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Spatiotemporal distribution of ecological risk of antibiotics in seven major river basins of China: An optimized multilevel assessment approach

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ABSTRACT

Antibiotics have been recognized as emerging pollutants due to their ecological and human health risks. This paper aims to enhance the ecological risk assessment (ERA) framework for antibiotics, to illustrate the distribution of these risks across different locations and seasons, and to identify the antibiotics that pose high ecological risk. This paper focuses on 52 antibiotics in seven major basins of China. Relying on the optimized approach of ERA and antibiotic monitoring data published from 2017 to 2021, the results of ERA are presented in multilevel. Across the study area, there are marked variations in the spatial distribution of antibiotics' ecological risks. The Huaihe River Basin, the Haihe River Basin, and the Liaohe River Basin are the top three in the ranking of present ecological risks. The research results also reveal significant differences in temporal variation, underscoring the need for increased attention during certain seasons. Ten antibiotics with high contribution rates to ecological risk are identified, which is an important reference to formulate an antibiotic control list. The multilevel results provided both risk values and their ubiquities across a broad study region, which is a powerful support for developing ecological risk management of antibiotics.

Key words: antibiotics, ecological risk assessment, river basins, spatiotemporal distribution

HIGHLIGHTS

- Optimized the calculation method of multilevel ecological risks.
- Provided ecological risk spatiotemporal distribution of antibiotics for the seven major river basins of China (2017–2021).
- Provided the control priority for the seven major river basins.
- Provided the top concerned antibiotics with high contribution to ecological risks of the seven major river basins.

1. INTRODUCTION

Antibiotics, commonly used for treating bacterial infections, are harmful to ecosystems due to their toxicities and lead to antibiotic resistance of bacteria. Consequently, antibiotics are recognized as emerging pollutants internationally (Zhang & Li 2020). After consumption, 70–90% of antibiotics are excreted in their original form or as active metabolites. Current wastewater treatment technologies often fail to effectively remove them, leading to the continuous release of substantial amounts of antibiotics into the aquatic environment. This contributes to widespread and long-term ecological risks (Liu *et al.* 2018; Li *et al.* 2020; Zhang *et al.* 2021). It is often reported that China as a major producer and user of antibiotics has formed a habit of antibiotic abuse. Antibiotic ecological risk has become a focal point in environmental research and management in China (Lyu *et al.* 2020). The 'Action Plan for the Control of New Pollutants' issued by the Chinese State Council Office on 4 May 2022 clearly states that antibiotics are categorized as emerging pollutants and pose ecological and health risks for China. This action plan requires the initiation of a comprehensive investigation of chemical substances, environmental monitoring, and environmental risk assessment, followed by the formulation of supplementary lists, control strategies for emerging pollutants, as well as the development of local standards. Therefore, the ecological risk assessment (ERA) covering sufficient results of antibiotic monitoring can present the ecological risk of antibiotics at a national scale, which is a necessary step for environmental management.

