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Which Commonly Monitored Chemical Contaminant in the Bohai Region and the Yangtze and Pearl Rivers of China Poses the Greatest Threat to Aquatic Wildlife?

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Abstract: The present study assessed the relative risk of 29 chemical contaminants to aquatic wildlife in the Bohai region and the Yangtze and Pearl Rivers of China. River monitoring data from 2010 to 2015 for metals, pesticides, plasticizers, surfactants, polyaromatic hydrocarbons, flame retardants, and ammonia were collected. For each chemical, ecotoxicity data were compiled for Chinese-relevant aquatic species. The chemicals were ranked by relative risk either by comparing the ratios of the median river concentration divided by the median ecotoxicity concentration or by the percentage of river measurements which exceeded the lower 10th percentile ecotoxicity value. To provide context, these results were compared with the same analysis for rivers in the United Kingdom. From this collection of chemicals in Chinese rivers, the highest risks appear to be from Cu, closely followed by Zn, Fe, and Ni together with linear alkyl benzene sulfonate, nonylphenol, and NH_3. This risk, particularly from the metals, can be several times higher than that experienced in UK rivers when using the same analysis. Ammonia median concentrations were notably higher in the Pearl and Yangtze than in UK rivers. The results suggest that China should focus on controlling metal contamination to protect its aquatic wildlife. *Environ Toxicol Chem* 2018;37:1115–1121. © 2017 SETAC

Keywords: Risk; Ecotoxicology; Metal; Pesticide; Ammonia; China

INTRODUCTION

China's economic growth of the past 30 yr has staggered the world. Not only does China support its own fast-growing economy but it supplies much of the rest of the world with the finished goods and chemicals it needs. While it is under pressure to feed its growing population with traditional staple foods, such as rice, its growing affluence is also driving up livestock rearing. Although China has a vast landmass and big rivers to accommodate its growing population, industry, and agriculture, this has led to increasing pressures on its natural environment (Currell and Han 2017). Back in 2004 it was estimated that China's surface waters were receiving 22 billion tons of industrial wastewater and 29 billion tons of domestic wastewater per year (Shao et al. 2006). Not only has this waste discharge had consequences for the environment, but some have linked poor water quality with human health impacts (Wang and Yang 2016). As of April 2017, typing the words "China" and "pollution" and "water" into an academic search engine such as Web of Science™ returned over 6000 entries. Currently there are 1000 new papers on the topic coming out every year. Given the many thousands of chemicals used each year and the wide range of surface and groundwaters into which they are disposed in China, there is certainly no shortage of topics to study. Indeed, the literature is full of discussions on chemical *x* in location *y* and the risks it might pose to species *z*. Valuable as these studies might be, they give no indication of relative risk. Similarly, it is hard to put the levels of contamination in China into context, to say just how bad they are on a worldwide scale. In recent years China has stepped up its efforts to control pollution with the amended environmental policy of April 2014 managed by the Ministry of Environmental Protection (Zhang B et al. 2017). Local officials are now evaluated on the basis of their performance in

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environmental protection, not just economic growth. Concurrent with an improving legal status for water and the environment, there is a greatly increased consciousness and concern by citizens about water quality (Zheng and Shi 2017).

Any attempt to make such assessment of the relative risk of the different chemicals in China's rivers is necessarily limited by the amount of good-quality monitoring data available. But this situation is gradually improving thanks to research translating into scientific publications as well as through the efforts of the China National Environmental Monitoring Centre, which publishes annual reports on the concentrations of a wide range of chemicals throughout China. This enables China to report on a series of 5 different chemical water classes from I to V. Grade I is classed as source water, national natural conservation area; II is suitable for drinking water and as habitat for rare aquatic species; III may also be used as drinking water and for aquaculture; and the lower grades of IV and V may only be used for industry or agricultural needs. So these classes are somewhere between a grading for suitability for human exploitation and a guide to environmental quality such as that used in the Water Framework Directive in Europe. Back in 2004, over 28% of monitored sites were below class V, the lowest status (Shao et al. 2006). China is now actively considering how to link more explicitly contaminant concentrations to water quality criteria for protection of wildlife (Jin et al. 2014).

