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Antibiotics in Coastal Waters of Northern China: Occurrence and Environmental Risk

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Abstract. In recent years, antibiotic pollution has become more serious in the coastal waters of China. This paper summarizes the distribution of antibiotic pollution in the coastal waters of northern China (Bohai Bay, Laizhou Bay, and the northern Yellow Sea coast), calculates and evaluates the environmental risk level. It was shown that 5 types of 29 antibiotics were detected in the coastal waters of northern China, including 13 sulfonamides (SAs), 5 fluoroquinolones (FQs), 5 macrolides(MLs), 4 tetracyclines(TCs), 2 chloramphenicols(CPs) and 1 sulfonamide synergist, with the highest frequency of SDZ and SMZ detection. The average concentrations of antibiotics in Laizhou Bay and Bohai Bay are significantly higher than North Yellow Sea Coast, with a total average of 210.5 ng/L and 180.15 ng/L, respectively. The maximum concentration of antibiotics appeared in Laizhou Bay, with a TPM of 330 ng/L. The Environmental Risk Index shows that in terms of environmental risk, Laizhou Bay>Bohai Bay>North Yellow Sea Coast. SMX has high environmental risks, NOR, ENRO, and RTM have moderate environmental risks.

Introduction

Antibiotics are organic substances that selectively inhibit or affect biological functions at low concentration [1]. Since Fleming first discovered the antibiotic penicillin in 1929 [2], more and more antibiotics have been discovered or synthesized. As an important drug for preventing and treating many common diseases in humans and animals and for promoting animal growth in agricultural aquaculture, it is widely used in the medical industry and animal husbandry. However, they are not completely absorbed by the organism and 90% of the antibiotics are excreted in the form of metabolites through the feces and urine and are directly or indirectly introduced into the natural world. With the increasing use of emerging pollutants such as antibiotics [3], antibiotics widely remaining in nature are attracting more and more attention. These residues will promote the development of antibiotic resistance genes (ARGs) in bacterial populations. Screening has become an emerging pollutant and has led to bacterial antibiotic resistance and potential adverse effects on ecosystem health and food safety [4]. Antibiotic resistance has become a global and imminent health threat. In addition, antibiotics have toxic effects on marine ecosystem animals, 80% of which are toxic to algae, and more than 50% of antibiotics are toxic to fish. Therefore, the treatment of antibiotic pollution is imminent.

China is the world's largest producer and user of antibiotics. In 2013, the total use of antibiotics in China was approximately 162,000 tons, of which 48% were meant for human antibiotics and the rest were veterinary antibiotics. Marine water can be used as the main carrier of antibiotic residues. A large number of antibiotics penetrate the coastal ecosystem through three channels, namely, river input, wastewater discharge from wastewater treatment plants, and direct discharge of untreated wastewater from coastal farming [5]. Therefore, this study aims to evaluate the concentration and distribution of antibiotics in coastal areas of China, and assess and compare the environmental risks of

antibiotics in coastal waters of northern China based on the toxicity of algae, invertebrates and other typical indicator organisms [6].

Materials and Methods

Antibiotic Contamination Data Analysis Method. Through the computer retrieval pubmed, Chinese biomedical literature database and relevant literature traceability and other ways to collect information on the review of antibiotic residues in the water environment and survey literature. Used to assess antibiotic contamination in coastal areas of China.

Environmental Risk Assessment Method. Assessment of the potential environmental risks of antibiotics in the aquatic environment based on the Risk Quotient (RQ) in the European Risk Assessment Technical Guidance Document [7]. The individual RQ was calculated by dividing the measured actual concentration (MEC) of the predicted ineffective concentration (PNEC) (Eq. 1).

$$RQ = MEC/PNEC. \quad (1)$$

PNEC is calculated based on the toxicity data.

The estimated risk level can be divided into four categories, namely no risk (RQ<0.01), low risk (0.01<RQ<0.1), medium risk (0.1<RQ<1) and high risk (RQ>1).

Antibiotic Pollution in Coastal Waters in Northern China

Antibiotic Types. A total of 29 antibiotics were detected in the Bohai Bay [8], Laizhou Bay [9] and North Yellow Sea [10] regions, including 13 sulfonamides(SAs), 5 fluoroquinolones(FQs), 5 macrolides(MLs), 4 tetracyclines(TCs), 2 chloramphenicols(CPs) and 1 sulfonamide synergist(Fig. 1). Among them, SDZ and SMZ have the highest frequency of detection and are detected in all three regions. The next is ETM, NOR, OTC, RTM, STZ, and TC. Most of the antibiotics were detected in Bohai Bay, including SAs, TCs, MLs, and FQs. The number of antibiotics detected the North Yellow Sea coast was the largest, among which SAs were the most, accounting for 64.7%.

