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LETTER

WATER POLLUTION AND AGRICULTURE: MULTI-POLLUTANT PERSPECTIVES

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KEYWORDS

water quality, agriculture, multi-pollutant assessment, hotspots, interactions

HIGHLIGHTS

- Four highlights are identified for agriculture and water from the multi-pollutant perspective.
- Large variations in time and space for multiple pollutants in waters and their sources.
- Scientific agenda should account for multiple pollutants in agricultural strategies.

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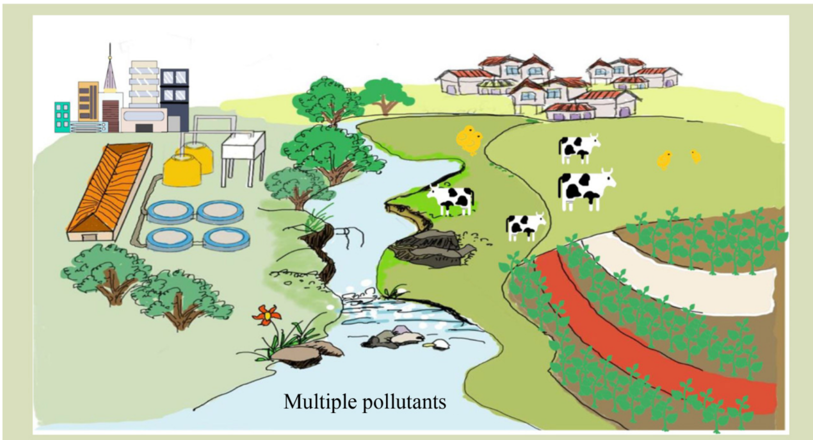
GRAPHICAL ABSTRACT



ABSTRACT

Agriculture is an important cause of multiple pollutants in water. With population growth and increasing food demand, more nutrients, plastics, pesticides, pathogens and antibiotics are expected to enter water systems in the 21st century. As a result, water science has been shifting from single-pollutant to multi-pollutant perspectives for large-scale water quality assessments. This perspective paper summarizes and discusses four main highlights related to water pollution and agriculture from the multi-pollutant perspective. These highlights reveal the spatial and temporal distribution and main sources of multiple pollutants in waters. Based on the highlights, a scientific agenda is proposed to prioritize solutions for sustainable agriculture (UN Sustainable Development Goal 2) and clean water (UN Sustainable Development Goals 6 and 14). This agenda points out that when formulating solutions for water pollution, it is essential to take into account multiple pollutants and their interactions beyond biogeochemistry.

1 INTRODUCTION

Today, water quality, when influenced by multiple pollutants, remains poorly understood, especially at large scales such as national and global[1]. In the past, water pollution issues were often related to nutrients and eutrophication[2,3]. Today, it is different[4,5]. With the increase in agricultural activities and technological developments, water systems experience new challenges associated with emerging pollutants such as plastics, antibiotics and chemicals[6–12] (Fig. 1). Climate change[16–19] and unforeseen crises (e.g., COVID-19)[10] accelerate these challenges[20,21]. For example, the intensified agricultural activities are one of the main sources that release various pollutants to water systems, such as nutrients (e.g., used as fertilizers for crop production), plastics (e.g., used for crop mulching), antibiotics (e.g., used for animal disease protection), pathogens (e.g., via animal manure) and pesticides (e.g., used for crop protection). As a result, many water systems such as groundwater[22,23], rivers[2], lakes[24,25] and coastal seas[26] are more polluted than in the past. The urgency is clear; water science must shift from a single-pollutant toward a multi-pollutant focus[4,5,20]. In this perspective paper, we summarize and discuss four highlights related to water pollution and agriculture. Our highlights focus on (1) reasons that call for more focus on multi-pollutant water pollution issues, (2) the spatial and temporal distribution of multi-pollutant hotspots, (3) agriculture as an important source of multiple pollutants, and (4) interactions of multiple pollutants in water systems beyond biogeochemistry. These highlights are supported by literature and the results of the MARINA (Models for Assessing River Inputs of pollutaNts to seAs) model family on multi-pollutant issues. We provide examples on water pollution issues worldwide and for China. Based on the discussed highlights, we set a scientific agenda for water research in exploring solutions for future water pollution control.