In principle, the ecological risk of a substance is determined by its environmental monitoring concentration (MEC) and toxicity to organisms. Normally, predicted no-effect concentration (PNEC) is used to indicate the toxicity. Logically, lower PNEC represents the lower MEC that can cause ecological hazards, which means higher toxicity could lead to more serious ecological risk. Based on this principle, monitoring of antibiotics in surface waters and ERA has been widely conducted, yielding abundant research outcomes (Li *et al.* 2020; Shaheen *et al.* 2022). However, these efforts have evident limitations. Firstly, most studies primarily utilize the risk quotient (RQ), which is a simple, deterministic quotient expressed as MEC divided by PNEC (Xu *et al.* 2021; Mheidli *et al.* 2022). When MEC and PNEC values for antibiotics are numerous in a study region, the single method of RQ cannot comprehensively characterize ecological risk, causing results to be overestimated or underestimated (Yang *et al.* 2023). Moreover, it is inevitable that there may be outliers within either MEC or PNEC that may lead to bias or misinterpretation of risks (Donnachie *et al.* 2016). Thus, a probabilistic ERA of antibiotics is emerging. Based on RQ, a semi-probabilistic method is used to determine the risk values by multiplying RQ with the frequency of concentrations exceeding PNEC (Liu *et al.* 2020a, 2020b). The advantage of this method is that it provides risk assessment results reflecting the risk overflow rate that represents the ubiquity of ecological risk levels across a broad study region as opposed to point estimates provided by the traditional deterministic ERA, thereby reducing the influence of outliers (Chen *et al.* 2023). When considering PNEC as another determinant of RQ, the related probabilistic methods should also be included, such as probabilistic ERA and species sensitivity distribution (Hu *et al.* 2018; Le Page *et al.* 2019). However, these methods require plentiful ecological toxicity data, while most antibiotics, as emerging pollutants, do not have sufficient PNEC values at present (Yang *et al.* 2018; Liu *et al.* 2020a, 2020b; Rathi *et al.* 2021). The semi-probabilistic method focusing on MEC therefore is suitable for conducting the present ERA of antibiotics. Secondly, there is a wide variety of antibiotics, over a hundred commonly used in clinical practice and dozens with environmental monitoring data. On the other hand, existing researches have focused solely on monitoring and risk assessment of a limited number of antibiotics (Xi *et al.* 2019; Yin 2021). Thirdly, the majority of existing articles concentrate on localized antibiotic monitoring without extensive comparison and risk screening, making it difficult to provide comprehensive data support for the formulation of priority lists of antibiotics. Lastly, some studies fail to consider the seasonality of the ecological risk of antibiotics, which hinders them from reflecting the temporal distribution patterns nationally or in the study region. As a result, there is a lack of data supporting the ecological risk management of antibiotics, especially for policy making.

Considering the above-mentioned issues, the first objective of the paper was to optimize the assessment method of ecological risk by combining RQ with the overflow probability (F) to form the priority index (PI). The second objective was to analyze monitoring data of antibiotics (from 2017 to 2021) in seven major river basins of China where the human populations are concentrated and then to calculate multilevel ecological risk values. The third objective was to reveal whether antibiotic ecological risks vary with season and region, and to list high-risk antibiotics across seven major river basins of China, intending to provide data support for the prioritized monitoring of antibiotics and their ecological risk management.

2. METHODOLOGY AND DATA SOURCES

2.1. Multilevel ecological risk calculation

The multilevel ecological risk calculation in this study comprises three specific levels, which are described in Figure 1. At the first level, RQ calculation is depicted in the European Commission's technical guidance document (European Commission 2003). For an antibiotic, its RQ is determined by the ratio of MEC to PNEC. RQ values are categorized into high (RQ_H, $RQ \geq 1$), medium (RQ_M, $0.1 \leq RQ < 1$), and low (RQ_L, $RQ_L \leq 0.01$) risk. Various formats of MEC are expressed in environmental monitoring publications. To unify its format, this study gives priority by the following sequence: mean value (MEC_{AVG}) > median value (MEC_{MED}) > maximum value (MEC_{MAX}) > minimum value (MEC_{MIN}), which is achieved by the IF function of logical operations in Figure 1. Toxicity data (C_{TOX}) include both the acute toxicity data such as the lethal concentration (LC_{50}), the effective concentration for 50% of organisms (EC_{50}), and the inhibitory concentration for 50% of organisms (IC_{50}), and the chronic toxicity data such as the lowest observed effect concentration (LOEC) and the no observed effect concentration (NOEC). PNEC is the ratio of C_{TOX} to assessment factors (AF). Depending on the type and number of C_{TOX}, PNEC can be calculated in four distinct scenarios.