The traditional approach to prioritize chemicals for regulation is on the basis of their possessing hazardous properties, particularly being persistent, bioaccumulative, and toxic, the so-called PBT chemicals. Having carcinogenic or mutagenic properties may raise their priority still further (Hansen et al. 1999; Wilkinson et al. 2007; Daginnus et al. 2010). China has also considered a "black list" of high PBT chemicals, being the ones deserving the most attention (Jin et al. 2014). Linking risk assessment to regulation has tended to use a threshold value which may be termed an "environmental quality standard" for chemicals of concern. Typically, this is linked to the toxicity of the chemical and is based on a predicted-no-effect concentration (PNEC). This may be derived from a species sensitivity distribution, which can be employed when data are available for at least 20 different species. But where less information is available, the lowest effect concentration for an aquatic species must be found. From such information, an additional safety or adjustment factor is added to derive the PNEC, a level which, if not exceeded, should protect all aquatic wildlife in the absence of other pressures. When a PNEC is compared to the measured environmental concentration (MEC), some sort of risk quotient is generated, which could be used for comparative risk analysis of different chemicals. However, the problem is that, depending on our knowledge or lack of it, different chemicals will receive different adjustment factors, which may be up to 1000 for one substance and only 5 for another (Hansen et al. 1999; von der Ohe et al. 2011). Thus, despite their popularity, these methods have significant drawbacks: firstly, the potentially distorting effect of differing adjustment factors being applied to different chemicals, making relative risk hard to judge, and, secondly, the use of the highest MEC. Thus, when the most high-priority chemical is selected, this may be the result of a combination of an overly precautionary adjustment factor being applied (perversely simply because less ecotoxicity information was available on that chemical) and comparison with some extremely rare high concentrations being recorded in a river. Together, both could distort the risk assessment by overlooking the chemical causing the most frequent damage to wildlife. To avoid these potential errors, a different risk-ranking method has been proposed where a median or percentile of the ecotoxicity data set is compared against the median or a percentile of the MEC, and this has been recently applied to a range of chemicals in the United Kingdom (Donnachie et al. 2014, 2016; Johnson et al. 2017) and in China (Su et al. 2017; Zhang M et al. 2017; Zhang Y et al. 2017). Thus, the ecotoxicity value used in the present study is bounded firmly within the data set and is not a prediction beyond what has been recorded in ecotoxicity testing such as is used to generate a PNEC.

Through gathering ecotoxicological data sets for the selected chemicals for Chinese-relevant wildlife species and by comparing against river measurements from the literature and China National Environmental Monitoring Centre reports, the aims of the present study were as follows. First, to use the risk-ranking approach to identify, from the range of chemicals regularly monitored, which might be of greatest concern in the Bohai coastal region and the Yangtze and Pearl Rivers. Second, to compare the relative risk for these chemicals with the situation in the rivers of England and Wales (UK). Third, to identify from the range of chemicals regularly monitored and of concern which might be having the greatest impacts on wildlife, by examining what percentage of Chinese river measurements exceed the lowest 10th percentile ecotoxicity value (most sensitive organisms).

Approaches, such as the one proposed in the present study, may be valuable in the future to a country like China, in helping it to focus resources where they might have the greatest impact on improving the ecology of freshwaters.