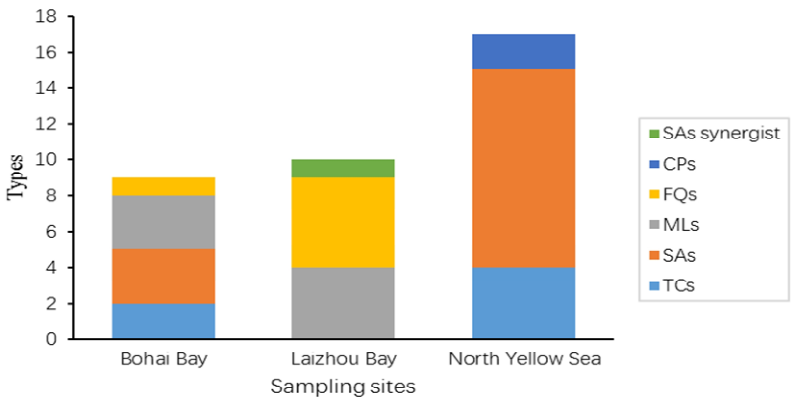

Fig. 1 Types of antibiotics in coastal areas in northern China

Antibiotic Concentration. The total concentration of antibiotics in the Bohai Bay, Laizhou Bay and the Northern Yellow Sea is as high as 400.68 ng/L. The highest concentration of antibiotics in Laizhou Bay reached 210.5 ng/L, followed by Bohai Bay (180.15 ng/L), and the concentration of antibiotics in the northern Yellow Sea was the lowest, only 10 ng/L. The maximum values of TMP and ENO in the Laizhou Bay area were as high as 330 ng/L and 209 ng/L, respectively. The maximum concentrations of TC and RTM in Bohai Bay were as high as 98.31 ng/L and 96.31 ng/L, respectively. The reason for this phenomenon is assumed to be as follows: First, Bohai Bay and Laizhou Bay are located within the Bohai Sea. The Bohai Sea is a closed inland sea, where water-phase interactions are

slow, and antibiotics do not easily extend to cause accumulation of antibiotics. Yellow Sea as an open sea, antibiotics easily spread, resulting in lower antibiotic concentrations. Second, there are numerous rivers that enter the sea upstream in the Bohai Bay and Laizhou Bay areas, carrying large amounts of antibiotics into the sea and accumulating in coastal areas. Third, the surrounding cities of Bohai Bay and Laizhou Bay are dense and have a larger population. Therefore, a large number of therapeutic antibiotics indirectly enter the coastal waters through the sewage system. Fourth, coastal aquaculture in the Bohai Bay and Laizhou Bay areas is relatively intensive, and a large number of antibiotics injected into the coastal areas are discharged without treatment, resulting in high concentrations of antibiotics in the area.

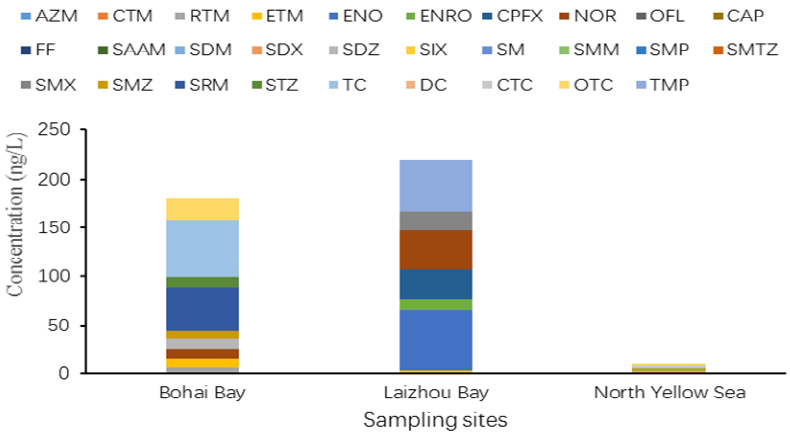

Fig. 2 Antibiotic concentration in coastal waters in northern China (ng/L)

Environmental Risk in Coastal Waters in Northern China

The environmental risk assessment of antibiotics adopts the EU environmental risk assessment method, namely RQ = MEC/PNEC. Based on the maximum impact considerations, the RQ values calculation uses the PNEC that screens the most sensitive species and calculates the maximum concentration of antibiotics. The results are shown in Table 1.