2 FOUR HIGHLIGHTS

2.1 From single- to multi-pollutant assessments

Over the last several decades, water science has been changing its focus, from single-pollutant to multiple-pollutant assessments of water quality. We identify five main reasons that underline this shift.

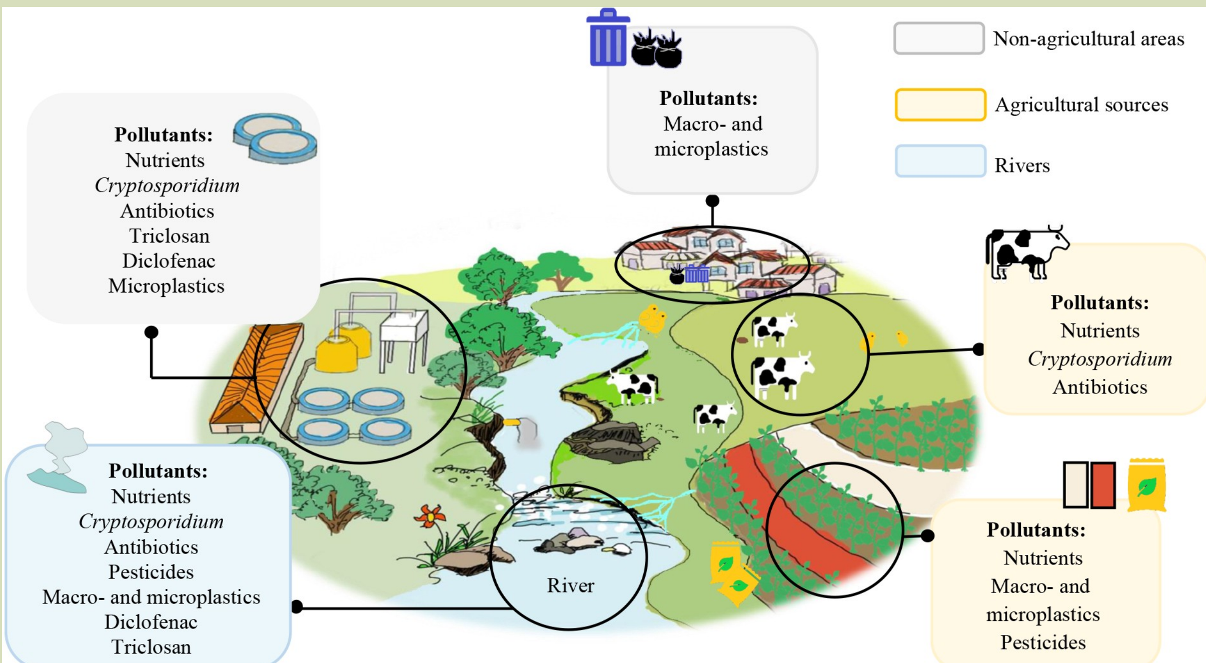


Fig. 1 Concept model of multiple pollutant inputs to rivers from agricultural sources and non-agricultural areas. This concept model includes pollutants for which global or regional modeling approaches exist. Table 1 summarizes the information from conceptual modeling approaches. Agricultural sources include pollutants from crop production and livestock production (yellow box). Pollution sources from non-agricultural areas include pollutants from mismanaged plastic waste and sewage systems (gray box). The blue box indicates all pollutants from agriculture sources and non-agricultural areas enter rivers. Sources: MARINA models (Model to Assess River Inputs of pollutaNts to seAs)[2,13–15].