MATERIALS AND METHODS

Location

To set the scene it is helpful to compare the geography (Table 1) at a basic level of the 3 areas selected in China to that of England and Wales (UK). England and Wales is included in the present study as a benchmark of a developed Western country with an established environmental protection infrastructure. In the present study, the Bohai region rivers were considered to include the Beijing area and the area to the west of Beijing draining into the Bohai Sea (this includes only a part of the basin of the Yellow River; Zhang Y et al. 2017). For the Pearl and Yangtze Rivers the whole basins were considered. It should be noted that these rivers rise in the very sparsely populated western region of China. All of these rivers flow from west to east, with the Bohai region in the north, Yangtze in the center, and Pearl in the south of China. Together the area drained by these water courses accounts for 29% of China's landmass and 58% of its human population (Table 1).

Collection of data on the selected chemicals

Although thousands of chemicals may be present in the aquatic environment, only a few of these are measured regularly

TABLE 1: Overview of the surface waters examined in China and the United Kingdom[a]

	Yangtze basin	Pearl basin	Bohai rivers (Liaohe, Daling, Haihe, Yellow River basins)	England and Wales, UK
Area (km^2)	1 800 000	442 100	523 156	151 040
Proportion of land mass (%)	19	5	5	62
Population inland	430 million	90 million	253 million	32 million
Proportion of the population (%)	32	7	19	53
Population density (people/km^2)	239	203	483	212
Mean annual flow (m^3/s)	31 900	10 654	902	2195
Dilution available per person (m^3/cap/d)	6.4	10.3	0.3	5.9

[a]Sources: Area, population, and flow data for China compiled from National Bureau of Statistics (2014), Bureau of Statistics of Liaoning Province (2015), Bureau of Statistics of Tianjin (2015), Bureau of Statistics of Hebei Province (2015), and Bureau of Statistics of Shandong Province (2015). Flow data for Liaohe River basin, Daling River, Haihe River basin, and Yellow River basin (1999) (Li et al. 2009; Wu et al. 2011; Sun et al. 2012; Li et al. 2016). For the United Kingdom the data are from Marsh et al. (2015) and Johnson et al. (2011).

in surface waters. However, these tend to be the chemicals considered of high concern because of their toxic effects. Thus, regular monitoring data on 29 chemicals could be found across all of the Chinese rivers in these regions covering 8 different classes (Table 2). In the present study, concentration data for these chemicals in these rivers were collected both from the scientific literature for the period 2009 to 2015 and from data published in the National Report on Environmental Quality of China for 2013 (Ministry of Environmental Protection 2013). For the Bohai region insufficient measurements were available for the persistent organic pollutants and pesticides of a hydrophobic nature in the water column (with the exception of perfluorooctane sulfonic acid and perfluorooctanoic acid). However, abundant sediment values were available, so water concentrations were estimated based on the K_{OC} value for the chemical and the organic carbon content of the sediments from which they originated (Zhang Y et al. 2017). In the case of ammonium, the most toxic form is the un-ionized NH_3 molecule, but the water measurements are for total ammonium, which is mainly the NH_4^+ ion. However, the proportion of NH_3 present in the water can be calculated if the pH and temperature are also known (Emerson et al. 1975). The quantity and summary of river measurements collected per chemical are shown in Supplemental Data, Tables S1 through S4. Overall, 20 887 different river measurement values were collected for these Chinese surface waters.

For rivers in England and Wales (UK) measured data for the chemicals were collected from the scientific literature (from 2000 onward) but largely from the UK Environment Agency monitoring data ("WIMS" data), using data from 2010 through 2012 (Johnson et al. 2017). Because the ecotoxicity of metals pertains to their dissolved concentration, only dissolved metal measurements in the environment were collected.

Where measurements were recorded as less than the limit of quantification (LOQ), half of the given quantification limit was used. In a few cases the literature reported only summary information, such as number of samples, with range and average. To reflect the number of measurements taken in such a case, the minimum, maximum, and $n - 2$ times the average were entered.

