Which chemicals in the Bohai Region, Yangtze and Pearl Rivers of China pose the greatest threat to aquatic wildlife?

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ABSTRACT

This study assessed the relative risk of 29 chemical contaminants to aquatic wildlife in the Bohai Region, Yangtze and Pearl Rivers of China. River monitoring data from 2010-2015 for metals, pesticides, plasticisers, surfactants, poly-aromatic 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 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 UK. 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 (LAS), nonylphenol (NP) 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 China should focus on controlling metal contamination to protect its aquatic wildlife.

INTRODUCTION

China's economic growth of the past 30 years 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. Whilst 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. Whilst China has a vast landmass and big rivers to accommodate its growing population, industry and agriculture, this has led to an increasing pressures on its natural environment [1]. 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 [2]. Not only has this waste discharge had consequences for the environment, but some have linked poor water quality with human health impacts [3]. As of April 2017, typing the words China and pollution and water into an academic search engine such as Web of Science™ returns over 6000 entries. Currently there are 1000 new papers on the topic coming out every year. Given the many 1000's of chemicals used each year and the wide range of surface and groundwaters into which they are disposed in China, there are 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 [4]. Local officials are now evaluated on the basis of their performance in 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 [5].

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 but also through the efforts of the China National Environmental Monitoring Centre (CNEMC) who publish annual reports on the concentrations of a wide range of chemicals throughout China. This enables China to report on a series of five different chemical water classes from I to V. Grade I is classed as 'source water, national natural conservation area, II as suitable for drinking water and suitable habitat for rare aquatic species, III may also be used as drinking water and for aquaculture whilst the lower grades of IV and V may only be used for industry or agriculture needs. So these classes are somewhere between a grading for suitability for human exploitation and a guide to environmental quality such as used in the Water Framework directive in Europe.

Back in 2004, over 28% of monitored sites were below class V, the lowest status [2]. China is now actively considering how to link more explicitly contaminant concentrations to water quality criteria for protection of wildlife [6].

The traditional approach to prioritise chemicals for regulation is on the basis of their possessing hazardous properties, particularly being persistent, bio-accumulative and toxic, the so-called PBT chemicals. Having carcinogenic or mutagenic properties may raise their priority still further [7-9]. China has also considered a ‘black list’ of high PBT chemicals being the ones deserving the most attention [6]. 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 (SSD), 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 (AF) 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 AFs, which may be up to 1000 for one substance and only 5 for another [7, 10]. Thus, despite their popularity, these methods have significant drawbacks; firstly the potentially distorting effect of differing AFs 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 due to a combination of an over-precautionary AF being applied (perversely simply because less ecotoxicity information was available on that chemical) and compared 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 dataset is compared against the median or a percentile of the MEC and this has been recently applied to a range of chemicals in the UK [11-13], and in China [14-16].

Through gathering ecotoxicological datasets for the selected chemicals for Chinese relevant wildlife species and by comparing against river measurements from the literature and CNEMC reports, the aims of this study were to:

- Use the risk ranking approach to identify the chemicals of greatest concern in the Bohai coastal Region, Yangtze and Pearl Rivers
- Compare the relative risk for these chemicals with the situation in the rivers of England and Wales (UK)
- Identify which chemicals might be having the greatest impacts on wildlife. This will be by examining what percentage of Chinese river measurements exceed the lowest 10th percentile ecotoxicity value (most sensitive organisms).

MATERIALS AND METHODS

Location

To set the scene it is helpful to compare the geography (Table 1) at a basic level of the three areas selected in China against to that of England and Wales (UK). England and Wales is included here as a form of benchmark of a developed Western Country with an established environmental protection infrastructure. In this study, the Bohai Region rivers were considered to include the Beijing area and to the west of Beijing draining into the Bohai Sea (this includes only a part of the basin of the Yellow River) [14]. 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 these rivers flow from west to east with the Bohai Region in the north, Yangtze in the centre 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).