The RQ method, while indicating risk values for specific water bodies, is inadequate for capturing the ubiquity of ecological risk levels across a broad study region. Thus, the second level involves F to express the proportion of MEC in the river basin that exceeds PNEC, aiming to comprehensively reflect the possibility of ecological risk throughout the river basin (NORMAN

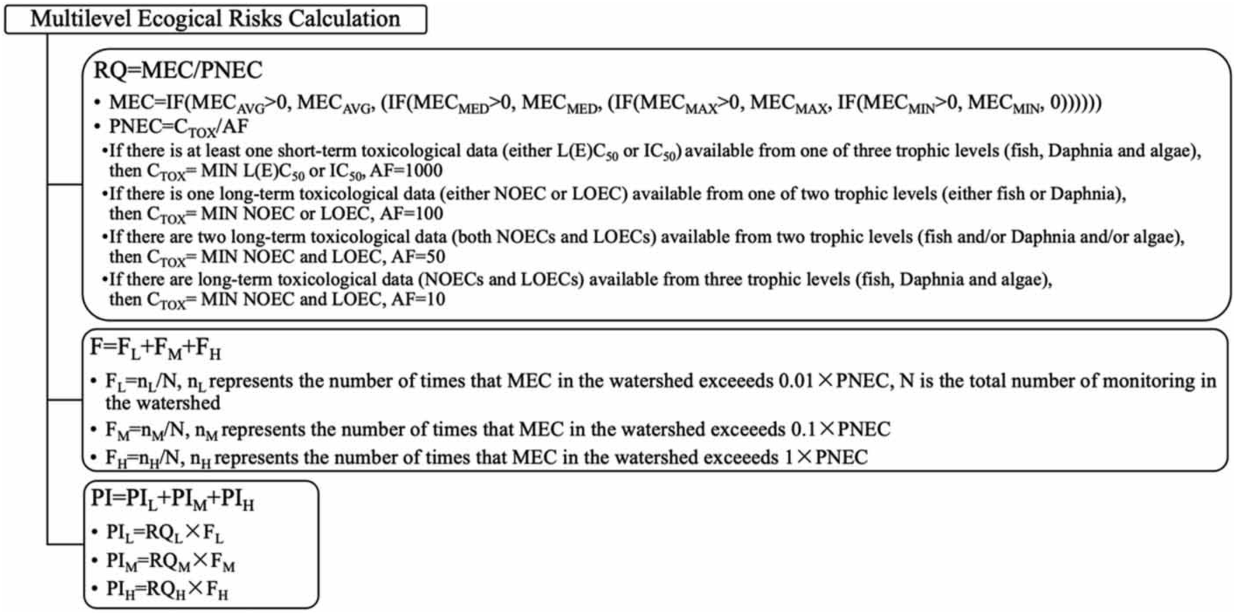

Figure 1 | Multilevel ecological risk calculation process.

Association 2013). Corresponding to the three classifications of RQ in the first level, F is divided into low-risk overflow rate (F_L), medium-risk overflow rate (F_M), and high-risk overflow rate (F_H).

Based on the results of the above two levels' calculations, the third level employs PI to express the risk value as the product of RQ and F. According to three classifications of both RQ and F, PI is expressed as PI_L, $PI_{M,}$ and PI_H. The summation of the three classifications of PI presents the overall ecological risk, which is used to compare ecological risks within the study region of large-scale.

2.2. Study area and data sources

This study focuses on the surface waters of seven major river basins in China, including Heilongjiang, Liaohe, Haihe, Yellow River, Huaihe, Yangtze, and Pearl River Basins (Zhang *et al.* 2018). Seven major river basins, covering 47.3% of China's land area, contribute 82.6% of China's gross domestic product (GDP) and contain 82.7% of China's population (Song 2023). The monitoring data are sourced from environmental monitoring publications, master's and doctoral theses published on the China National Knowledge Infrastructure (https://www.cnki.net) from 2017 to 2021, as well as papers published on the Web of Science. The monitoring data are drawn from 75 publications, covering 70 surface water bodies and 107 types of antibiotics. The statistics on monitoring data are presented in Table 1. The monitoring data are categorized into dry and wet