To assist the collection of aquatic ecotoxicity data for an individual chemical, it was found that the US Environmental Protection Agency's ECOTOX database was a good starting point, and this was supplemented by searching the Web of Science database using a series of keywords (Donnachie et al. 2014, 2016). Ecotoxicity data for Chinese local freshwater species and standard test species were selected for each chemical (see Supplemental Data, Table S5 for the species included). Although the response of Chinese species to toxic chemicals is not expected to be markedly different from others worldwide (Jin et al. 2015), it may bring a little extra precision and reassurance to the present study. To help compare results, for the present study the UK surface water measurements were also ranked using these Chinese-relevant species. A range of effect measurements was present in the literature including lowest-observed-effect concentration, median effect concentration (EC50), median lethal concentration (LC50), and acute and chronic toxicity; and all of

TABLE 2: The 29 different chemicals examined in the present study and their different classes

Class	Origin	Examples studied
Metals	Industry and some domestic products	Cu, Zn, Ni, Fe, Cd, As, Pb, Hg, Cr, Mn
Pesticides	Agriculture	DDT, DDE, Endrin, heptachlor, hexachlorocyclohexane (γ, α), hexachlorobenzene
Surfactants or their degradation products	Industry and domestic sources	Linear alkylbenzene sulfonate, nonylphenol, octylphenol
Persistent organics	Industrial and domestic combustion	Phenanthrene, fluoranthene, benzo[*a*]pyrene
Flame retardants	Domestic	Hexabromocyclododecane
Perfluorinated compounds	Industry and domestic sources	Perfluoro-octane sulfonic acid, perfluoro-octanoic acid
Sanitary waste product	Domestic and agriculture (some industry also possible)	NH_3
Plasticizer	Industry and domestic sources	Di(2-ethylhexyl)phthalate, bisphenol-A

DDE = dichlorodiphenyldichloroethylene; DDT = dichlorodiphenyltrichloroethane.

these were collected. The effect data of LC50 and EC50 were preferred for each species in each study. The widest ranges of species and endpoints were considered, to ensure that as representative a picture of species and possible effects was obtained. The total number of ecotoxicity values collected was 6989 with an average of 241 per chemical. A summary of this data is shown in Supplemental Data, Tables S1 through S4. Where several studies reported effect concentrations using the same or different endpoints for one species, the EC50 for a single species was noted. Thus, the final ecotoxicity data set allocated a single value for this single species for the purpose of calculation of the median or percentiles. This refinement was to ensure that the median ecotoxicity value was not swayed by say hundreds of values for *Daphnia* compared to say a few for *Gammarus* and *Lemna*. The reason for selecting one value per species is that it reveals clearly to the viewer the number of different species available for analysis and does not give undue weight to commonly studied species.

Assessment of risk

Once the data sets for ecotoxicology and environmental concentration measurements were considered sufficient, the information in them could be plotted and the medians noted. The final median ecotoxicity value for a chemical was selected from the collection of medians identified for each single species and endpoint. The difference between these medians can be described as a risk ratio, which can be used to rank concern; the larger the value, the greater the concern (Equation 1).

$$\text{Risk} = \frac{m\boldsymbol{W}}{m\boldsymbol{T}} \tag{1}$$

In Equation 1, mW is the median river water concentration (micrograms per liter) and mT is the median effect (i.e., toxicity) concentration (micrograms per liter). Using the medians as a comparator provides a robust method to compare the relative risk of chemicals. However, this relative risk index does not reveal to what degree any of the chemicals might actually be harming local wildlife. It is tempting to compare the concentration affecting the most sensitive species against the highest reported measurement in a river, but this may not be robust and, hence, is open to challenge. This is because there can be concerns over the potential quality of reports on the most sensitive effects on wildlife (Harris et al. 2014) and for the highest measurements in rivers (the extremes; Johnson et al. 2008), so another approach was included. This was to provide a percentage for the number of environmental concentrations which exceeded the lowest 10% of the ecotoxicity data (this can only be provided for the chemicals where this overlap actually occurs).