Table 1. Overview of the surface waters examined in China and the UK

	Yangtze Basin	Pearl Basin	Bohai Rivers (Liaohe, Daling, Haihe, Yellow river basins)	England & Wales
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/sec)	31,900	10,654	902	2,195
Dilution available per person (m^3/cap/d)	6.4	10.3	0.3	5.9

Sources: Area, population and flow data for China compiled from National Bureau of Statistics [17], Liaoning Statistical Yearbook 2015 [18], Tianjin Statistical Yearbook 2015 [19], Hebei economic Yearbook 2015 [20] and Shandong Statistical Yearbook 2015 [21]. Flow data for Liaohe River Basin, Daling River, Haihe River Basin and Yellow River Basin (1999) was compiled from literature [22-25]. For the UK the data is from Marsh, Sanderson and Swain [26] and Johnson, Yoshitani, Tanaka and Suzuki [27]

Collection of data on the selected chemicals

Whilst 1000s of chemicals may be present in the aquatic environment, only a few of these are measured regularly in surface waters. However, these tend to be the chemicals considered of high concern due to 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 this 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 [28]. 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 PFOS and PFOA). However, abundant sediment values were available and 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 [14]. In the case of ammonium, the most toxic form is the un-ionised 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 [29]. The quantity and summary of river measurements collected per chemical is shown in SI Table SI 1-4. Overall 20,887 different river measurement values were collected for these Chinese surface waters.

For England and Wales (UK) river measured data for the chemicals was collected from the science literature (from 2000 onwards where few data existed), but largely from the UK Environment Agency monitoring data (“WIMS” data), using 2010-2012 data [11]. As the ecotoxicity of metals pertains to their dissolved concentration, only dissolved metal measurements in the environment were collected.

Where measurements were recorded as <LOQ half of the given quantification limit was used. In a few cases the literature reported only summary information, such as number of samples (n), 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 EPA ECOTOX database was a good starting point https://cfpub.epa.gov/ecotox/ , and this was supplemented by searching the Web of Science^{TM} database using a series of key words [12, 13]. Ecotoxicity data for Chinese local freshwater species and standard test species were selected for each chemical (See SI Table 5 for the species included). Whilst the response of Chinese species to toxic chemicals is not expected to be markedly different from others worldwide [30], it may bring a little extra precision and reassurance to this study. To help compare results, for this study the UK surface water measurements were also ranked using these Chinese relevant species. A range of effect measurements were present in the literature including LOEC, EC_{50}, LC_{50}, acute and chronic toxicity and all of these were collected. The effect data of LC_{50} and EC_{50}, was preferred for each species in each study. The widest range of species and end-points were considered, to ensure that as representative a picture of species and possible effects was obtained. The total number of ecotoxicity values collected were 6,989 with an average of 241 per chemical. A summary of this data is shown in SI Tables SI 1-4. Where several studies reported effect concentrations using the same or different end-points for one species, then the median effect concentration for a single species was noted. Thus, the final ecotoxicity dataset 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 100s' 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.

Table 2. The 29 different chemicals examined in the 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-HCH, α-HCH), Hexachloro-benzene (HCB)
Surfactants or their degradation products	Industry and domestic sources	Linear alkylbenzene sulfonate (LAS), nonylphenol, octylphenol
Persistent organics	Industrial and domestic combustion	phenanthrene, fluoranthene, benzo[a]pyrene (B[a]P)
Flame retardants	Domestic	Hexabromo-cyclododecane (HBCD)
Perfluorinated compounds	Industry and domestic sources	Perfluorooctane sulfonic acid (PFOS), perfluoro octanoic acid (PFOA)
Sanitary waste product	Domestic and agriculture (some industry also possible)	NH_3
Plasticiser	Industry and domestic sources	Di(2-ethylhexyl) phthalate (DEHP), Bisphenol-A (BPA)

Assessment of risk

Once the datasets for ecotoxicology and environmental concentrations 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 end-point. 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).

$$\text{Risk} = \frac{m\boldsymbol{W}}{m\boldsymbol{T}} \qquad \text{(Equation 1)}$$

Where mW is the median river water concentration (μg/L) and mT is the median effect (i.e. toxicity) concentration (μg/L). 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 [31] and also for the highest measurements in rivers (the extremes) [32] so another approach was included. This was to provide a percentage for the number of environmental concentrations which exceeded the lowest 10%ile 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 (Supporting Information (SI) 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. Note maps showing the sampling points in the Yangtze River and Bohai Region rivers are available as SI figures S2 and S3.

