial and temporal scales as well as *in situ* toxicity data, and to increase our knowledge about the existing site specific species and their importance in the structure and function of field ecosystems.

3 Conclusions

The two PRA methods reached the similar conclusion, and were able to greatly decrease the level of uncertainty. The potential adverse effects of individual CPs, including 2,4-DCP and 2,4,6-TCP were relatively small for both the median and maximum cases. The joint probability was < 0.1 and the probability of a concentration exceeding the concentration likely to adversely affect 5% of species was $< 4\%$. However, mixtures of CPs, including PCP, are potential of greater concern. Joint probabilities for PCP were 0.271 for the maximum concentrations and 0.111 for the median concentrations. The probabilities of affecting 5% of aquatic species were 21.36% and 5.99% for the maximum and median concentrations, respectively. The joint probabilities for mixtures of CPs were 0.503 and 0.171 for the maximum and median concentrations and the probabilities of affecting 5% of aquatic organisms were 49.83% and 12.72% for the maximum and median concentrations.

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