The RQ values shown in Table 1, it can be seen that Laizhou Bay> Bohai Bay> North Yellow Sea Coast, where SMX has the highest RQ value, indicating that it has high risk for algae, Laizhou Bay Medium NOR and ENRO have moderate environmental risks, RTM has a low environment risk. The NOR and RTM in the Bohai Bay have moderate environmental risks, while the TC, OTC and SDZ have low environmental risks, and the low concentrations of antibiotics in the northern Yellow Sea coastal areas are risk-free.

Table. 1 RQ of antibiotics in coastal waters in northern China

	Non-target organism	Toxicity	PNEC ng/L	Bohai Bay	Laizhou Bay	North Yellow Sea
TMP	*R. salina*	Acute	16,000	0	0.020625	0
SMX	*S. leopoliensis*	Acute	27	0	3.037037	0
SMZ	*Lemna minor*	Acute	17400	0.001002	0.0000862	0.0001282
SDZ	*S. capricornutum*	Acute	2200	0.033136	0.000195	0.0009318
RTM	*P. subcapitata*	Chronic	100	0.9804	0.015	0
TC	*P.subcapitata*	Acute	3310	0.029701	0	0.0011541
OTC	*P.subcapitata*	Acute	1040	0.08126	0	0.0060385
NOR	*V.ficheri*	Chronic	103.8	0.773699	0.992293	0
ENRO	*V.ficheri*	Chronic	28.8	0	0.263889	0

Conclusions

The contamination of antibiotics in the coastal waters of northern China is relatively complex. A total of 29 antibiotics were detected, including 13 sulfonamides(SAs), 5 fluoroquinolones(FQs), 5 macrolides(MLs), 4 tetracyclines(TCs), 2 chloramphenicols(CPs) and 1 sulfonamide synergist. SDZ and SMZ have the highest detection frequency, followed by ETM, NOR, OTC, RTM, STZ, and TC.

The antibiotic pollution in the coastal waters of northern China is more serious. The concentration of antibiotics in Laizhou Bay and Bohai Bay in the Bohai Sea is relatively high. The total average is as high as 330ng/L and 209ng/L, respectively. The maximum TPM in Laizhou Bay is as high as 330ng/L.

The environmental risk of antibiotics in the coastal waters of northern China is relatively high, and the environmental risk assessment index as a whole is Laizhou Bay> Bohai Bay> North Yellow Sea Coast. Among them, SMX has high environmental risk, and NOR, ENRO, and RTM have moderate environmental risks. TC, OTC and SDZ have low environmental risks.

The above analysis shows that the status of antibiotic pollution in the coastal waters of northern China is relatively serious, and the Bohai region has a high concentration of antibiotics and is relatively high due to the fact that it is a closed inland sea, and there are many rivers flowing into the sea, and the resident population is dense. Environmental risks pose a certain potential threat to human health. Therefore, we need to control the use of antibiotics in coastal areas and adopt rational and effective antibiotic degradation methods.

Acknowledgements

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References

[1] A.K. Sarmah, M.T. Meyer, A.B.A. Boxall: Chemosphere. Vol. 65 (2006), p. 725–759.

[2] A. Fleming: Exp. Pathol. Vol. 10 (1929), p. 226–236.

[3] K. Eguchi, H. Nagase, M. Ozawa, Y.S. Endoh, K. Goto, K. Hirata, K. Miyamoto, H. Yoshimura: Chemosphere. Vol. 57(2007), p. 1733-1738.

[4] A.B.A. Boxall, D.W. Kolpin, B. Halling, J. Tolls: Environ. Sci. Technol. Vol. 37 (2003), p. 286–294.

[5] P.K Mutiyar, A.K. Mittal: Environ. Sci. Pollut. Res. Vol. 21 (2014), p. 7723–7736.

[6] M.D. Hernando, M. Mezcua, A.R. Fernandez, D. Barcelo: Talanta. Vol. 69 (2006), p.334–342.

[7] S. Li, W.Z. Shi, W. Liu, H.M. Li, W. Zhang, J.R. Hu, Y.C. Ke, W.L. Sun, J.R. Ni: Science of the Total Environment. Vol. 615 (2018) , p. 906–917

[8] D.M. Cheng, Y.J. Xie, Y.J. Yu, X.H. Liu, S.N. Zhao, B.S. Cui, J.H. Bai: Wetlands. Vol. 36 Suppl 1 (2016), p. S167–S179

[9] R. Zhang, G. Zhang, Q. Zheng, J. Tang, Y. Chen, W. Xu, Y. Zhou, X. Chen: Ecotoxicology and Environmental Safety Vol. 80 (2012b), p. 208-215.

[10] G.S. Na, X.D. Fang, Y.Q. Cai, L.K. Ge, H.M. Zong, X.T. Yuan, Z.W. Yao, Z.F. Zhang: Marine Pollution Bulletin. Vol. 69 (2013), p. 233–237