RESULTS AND DISCUSSION

This form of chemical risk ranking is attempting 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 the data used can be shown, such as for the Yangtze (figure 1, see also SI 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 (figure 3, see also 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 two were also highlighted for a large lake in Eastern China [33]. This finding, that the highest risks tend to be associated with metals is similar to the UK [11]. We must be careful to state, that this is a preliminary finding, as the fraction of bioavailable metal will be less than the dissolved concentration although this is unlikely to change their prominence. It will be noted that the surfactant LAS is in the top five for risk although the method used by the CNEMC for measurement in Chinese waters with methylene blue could be of questionable quality. The next highest risk organic chemical in this group is the plasticiser DEHP and then the surfactant breakdown product nonylphenol. Of the three 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 [34, 35]. Although we can see that overall the risks to wildlife from chemicals will be higher in Chinese rivers than the UK, there appear to be some modest exceptions. In this case, the risks from bisphenol-A, benzo[a]pyrene, DDT, HCB and heptachlor remain higher in UK rivers than in the Chinese ones (figure 2).

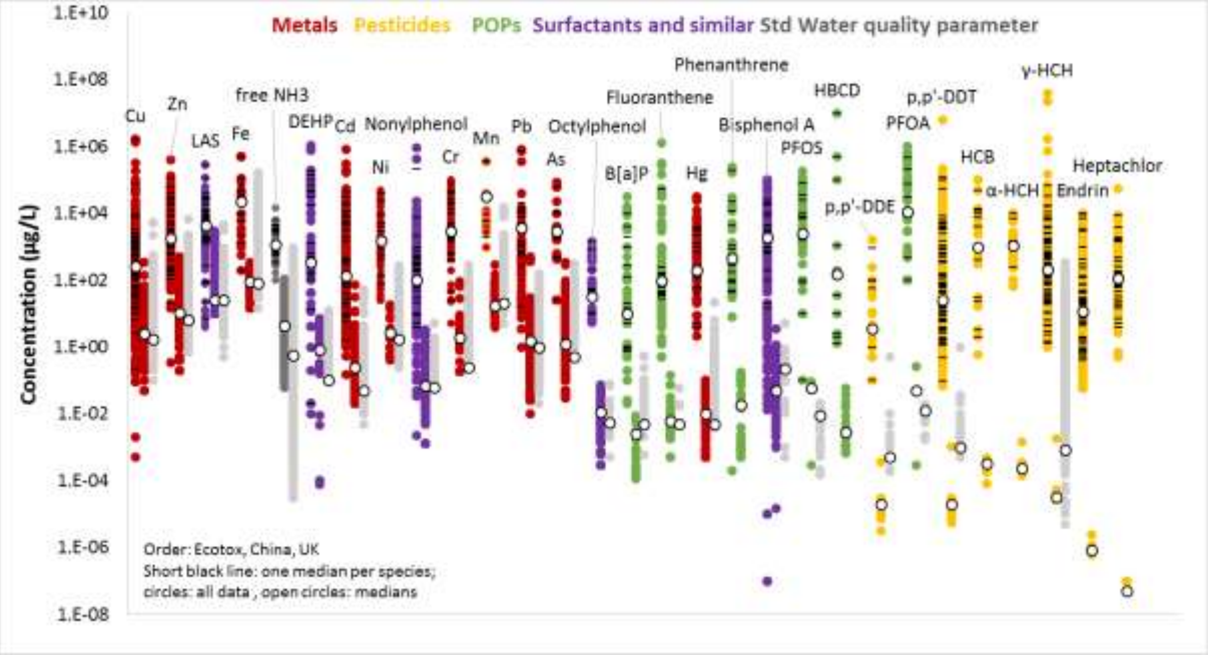

Figure 1. Paired data of all the collected ecotoxicity effect and measured river concentrations for 29 chemicals in the Yangtze River network. For each chemical, three rows of data are plotted side by side with the ecotoxicity values on the left, Chinese environmental data in the middle and -for comparison (in grey)- measurements for England and Wales on the right.