Table 1 | Statistics on antibiotic monitoring data of surface water in seven major river basins from 2017 to 2021

Basins	Surface waters involved	Antibiotics involved	Sampling sites (publication involved)	Population density, people/square kilometer (Song 2023)
Heilongjiang	2	21	42 (4)	82.8
Liaohe	3	22	74 (3)	131
Haihe	6	39	207 (7)	470
Yellow River	6	30	101 (7)	152.1
Huaihe	2	37	72 (2)	617.8
Yangtze River	34	91	595 (36)	256.3
Pearl River	17	71	186 (16)	338.5
SUM	70	107	1,277 (75)	–

periods based on the sampling date. Kinds of literature with data sources and monitoring sites of each basin are listed in Supplementary Table S1. Boundary data for seven major river basins are obtained from the National Earth System Science Data Center (http://www.geodata.cn/).

2.3. Toxicity data

Toxicity data of antibiotics for three trophic levels (fish, daphnia, and algae) were obtained from the US EPA ECOTOX toxicity database (https://cfpub.epa.gov/ecotox/). When selecting parameters of toxicity data, five indicators were set as toxicity endpoints (LC_{50}, EC_{50}, IC_{50}, LOEC, and NOEC), and freshwater was chosen as the exposure medium. After querying and calculations, 52 antibiotics with toxicological data were listed in Supplementary Table S2.

3. RESULTS AND DISCUSSION

3.1. Spatial distribution of ecological risk of antibiotic

Using monitoring data from a total of 1,277 sampling sites across seven major river basins from 2017 to 2021, the average RQ, F, and PI values were calculated. The ranking of multilevel ERA results is shown in Table 2. In terms of ranking order, it can be observed that there are relatively minor differences between RQ and PI. The Huaihe River Basin, the Liaohe River Basin, and the Haihe River Basin are the top three concerning both RQ and PI rankings, while the Pearl River Basin is the smallest and ranked last. Compared to the previous research results shown in the second column on the right of Table 2 (Yin 2021), the ranking positions for most basins are similar, except for the Huaihe River Basin and the Pearl River Basin. The exception could be due to the different sources of exposure and toxicity data. The exposure data used by Yin 2021 were from 2010 to 2020 and related to only 13 antibiotics, and toxicity data were collected from the literature rather than the toxicity database used by this article.

The risk values of ERA are shown in Figure 2. RQ values of the top three ranked river basins are greater than 0.1, indicating the medium-risk level based on the RQ criteria. As the second level of the multilevel ERA, F expresses the ubiquity of RQ across a broad study region. The occurrence frequency of high-risk, F_H, in the Huaihe River basin, the Liaohe River basin, and the Haihe River basin are also ranked in the top three, further confirming their risk level and indicating that more attention should be directed to future monitoring and management of these river basins.

The spatial distribution of ecological risks depends on many factors that could be related to social and environmental conditions, such as population density, precipitation, temperature, water flow, photolysis, biodegradation, etc. (Chen *et al.* 2018; Qiao *et al.* 2018; Huang *et al.* 2019). As half of antibiotics are used in animal farming, the ratio of animal husbandry in the economic structure of a study area is also a significant factor for the distribution (Zhou *et al.* 2020; Patyra *et al.* 2023). The population density of each basin shown in Table 2 presents a certain relationship with the spatial distribution, except for the Liaohe River Basin and the Pearl River Basin. Other factors in social and environmental conditions could play stronger roles for the exception.