The first reason is that increasing amounts of emerging pollutants are being released into water systems in many places worldwide. Emerging pollutants are defined as those that have adverse impacts on nature and society, and for which monitoring programs are typically not designed[27]. According to the NORMAN Substance Database, more than 700 substances have been identified. These pollutants are often not included in the monitoring programs but pose additional stress on water systems. Modeling tools are used to estimate pollution levels of some of the emerging pollutants. Examples are microplastics released from sewage systems into rivers and seas that are expected to continue because of urbanization activities (e.g., more people consuming more plastic products). A recent paper[28] reported that almost 40% of the river sub-basins worldwide are dominated by microplastics from sewage systems. This includes the European rivers draining into the Black Sea that contribute today over half of the microplastics in the sea[29]. Another example is microplastics in Chinese rivers from crop production (mulching and greenhouses) and sewage systems[13]. All this highlights a need for a better understanding of pollution levels of emerging pollutants and their causes.

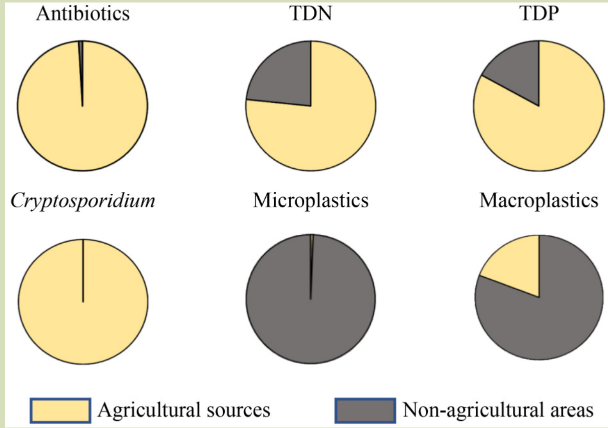


Fig. 2 Multi-pollutant problems in river sub-basins associated with annual inputs of more than one pollutant to rivers from agricultural sources and non-agricultural areas in China in the 2010s. This figure shows the percentages of multiple pollutants to Chinese rivers from agricultural sources and non-agricultural areas, The data presented covers results from the MARINA models for 2013 and 2015. The annual inputs of pollutants to rivers are derived from the MARINA-Multi, MARINA-Nutrients, MARINA-Antibiotics and MARINA-Plastics[2,13–15]. Six pollutants are considered: antibiotics, *Cryptosporidium*, diclofenac, triclosan, microplastics, macroplastics, total dissolved phosphorus (TDP), and total dissolved nitrogen (TDN).

The second reason is that multi-pollutant problems spread worldwide in many river basins. Globally, more water systems become contaminated with multiple pollutants. Thus, multi-pollutant problems have already become a global issue[2,4,13,14,28]. Several of the pollutants in water have already been reported on larger scales (continental and global). This holds for microplastics[30] and triclosan[12] from personal care products, diclofenac (pharmaceutical)[31], macroplastics[26], antibiotics (drugs)[32] and pesticides (biocides)[33]. Thus, multi-pollutant problems become large-scale issues in the 21st century[2]. As a result, the number of multi-pollutant studies has also been increasing for continental and global analyses[20,34] (examples are given in Fig. 2).

The third reason is that many pollutants enter waters from the same sources. Agriculture takes an important share here (Table 1, Fig. 1 and Fig. 2). For example, more than three-quarters of nitrogen, phosphorus, and oocysts of *Cryptosporidium* (pathogen) entered rivers from pig, cattle and chicken manure that was applied to grow crops in 2010 globally[14]. Crop production can be a source of not only nutrients in waters[19,35], but also macro- and microplastics through mulching activities and greenhouses as well as pesticides through their spray on cropland (Table 1 and Fig. 2). In addition, antibiotics are often used in animal feed[36,37]. As a result, manure can also contain those antibiotics. Therefore, the use of manure to grow crops can pollute waters with

Table 1 Examples summarize the importance of agricultural activities in water pollution control, focusing on multi-pollutant perspectives

	N	P	*Cryptosporidium*	Triclosan	Diclofenac	Antibiotics	Plastics	Pesticides
Crop production	X	X	X	–	–	X	X	X
Animal production	X	X	X	–	–	X	–	–
Sewage	X	X	X	X	X	X	X	
Model version	a, b		b, c	c,d	d	e	d, f	g