The ecotoxicity dataset shows all values used as coloured dots with the median for a particular species as a black horizontal line. The 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 colours refer to the chemical groups

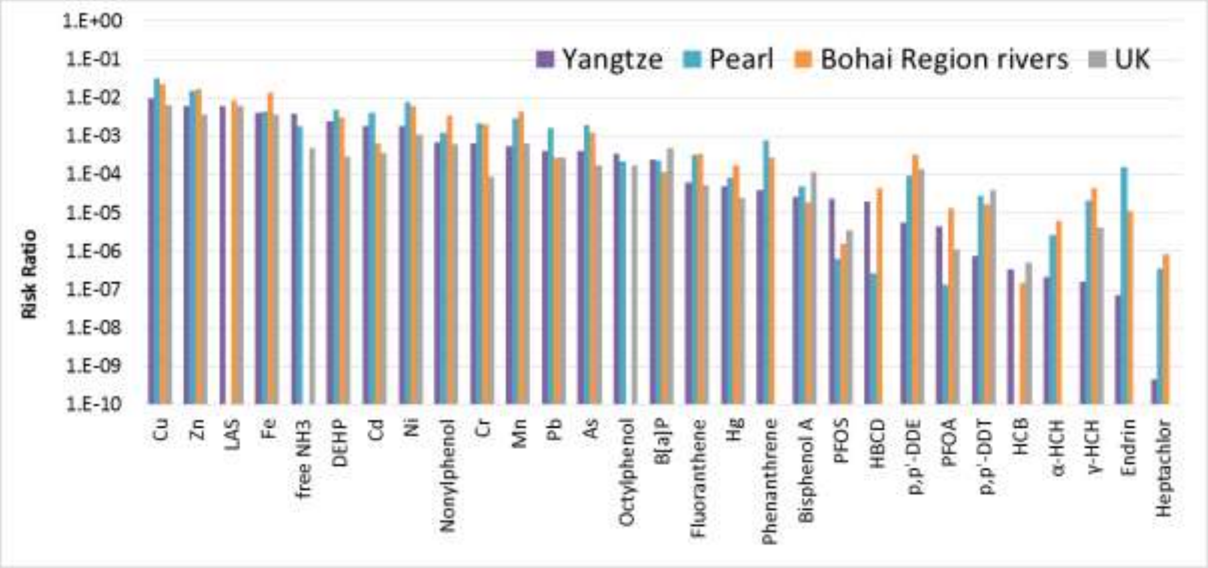

Figure 2. Risk ratios from the median ecotoxicity value compared to the median environmental value for each river basin. The larger the value the higher the risk (ordered by risk ratio in the Yangtze river).

Whilst using the medians is arguably both a robust and 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 a 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 bench-mark, for the Pearl River, 13-14% of monitoring values for Cu and Zn exceed the 10th percentile ecotoxicity point, whilst 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.