There are 1,277 sampling sites across seven major river basins from 2017 to 2021. Figure 2 also shows the distribution of monitoring sites and locations of megacities. All 22 megacities with a population of more than 5 million people are distributed in seven major river basins. The distribution reveals that the monitoring efforts are currently concentrated around megacities,

Table 2 | Ranking of multilevel ERA for seven major river basins of China

Basins	RQ_{SUM}	F			PI	RQ_{SUM} (Yin 2021)	Population density
		F_L	F_M	F_H			
Huaihe River	1	7	3	3	1	7	1
Liaohe River	2	5	1	1	3	2	6
Haihe River	3	2	4	2	2	1	2
Yellow River	4	3	2	6	6	5	5
Yangtze River	5	4	5	5	4	4	4
Heilongjiang River	6	1	6	7	5	6	7
Pearl River	7	6	7	4	7	3	3

Note: The ranking decreases in the order of 1–7.

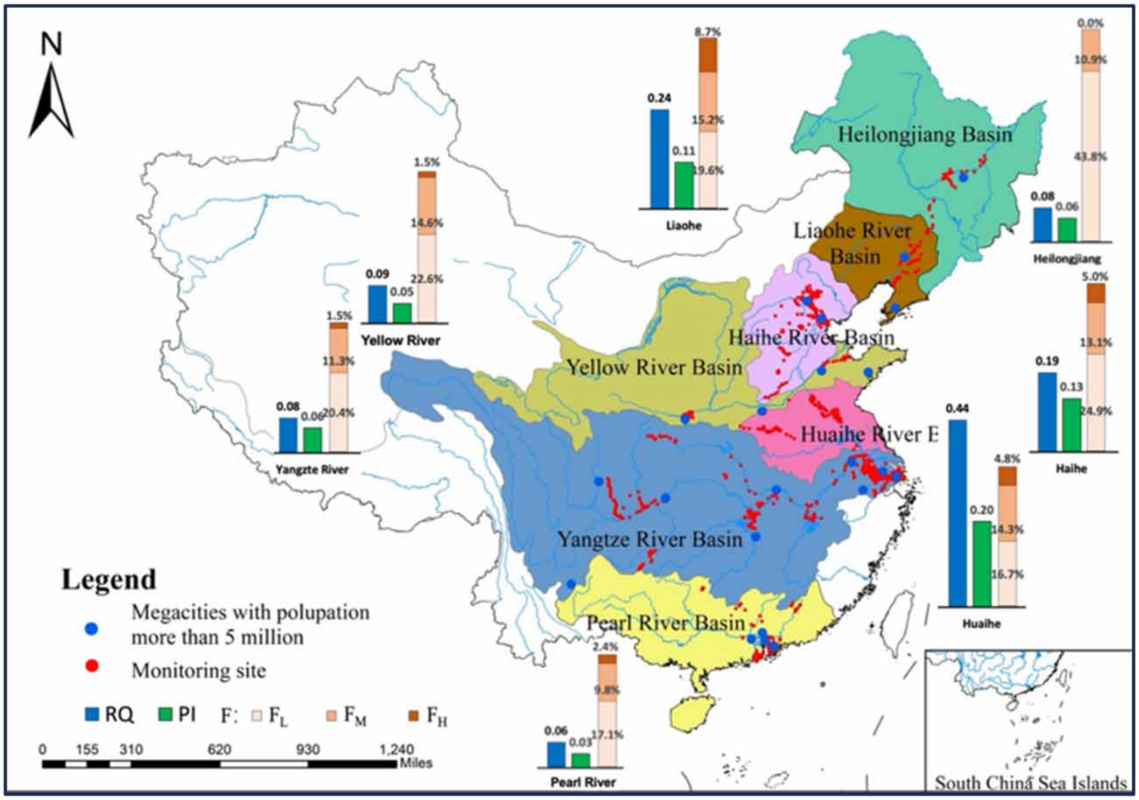

Figure 2 | Results of multilevel ecological risk assessment of seven major basins in China, and monitoring sites distribution.