Sampling locations

A conclusion on environmental risk for chemicals in a river can only be as comprehensive as the monitoring network. An example for the Pearl River is shown (Supplemental Data, Figure S1) where a good coverage for metals and NH_3 is evident throughout the basin, but most measurements for organic chemicals are found only in the downstream reaches. Maps showing the sampling points in the Yangtze River and Bohai region rivers are available as Supplemental Data, Figures S2 and S3.

RESULTS AND DISCUSSION

This form of chemical risk ranking attempts to identify the chemical likely to be having effects on the widest range of species in the widest range of locations/times. An advantage of this risk-ranking method is its transparency: all of the data used can be shown, such as for the Yangtze (Figure 1; see also Supplemental Data, Figures S4 and S5) without the further complexity of hazard-based scoring systems making assessments difficult to assess. To simplify matters further, the risk ratio of the median ecotoxicity and median river measurement can be shown and compared for all of the rivers combined (see Figure 2 and Supplemental Data, Figures S6–S8, for the individual rivers). Focusing on the Chinese situation, from this group of chemicals of concern, the greatest risks appear to be from the metals, most prominently from Cu and Zn; and these 2 were also highlighted for a large lake in eastern China (Fu et al. 2016). This finding, that the highest risks tend to be associated with metals, is similar to the UK data (Johnson et al. 2017). We must be careful to state that this is a preliminary finding because the fraction of bioavailable metal will be less than the dissolved concentration, although this is unlikely to change their prominence. It is noted that the surfactant linear alkyl benzene sulfonate is in the top 5 for risk, although the method used by the China National Environmental Monitoring Centre for measurement in Chinese waters with methylene blue could be of questionable quality. The next highest risk organic chemical in this group is the plasticizer bis(2-ethylhexyl) phthalate and then the surfactant breakdown product nonylphenol. Of the 3 Chinese rivers/regions, the relative risks of these chemicals tended to be lower in the Yangtze (Figure 2). Others have shown that concentrations of chemical pollutants in the Yangtze are not excessive by world standards, although the loads carried inevitably are (Muller et al. 2008; Floehr et al. 2013). Although we can see that overall the risks to wildlife from chemicals will be higher in Chinese rivers than in UK rivers, there appear to be some modest exceptions. In this case, the risks from bisphenol-A, benzo[*a*]pyrene, DDT, hexachlorobenzene, and heptachlor remain higher in UK rivers than in Chinese rivers (Figure 2).

Although using the medians is arguably both a robust and a fair way to compare relative chemical risks, an alternative is to identify the relative predicted impact on wildlife in these rivers. Thus, the percentage of monitoring values (which include data from different years and different stretches of the river) which exceed the lowest 10th percentile of the ecotoxicity values can be identified (Figure 3). In this case, it would appear that one-third of monitoring values for Fe and Cu would be harming the most sensitive 10th percentile of the species (if it were all bioavailable) in the Bohai region rivers. Using the same benchmark, for the Pearl River, 13 to 14% of monitoring values for Cu and Zn exceed the 10th percentile ecotoxicity point,