Note: a, MARINA-Nutrients (Global-2.0); b, MARINA-Multi (Global-2.0); c, MARINA-Multi (Global-1.0); d, MARINA-Multi (Global-3.0); e, MARINA-Antibiotics (China-1.0); f, MARINA-Plastics (China-1.0); g, MARINA-Pesticides (Global 1.0). X indicates the source of pollutants in rivers that are considered in the existing versions of MARINA models. Sources: MARINA models (Model to Assess River Inputs of pollutaNts to seA)[2,13–15].

antibiotics[38,39] (Table 1, Fig. 2 and examples in Section 2.3). In addition, increasing consumption of personal products, medicines, and plastics due to urbanization has made cities a common source of multiple pollutants to waters[2]. As a result, water systems become cocktails of pollutants from both agricultural (i.e., animal antibiotics, nutrients, pathogens, pesticides and plastics)[13,40,41] and urban (i.e., human antibiotics, nutrients, painkillers, personal care products and plastics) activities[2,14,15].

The fourth reason is that multiple pollutants could lead to multiple impacts on society and nature. It is well known that accumulation of nutrients in water bodies can lead to eutrophication, which triggers harmful algal blooms, and creates dead zones in aquatic environments. While individual impacts of some pollutants are understood, more pollutants in water could likely increase the stress level on aquatic organisms and make water unsuitable for human needs. However, such a field is still very limited in scientific evidence and needs more attention. We need to better understand interactions between pollutants for impacts (Section 2.4).

The fifth reason is that global multi-pollutant water quality models have been developed in recent years to support multi-pollutant assessments of water quality. In the past, water quality models were developed with a large focus on single pollution type. Models[35,42–44] can provide insights into what pollutants exist in water, why and where, and how to reduce them. Recently, more scientific attention is on developing multi-pollutant water quality models to better understand multi-pollutant hotspots and their causes. The MARINA models are promising examples and have been developed to address multi-pollutant problems, their sources and explore future trends (Fig. 1 and Table 1). The models are largely process-based and oriented to scenarios. The MARINA models (Table 1) integrate existing approaches for nutrients[14], macroplastics[13,15], microplastics[2,13,15], *Cryptosporidium* (pathogen)[2,14], triclosan (chemical)[2,15], diclofenac[15], antibiotics and pesticides. The model runs at the sub-basin scale for rivers worldwide. Recent developments in this multi-pollutant modeling approach allow for a better understanding of the causes of multiple pollutants in waters (Section 2.2), their hotspots (Section 2.3), and how to reduce them (Section 2.4) (Fig. 1 and Fig. 2).

2.2 Spatial and temporal distribution of multi-pollutant hotspots

We discuss this highlight using examples from the MARINA models. Current multi-pollutant hotspots are mainly distributed in south Asia, Europe, and parts of North America, as illustrated by MARINA studies of inputs of eight pollutants to global sub-basins[2,14,15,29]. These eight pollutants are antibiotics, *Cryptosporidium,* diclofenac, microplastics, macroplastics, nitrogen, phosphorous, and triclosan can impact both societies (e.g., diarrhea) and nature (e.g., eutrophication). Hotspots are sub-basins with more than four pollutants exceeding the global average levels. These studies basins[2,13–15,29] found that in 2010, nearly 10% of the global sub-basins (1072 of 10,226) will likely face water pollution problems caused by more than four pollutants (Table 2). These hotspots cover 14% of the total global sub-basin area but support 67% of the global population (Table 2).

The multi-pollutant issue will likely increase in the future and expand to other world regions. For example, Africa is becoming a hotspot for multi-pollutant water pollution, since African surface waters are being polluted by at least two contaminants[2,15]. The Congo and Niger Rivers are particularly polluted for more than four pollutants (i.e., nitrogen, phosphorus, micro and macroplastics), despite some sub-basin areas of the Nile River being cleaner than in 2010. In sub-basins of India, the Ganges and Indus Rivers will remain particularly polluted. The same holds for Chinese sub-basins

Table 2 Characteristics of sub-basins that receive inputs of more than four pollutants to rivers globally