thus results of this study largely represent the ecological risks of the megacities. Moreover, most of the monitoring sites are located in downstream areas of the main rivers, particularly in the Yellow River Basin and the Pearl River Basin. Therefore, due to the uneven distribution of monitoring sites, there could be significant variations of ecological risk in other regions of the basins, suggesting more surveillance of antibiotics over a broader geographical region. This finding and suggestion have been confirmed by other articles (Li *et al.* 2018; Liu *et al.* 2018). Qiao *et al.* (2018) highly recommended the establishment of a systematic surveillance network that includes regular, continuous measurement of antibiotics (Qiao *et al.* 2018). Therefore, this article strongly urges the necessity of enhancing both monitoring and related ERA to verify and manage the significant risks identified herein.

3.2. Temporal distribution of ecological risk of antibiotic

The temporal variation of average ecological risk values in seven major river basins during the dry and wet periods of the year is presented in Figure 3. Except for the Huaihe River Basin and the Pearl River Basin, the RQ, *F*, and PI values during the dry period in the other basins are higher than in the wet period. In the Liaohe River Basin, especially, the RQ and PI values of dry periods are nearly 10 times higher than wet periods, while around five times for the Haihe River Basin and the Heilongjiang River Basin. Of particular concern is that the RQ_H in the Liaohe River Basin during the dry period is approaching the high-risk range. This finding is observed by other researchers who carried out ERA in the perspective of seasonal variation (Lei *et al.* 2019; Khan *et al.* 2020). The temporal distribution depends on many factors, such as lower surface water volume during the dry period, higher per capita antibiotic usage in the autumn and winter, and lower levels of solar radiation and biological activity in winter leading to antibiotic degradation (Li *et al.* 2019; Zhou *et al.* 2019; Zhang *et al.* 2020). In addition, the prevalence of human antibiotic use in flu season (normally referring to winter) plays an important role in the temporal distribution (Zhang *et al.* 2019). Regarding measures for controlling antibiotics in dry seasons, it is necessary to strengthen public awareness of the rational use of antibiotics and to improve the removal efficiency of antibiotics in the wastewater treatment process (Binh *et al.* 2018). The Pearl River Basin exhibits a relatively small deviation from this pattern, while the Huaihe