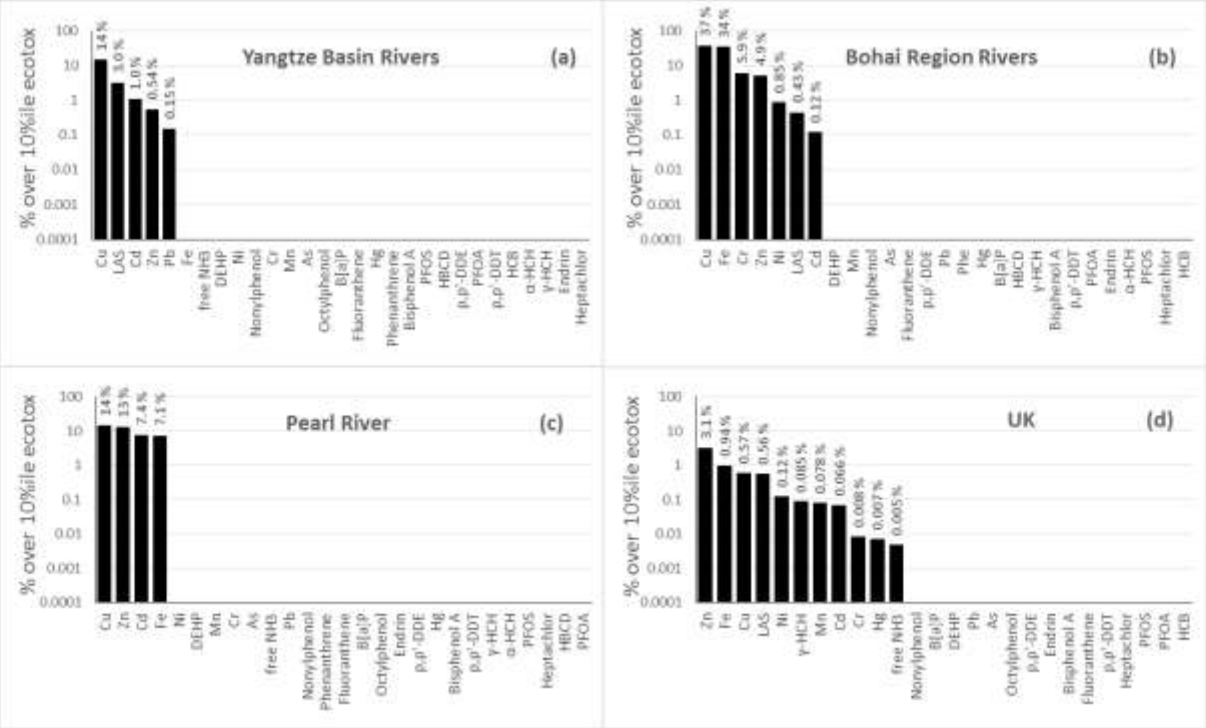

Figure 3. Number of monitoring values as a percentage that exceeds the 10th percentile (most sensitive) ecotoxicity value for (a) the Yangtze river basin; (b) the Bohai region rivers; (c) the Pearl river basin; (d) the UK (chemicals with no overlap are ranked by medians).

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 amongst the best available at this moment. It will be noted from SI Tables S1-3 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 as precautionary, since with so many non-detects it is likely that the real median concentration would 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 more than 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, whilst recognising that new information on other chemicals will become available in time and may change the relative risk. As the metals featured strongly as being of the highest risk, so a more thorough re-analysis 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 POPs. They are difficult to measure in water and there are no standardised ecotoxicity tests which take into account the environmentally relevant exposure through the food web. Thus, both the hazards and 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, due to 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 here, which may be commonly found at levels at our near toxic effect levels, will remain a concern. Indeed the highest ranked chemicals identified here 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. Whilst there has been improvement in reducing heavy metal pollution in China [15], perhaps more emphasis on the control of Cu, Zn and Fe is needed? Assuming a significant proportion of these metals are bioavailable, then damaging impacts on the local wildlife could be occurring. The results of this study would argue for a high priority to be given to continuous and resolute measures to control metal pollution to benefit Chinese wildlife.

It should be noted that ammonia median concentrations were notably higher in the Pearl and Yangtze than in UK Rivers (not examined in the Bohai Region Rivers in this study) and this may reflect either a lower standard of human waste treatment in China or losses from agriculture either from livestock waste or fertiliser use. The top organics of concern were the plasticiser DEHP, the surfactant LAS and surfactant by-product nonylphenol.

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

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Data Accessibility – All data used is found in publicly available databases as stated in the text or in the cited literature. More details are given in the supplemental information.

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Figure captions

Table of Content (TOC) art: Measured chemical concentrations in Chinese rivers were compared with toxicity data to rank the relative risks of dozens of chemicals to wildlife. The top 10 chemicals posing the greatest threat in each study area are shown.

Figure 1. Paired data of all the collected ecotoxicity effect and measured river concentrations for 29 chemicals in the Yangtze River network. For each chemical, three 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 grey)- measurements for England and Wales on the right. The ecotoxicity dataset shows all values used as coloured dots with the median for a particular species as a black horizontal line. The 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 colours refer to the chemical groups.

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).

Figure 3. Number of monitoring values as a percentage that exceeds the 10^{th} percentile (most sensitive) ecotoxicity value for (a) the Yangtze river basin; (b) the Bohai region rivers; (c) the Pearl river basin; (d) the UK (chemicals with no overlap are ranked by medians).