Characteristics	Polluted sub-basins	Percentages of the national value
Number of sub-basins	1072	11
Sub-basin area (million km^2)	20	14
Total population (billion people per year)	5	67
Population connected to sewage systems (billion people per year)	2	79

Note: The global average was estimated by dividing the global input of pollutants to rivers from agricultural sources and non-agricultural areas by the global surface area in 2010. Polluted sub-basins are defined as the annual input of pollutants into the river that exceeds the global average. The annual inputs of pollutants to rivers are derived from the MARINA-Multi, MARINA-Nutrients, MARINA-Antibiotics and MARINA-Plastics[2,13–15].

(Hai, Yangtze and Pearl Rivers). Around 59 sub-basins in China are identified as particularly polluted sub-basins, which exceed the national average for at least four out of the six pollutants (i.e., total dissolved nitrogen, total dissolved phosphorous, microplastics, macroplastics, *Cryptosporidium* and antibiotics), account for 23% of the total area of the sub-basins and accommodate 72% of the Chinese population (Table 3)[2,14,15]. By the end of the century, about half (51%) of the global population will be at risk of severe water contamination due to 5-6 pollutants, including nutrients, plastics and chemical pollution.

2.3 The importance of agricultural sources in multi-pollutant problems

Agriculture is expected to be an important source of multiple pollutants in water systems as a result of population growth, increasing food demand, and climate change. Below, we first discuss examples of pollution sources worldwide. Then, we zoom into China as an interesting case with intensive agricultural production.

Expansion of agricultural land, intensive livestock production and increases in agricultural materials inputs are expected to lead to more chemicals (e.g., antibiotics[45], pesticides[40], and plastics[46]), nutrients[35], and pathogens[43,47] in rivers from crop production and livestock production in the future. For the world, without any proactive actions, plastic pollution will only become worse, both in terms of microplastics (e.g., Africa, Asia, Europe and North America) and macroplastics (e.g., Africa, Australia and Asia)[28]. For many African basins, river pollution is projected to be by 11–18 times higher in the end of the 21st century compared to the level of 2010 for nutrients, microplastics, *Cryptosporidium* and triclosan from cities[2]. Also, globally, agricultural activities may contribute largely to nitrogen loadings to rivers, whereas human waste from sewage systems is expected to be an important source of phosphorus[35]. Climate change may also accelerate water quality degradation and amplify the difficulties caused by multiple pollutants[18].

China is an interesting case, in this context, due to its large and intensive agricultural production, and the resulting multi-pollutant water pollution issue. In China, agricultural production contributed more than 75% of antibiotics, *Cryptosporidium*, total dissolved nitrogen and total dissolved phosphorus in all rivers in the 2010s (Fig. 2). This means that compared to other pollutants, agricultural sources are more important for river pollution than non-agricultural areas. The application of synthetic fertilizer and animal manure on agricultural land, and the use of plastic films in food production are the dominant sources of agricultural water pollution in China. MARINA-Multi (China 1.0) projected an over 45% increase in inputs of total dissolved nitrogen, total dissolved phosphorus and *Cryptosporidium* to the central, eastern and southern sub-basins of China from agriculture by 2050. An increase of 16% in plastics in rivers from crop production is expected by 2050. Thus, strategies for water pollution reduction in China must consider improved management of pollutant compounds from agriculture.

2.4 Interactions beyond biogeochemistry

This shift of water science to multi-pollutant perspectives also reveals challenges in developing solutions for water pollution control. This is associated with the large diversity in interactions between multiple pollutants across spatial and temporal scales[48,49]. Today, when exploring solutions, the focus is often on single pollutants[29,50,51]. By taking multi-pollutant perspectives, existing science tends to consider biogeochemical interactions[48,49,52]. The interactions occur when two or more substances (i.e., pollutants) from different pollutant groups (e.g., pathogens and plastics) affect each other. These interactions are most uncertain because of the

Table 3 Characteristics of sub-basins that receive inputs of more than four pollutants to rivers in China