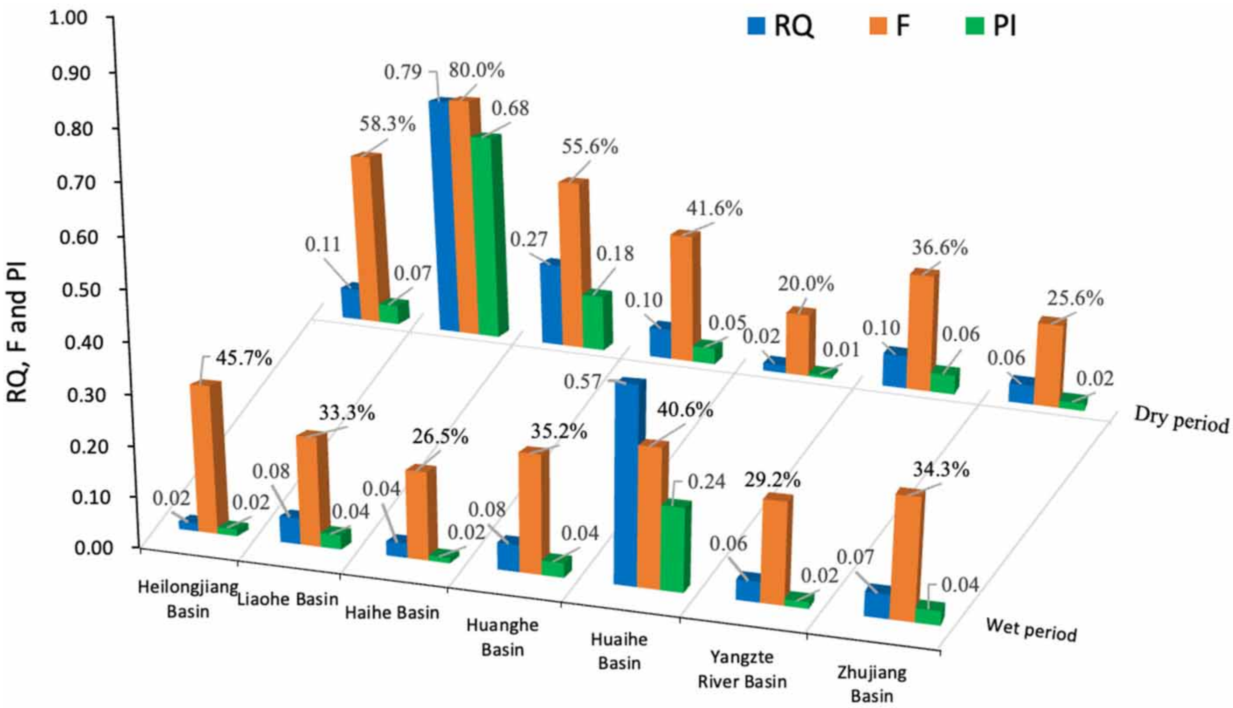

Figure 3 | Temporal distribution of multilevel ecological risk assessment in seven major river basins.

River Basin deviates more significantly in the opposite direction. The opposite temporal distribution was also observed in the articles that focused on fish farming (Wang *et al.* 2018; Ju *et al.* 2023). The higher antibiotic usage in regions with fish farming may result from disease outbreaks at high temperatures, leading to increased consumption of antibiotics to prevent and treat fish diseases during summer. Additionally, the opposite temporal distribution could be due to the previously mentioned factors, such as limited monitoring and monitoring sites concentrated in tributaries. Therefore, this study suggests that monitoring should be strengthened in key periods, especially for the Huaihe basin. Based on more monitoring results, the analysis should be done to find out which factors contribute to the opposite temporal distribution.

3.3. Identification of antibiotics of particular concern

PI is the product of RQ and *F*, depending on not only the risk level but also the occurrence frequency of it. To identify antibiotics of particular concern, the contribution rates of antibiotics to PI are exhibited in Figure 4. The antibiotic order from top to bottom depends on their PI cumulative contribution rate to seven major river basins. Figure 4 includes the top 15 among the 52 antibiotics that this article studied. There are significant differences in antibiotic types and contribution rates among seven major basins. Therefore, it is essential to formulate specific antibiotic monitoring plans and management measures for each basin.

The black-bordered boxes in Figure 4 highlight 10 antibiotics present in both wet and dry periods, underscoring the need for these antibiotics to receive special attention at a national level. Regarding the increase of ranking order from bottom to top, the higher ranking of antibiotics indicates the higher priority of concern at a national scale. There have been nine antibiotics listed in the watch list of substances of 'Union-wide monitoring in the field of water policy pursuant to Directive 2008' since 2015 (European Commission 2022), and seven of them are included in the figure, which are marked by the star in Figure 4. Most of these 10 antibiotics were pointed out by many review papers due to the low removal efficiency in the practical treatment process (Langbehn *et al.* 2021; Ilurdoz *et al.* 2022; Morin-Crini *et al.* 2022). Wastewater treatment is a key point of the fate of antibiotics in the aquatic environment, and the low removal efficiency could explain the high contribution of these antibiotics to river basins (Michael *et al.* 2013; Wang *et al.* 2019).