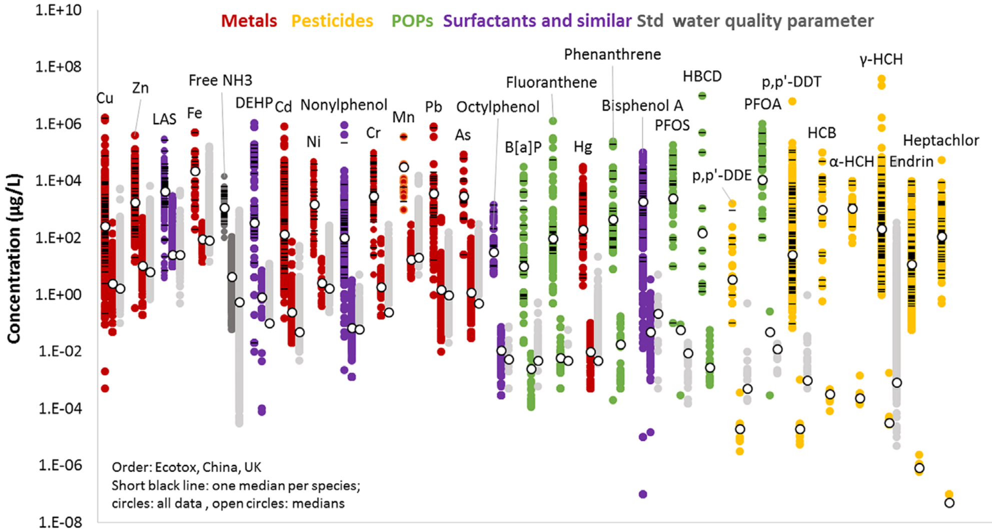

FIGURE 1: Paired data of all of the collected ecotoxicity effect and measured river concentrations for 29 chemicals in the Yangtze River network. For each chemical, 3 rows of data are plotted side by side with the ecotoxicity values on the left, Chinese environmental data in the middle, and, for comparison (in gray), measurements for England and Wales on the right. The ecotoxicity data set shows all values used as colored dots with the median for a particular species as a black horizontal line. Open circles denote the medians (of the species medians for the toxicology data and of all measurements for environmental data). The highest-risk chemicals for Chinese rivers are on the left and the lowest risk on the right. The colors refer to the chemical groups. B[a]P = benzo[a]pyrene; DDE = dichlorodiphenyldichloroethylene; DEHP = bis(2-ethylhexyl) phthalate; HBCD = hexabromocyclododecane; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane; LAS = linear alkylbenzene sulfonate; PFOA = perfluoro-octanoic acid; PFOS = perfluoro-octane sulfonic acid; POP = persistent organic pollutant.

whereas for the Yangtze this was 14% of Cu values. The potential impacts of the other metals appear less for wildlife in the Yangtze. By way of contrast, the greatest predicted impact for English and Welsh rivers (UK) is from 3% of Zn measurements exceeding this 10th percentile ecotoxicity value.

Limitations

The confidence we can put in this or any risk-ranking/prioritisation exercise is limited by the quantity and quality of available data. Not every chemical was measured across every part of these river networks (Figure 1), although metals tended to have the best coverage. Nevertheless, despite these sampling limitations, this coverage is among the best available at this moment. It can be noted from Supplemental Data, Tables S1 through S3 that for some chemicals in some rivers a high proportion of the information was reported as below the LOQ (e.g., 58% of heptachlor values in Bohai rivers). These are recorded as a value which is half the LOQ. In these cases, like for heptachlor, the medians become half the LOQ. This is not ideal, but it could be considered precautionary because with so many nondetects it is likely that the real median concentration would