Characteristics	Polluted sub-basins	Percentages of the national value
Number of sub-basins	59	15
Sub-basin area (million km^2)	3.4	23
Total population (million people per year)	981	72
Population connected to sewage systems (million people per year)	541	79

Note: National average was estimated as the division of pollutants input to rivers from agricultural sources and non-agricultural areas in China by national area in 2013 and 2015. Polluted sub-basins are defined as the annual input of pollutants into the river that exceeds the national average. The annual inputs of pollutants to rivers are derived from the MARINA-Multi, MARINA-Nutrients, MARINA-Antibiotics and MARINA-Plastics[2,13–15].

diversity in pollutants and their behaviors. The extent to which the interactions occur may vary among world regions due to different multi-pollutant combinations present in waters, climate change (e.g., temperature and hydrology), and sub-basin characteristics (e.g., human activities). Examples are interactions dependent on biofouling[53–56], sorption[57–59], toxic stress[60], supply[61,62] and leaching[63,64]. These biogeochemical interactions are important to understand the impacts of water pollution on nature.

We argue, however, that effective water solutions of the 21st century should go beyond biogeochemistry and consider interactions in the whole cause-effect chain. This includes societal, biogeochemical and impact-related interactions. The most relevant interactions are between population growth, economic development, and urban and rural areas. These interactions influence population prosperity (interactions between people and economy), food production (rural-urban), and sustainability (innovative technologies). Typically, these interactions are embedded in scenario developments and the UN Sustainable Development Goals (SDGs). Examples are Millennium Ecosystem Assessment[65,66] and Shared Socioeconomic Pathways (SSPs)[67–70]. The study of[3] identified 319 interactions between SDGs for clean water (SDGs 6 and 14) and other SDGs, of which 286 are positive (synergies) and 33 are negative (trade-offs) interactions. For example, water pollution control by reducing nutrient use in agriculture may reduce crop production and thus negatively influence the achievement of SDG 2 for food security. They conclude that it is essential to account for these interactions when developing water pollution control strategies to avoid negative impacts on other SDGs for society.

3 OUTLOOK

The four highlights presented recent findings in shifting water science from single-pollutant to multi-pollutant water quality assessments. Based on these highlights, we propose a scientific agenda to prioritize solutions for sustainable agriculture (SDG 2) and clean water (SDGs 6 and 14). Two priority areas to explore for sustainable agricultural management are (1) to address the multiple pollutants, and (2) to account for their interactions in the cause-effect chain.

The priority calls for future research to develop improved agricultural management to reduce emissions of multiple pollutants to water systems. In the 21st century, many emerging agricultural pollutants will cause water pollution worldwide. Today, solutions mainly focus on the management of nitrogen and phosphorus (e.g., fertilizer management, and manure management and treatment). Future research is needed to deal with the diversity in new pollutants. We need to better understand what technologies are available to address emerging pollutants such as antibiotics, pathogens, pesticides and plastics, and the impacts of these technologies on crop yield or animal production. It is also important to have more knowledge on the synergetic technologies that may have the potential to reduce multiple pollutants simultaneously (e.g., manure treatment for nutrients and pathogens), and to save labor and financial costs in agriculture.

The second priority calls for new knowledge and approaches that take into account interactions for multiple pollutants in the cause-effect chain. We emphasized that solutions for agricultural water pollution control should account for interactions beyond biogeochemistry (e.g., interactions between two or more pollutants in water, soil and sediments). The interactions between pollutants are diverse and vary largely across temporal and spatial scales. Often, existing science focuses on biogeochemical interactions. Future research needs to account for interactions in the whole cause-effect chain (e.g., between and within drivers, sources and impacts). This is essential when searching for effective solutions and will most likely require both single- and multidisciplinary research because emerging pollutants may need specialized expertise.

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Compliance with ethics guidelines

Mengru Wang, Qi Zhang, Yanan Li, Mirjam P. Bak, Sijie Feng, Carolien Kroeze, Fanlei Meng, Ilaria Micella, Vita Strokal, Aslıhan Ural-Janssen, and Maryna Strokal declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies with human or animal subjects performed by any of the authors.

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