As Table 1 shows, only a small number of publications are available about monitoring data of the Huaihe River Basin, the Liaohe River Basin, and the Heilongjiang River Basin, thus further monitoring and validation are needed for the abnormally

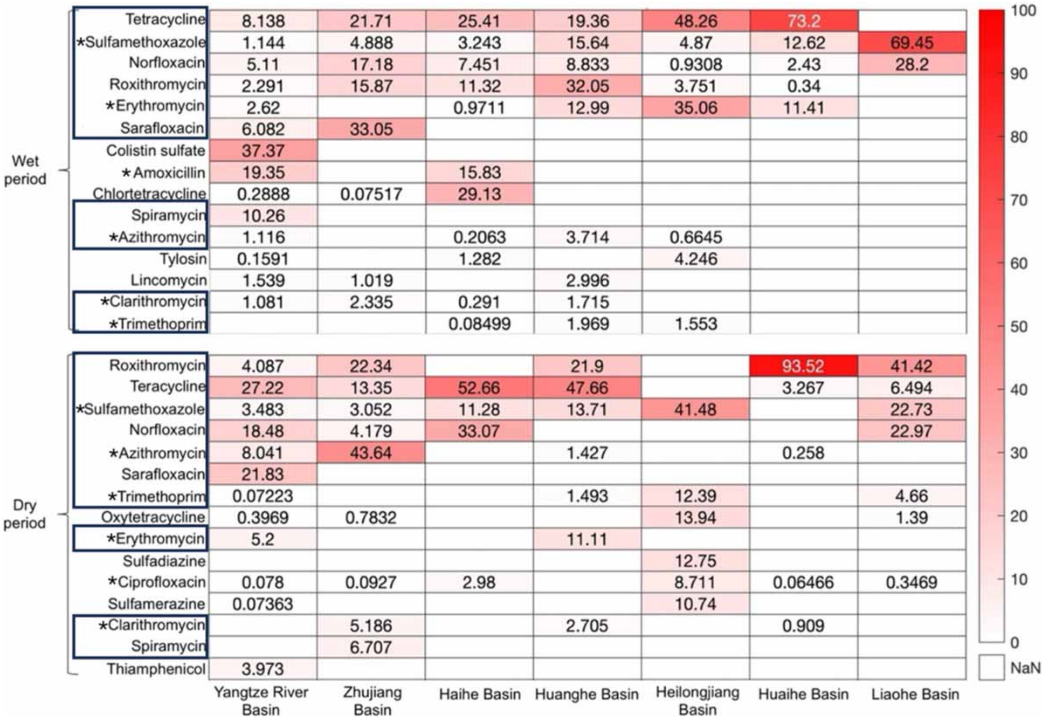

Figure 4 | Thermodynamic chart of the contribution rate of antibiotics to PI in seven major river basins (Unit: %). *Note*: the antibiotics marked by '*' have been listed in the watch list of substances by the European Union.

high contribution rates of tetracycline and roxithromycin. Regarding antibiotics with high contributions, it is necessary to seek alternative drugs with similar therapeutic effects but lower ecological risks.

4. CONCLUSIONS AND RECOMMENDATIONS

The optimized approach with the multilevel risk assessment presents not only the risk values but also the ubiquity of ecological risk levels across a broad study region. Based on the results of ERA by the optimized approach, it is recommended to preferentially strengthen the monitoring of antibiotics and ERA in the Huaihe River Basin, the Haihe River Basin, and the Liaohe River Basin, and to enhance control measures for antibiotic use. Considering seven major river basins cover most parts of China's land area, GDP, and population, the presented results and conclusions mostly stand for the ecological risk of antibiotics at the national scale. Monitoring density of antibiotics is highly uneven across the basins, and future efforts of monitoring of antibiotics should be extended to the upper and middle reaches of the main rivers, as well as other tributary regions.

Since ecological risk values of antibiotics exhibit significant differences between the dry and wet periods, it is advisable to focus on controlling antibiotic usage and improving wastewater treatment for the period with high ecological risk.

Antibiotics of particular concern are identified, which is helpful in formulating a national control list of antibiotics. On the other hand, there are certain differences between the river basins regarding certain types of antibiotics and their contributions to ecological risk, suggesting the formulation of antibiotic risk management on a regional level based on its antibiotic consumption and environmental conditions.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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