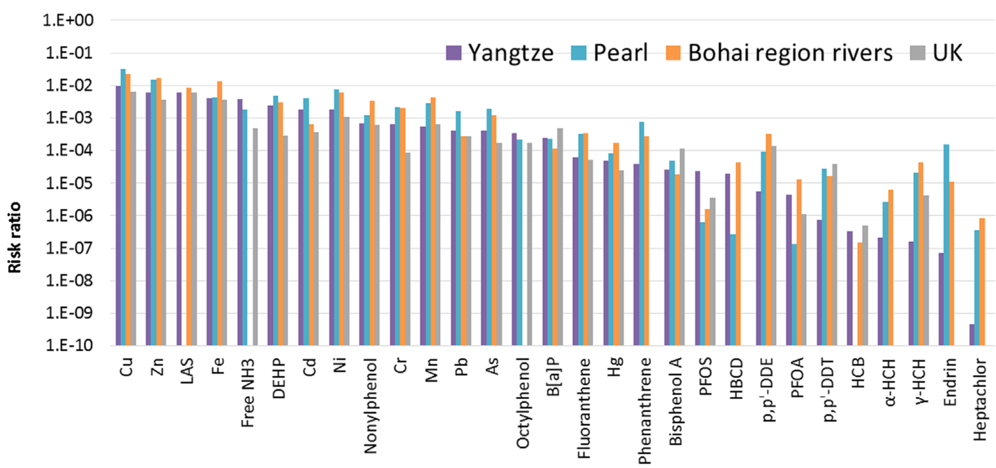

FIGURE 2: Risk ratios from the median ecotoxicity value compared to the median environmental value for each river basin. The larger the value, the higher the risk (ordered by risk ratio in the Yangtze River). B[a]P = benzo[a]pyrene; DDE = dichlorodiphenyldichloroethylene; DDT = dichlorodiphenyltrichloroethane; DEHP = bis(2-ethylhexyl) phthalate; HBCD = hexabromocyclododecane; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane; LAS = linear alkylbenzene sulfonate; PFOA = perfluoro-octanoic acid; PFOS = perfluoro-octane sulfonic acid; POP = persistent organic pollutant.

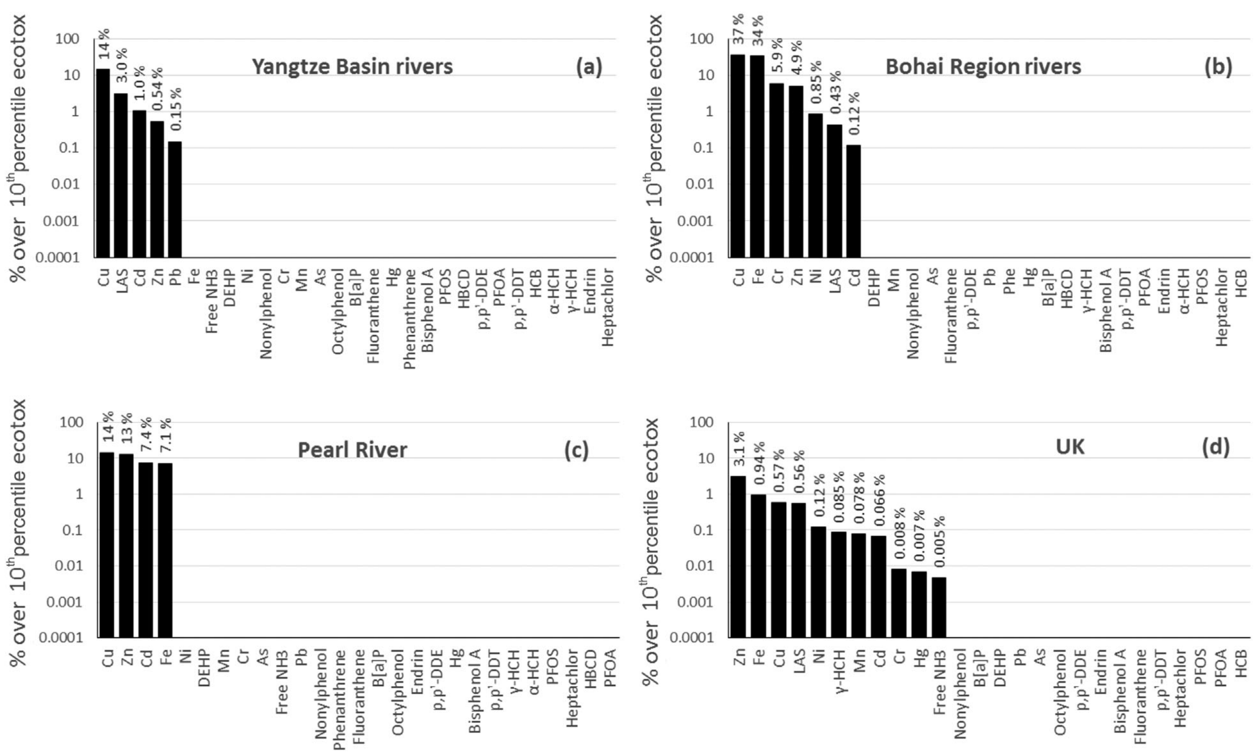

FIGURE 3: Number of monitoring values as a percentage that exceeds the 10th percentile (most sensitive) ecotoxicity value for (**a**) the Yangtze River basin, (**b**) the Bohai region rivers, (**c**) the Pearl River basin, and (**d**) the United Kingdom (chemicals with no overlap are ranked by medians). B[a]P = benzo[a]pyrene; DDE = dichlorodiphenyldichloroethylene; DDT = dichlorodiphenyltrichloroethane; DEHP = bis(2-ethylhexyl) phthalate; HBCD = hexabromocyclododecane; HCB = hexachlorobenzene; HCH = hexachlorocyclohexane; LAS = linear alkylbenzene sulfonate; PFOA = perfluoro-octanoic acid; PFOS = perfluoro-octane sulfonic acid; POP = persistent organic pollutant.

be lower than that. An alternative approach is to base the ranking not on the median but, for example, on the highest 10% of values. In that case a reliable value can be calculated so long as >10% of measurements were detectable and sufficient measurements have been taken to have several values in the top 10%. This risk-ranking exercise was limited to only 29 chemicals which are of high concern out of the many thousands of chemicals that are likely to be present in these rivers. But there is still a value in reviewing what we know now, while recognizing that new information on other chemicals will become available in time and may change the relative risk. Because the metals featured strongly as being of the highest risk, a more thorough reanalysis of their position following careful bioavailability considerations will be necessary.

It is unclear how best to assess the relative risk of hydrophobic chemicals such as the persistent organic pollutants (POPs). They are difficult to measure in water, and there are no standardized ecotoxicity tests which take into account the environmentally relevant exposure through the food web. Thus, both the hazards and the presence of such POPs may be underestimated.

There are also problems in dealing with highly toxic but rarely detectable chemicals such as insecticides. Most monitoring networks are not really appropriate to report concentrations of these chemicals because of their often limited use and short-term applications in agriculture.

Clearly a chemical-by-chemical analysis of risk to the environment ignores mixture effects. Nevertheless, the chemicals found in the present study, which may be commonly found at or near toxic effect levels, will remain a concern. Indeed, the highest-ranked chemicals identified in the present study could guide relevant mixture studies in the future.

CONCLUSIONS

From this collection of chemicals of concern in major Chinese rivers, the highest risks appear to be from the metals led by Cu, and this risk can be several times higher than that experienced in UK rivers. Although there has been improvement in reducing heavy metal pollution in China (Su et al. 2017), more emphasis on the control of Cu, Zn, and Fe may be needed. Assuming a significant proportion of these metals are bioavailable, damaging impacts on the local wildlife could be occurring. The results of the present study argue for a high priority to be given to continuous and resolute measures to control metal pollution to benefit Chinese wildlife.

Median concentrations of ammonia were notably higher in the Pearl and Yangtze than in UK rivers (not examined in the Bohai region rivers in the present study), and this may reflect either a lower standard of human waste treatment in China or losses from agriculture either from livestock waste or fertilizer use. The top organics of concern were the plasticizer bis(2-ethylhexyl) phthalate, the surfactant linear alkylbenzene sulfonate, and the surfactant by-product nonylphenol.

There is an argument that given the many thousands of chemicals undoubtedly present in China's rivers, ranging from new insecticides to pharmaceuticals, for which no systematic measurement as yet exists, it would be premature to draw too many conclusions from the present study. However, the present approach can be built on as more data become available. There can be no doubt that the data indicate that a strong probability of harm from metals remains, and this exceeds what has been found for the United Kingdom.

Supplemental Data—The Supplemental Data are available on the Wiley Online Library at DOI: 10.1002/etc.4042.

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Data availability—All data used were found in publicly available databases as stated in the text or in the cited literature. More details are given in the Supplemental Data.

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