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Multi-pollutant assessment of river pollution from livestock production worldwide

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ABSTRACT

Livestock production is often a source of multiple pollutants in rivers. However, current assessments of water pollution seldomly take a multi-pollutant perspective, while this is essential for improving water quality. This study quantifies inputs of multiple pollutants to rivers from livestock production worldwide, by animal types and spatially explicit. We focus on nitrogen (N), phosphorus (P), and *Cryptosporidium* (pathogen). We developed the MARINA-Global-L (**M**odel to **A**ssess **R**iver **I**nputs of polluta**N**ts to se**A**s for **L**ivetsock) model for 10,226 sub-basins and eleven livestock species. Global inputs to land from livestock are around 94 Tg N, 19 Tg P, and 2.9×10^{21} oocysts from *Cryptosporidium* in 2010. Over 57% of these amounts are from grazed animals. Asia, South America, and Africa account for over 68% of these amounts on land. The inputs to rivers are around 22 Tg Total Dissolved Nitrogen (TDN), 1.8 Tg Total Dissolved P (TDP), and 1.3×10^{21} oocysts in 2010. Cattle, pigs, and chickens are responsible for 74–88% of these pollutants in rivers. One-fourth of the global sub-basins can be considered pollution hotspots and contribute 71–95% to the TDN, TDP, and oocysts in rivers. Our study could contribute to effective manure management for individual livestock species in sub-basins to reduce multiple pollutants in rivers.

1. Introduction

Livestock production feeds one billion people globally (Sakadevan and Nguyen, 2017). However, intensified practices often pollute rivers with multiple substances including nitrogen (N), phosphorus (P), and pathogens (e.g., *Cryptosporidium*). Livestock provides manure for crop production. Manure contains nutrients (e.g., N, P) but often also pathogens such as oocysts of *Cryptosporidium*. These nutrients and oocysts can enter rivers through surface runoff from fertilized crop fields or direct discharges of manure (only for Chinese rivers) (Strokal et al., 2019; Vermeulen et al., 2019). With the population growth and changing diets, livestock production will increase to satisfy the global food demand (Herrero et al., 2016; Ilea, 2009). This may increase levels of multiple pollutants in rivers from livestock production in the future.

Currently, around two billion people in the world use contaminated water (WHO, 2019). In many countries, rivers contain nutrients (Strokal et al., 2019) and pathogens such as *Cryptosporidium* (Vermeulen et al., 2019). Excess nutrients cause eutrophication problems in many lakes, reservoirs, and coastal waters (Breitburg et al., 2018; Strokal et al., 2021a; Strokal and Kroeze, 2020). For instance, 78% of the coastal waters of the United States suffer from eutrophication (Malone and Newton, 2020) leading to harmful algae blooms, hypoxia (less oxygen), and loss of aquatic biodiversity (Gobler et al., 2017; Howard et al., 2017; Malone and Newton, 2020). *Cryptosporidium* is a protozoan parasite and can be found in infected human and animal feces (Vermeulen et al., 2017; Robertson et al., 2014). *Cryptosporidium* contamination occurs in many countries across the world (Bautista et al., 2018; Krumkamp et al., 2020). For instance, oocysts have been detected in surface and groundwater for domestic water use in Haiti (Balthazard-Accou et al., 2020). This situation leads to water-borne diarrhea that threatens public

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health, especially for young children (Krumkamp et al., 2020; Vermeulen et al., 2019).

Current assessments of water pollution from livestock production hardly take a multi-pollutant perspective. Existing studies focus either on nutrients (Beusen et al., 2015) or pathogens (Vermeulen et al., 2019), but not on both simultaneously. We observe two important knowledge gaps. First, the contribution of individual livestock species to river pollution with multiple pollutants is not well studied especially for sub-basins in the world. Second, there is a need for multi-pollutant assessments to evaluate effective pollution reduction options (Li et al., 2019). Reducing one pollutant from livestock may decrease (synergy) or increase (trade-off) other pollutants in rivers. A multi-pollutant assessments can help to account for synergies and trade-offs between pollutants in water pollution control and support the achievement of Sustainable Development Goals (SDG) 6 “Clean Water and Sanitation” and SDG 2 “Food security” (Mehrabi et al., 2020).

Existing global models quantify flows of pollutants from land to rivers, and focus largely on single groups of pollutants (Beusen et al., 2015; Vermeulen et al., 2017; Wang et al., 2020). The GloWPa-Crypto C1 (**glo**bal **w**aterborne **pa**thogen-***Crypto****sporidium* **C**oncentration) model quantifies oocyst concentrations in rivers from livestock and humans at 0.5° grid cell (Vermeulen et al., 2019). MARINA (Model to Assess River Inputs of pollutaNts to seAs) is another model for river pollution (Strokal et al., 2016). The first version of the MARINA model was developed for China to quantify river export of nutrients at the sub-basin scale (Strokal et al., 2016). MARINA has been updated with more pollutants and expanded to 10,226 sub-basins worldwide, resulting in a global version (MARINA-Global) (Strokal et al., 2021a). The MARINA-Global model includes nutrients and oocysts and integrates the approach of the GloWPa-Crypto model for human waste (Strokal et al., 2019; Strokal et al., 2021a). MARINA-Global does not yet model pollutants from livestock and does not distinguish the contribution of individual livestock species to river pollution. Moreover, a spatially and temporally consistent dataset for over ten thousand sub-basins and multiple pollutants in rivers from livestock production is still lacking.

This study aims to quantify inputs of multiple pollutants to rivers from livestock production worldwide, by animal types and spatially explicit. We focus on DIN (Dissolved Inorganic Nitrogen), DIP (Dissolved Inorganic Phosphorus), DON (Dissolved Organic Nitrogen), DOP (Dissolved Organic Phosphorus), and oocysts from *Cryptosporidium*.

2. Material and methods

2.1. MARINA-Global-L model

We develop the MARINA-Global-L model: **M**odel to **A**ssess **R**iver **I**nputs of pollut**N**ts to se**A** for **L**ivestock (Fig. 1). It quantifies annual inputs of nutrients and oocysts to rivers from individual livestock species in 10,226 sub-basins for 2010. This number of sub-basins is larger than in earlier studies. For example, Mayorga et al. (2010) focused on around 6000 basins. In our study, large basins are split into sub-basins (details are in Strokal et al., 2021a). In total, it results in 10,226 sub-basins in the world. Oocysts of *Cryptosporidium* in our MARINA-Global-L model are examples of pathogens. MARINA-Global-L is based on the existing MARINA-Global model (Strokal et al., 2021a), but integrates the three modeling approaches for livestock: the GloWPa-Crypto L1 (**Glo**bal **W**aterborne **Pa**thogen-***Crypto****sporidium* from **L**ivestock) and GloWPa-Crypto C1 model (**Glo**bal **W**aterborne **Pa**thogen-***Crypto****sporidium* **C**oncentration) for oocysts (Vermeulen et al., 2017, 2019) and the MARINA modeling approach of Chen et al. (resubmission) for nutrients. The MARINA-Global model and GloWPa models are described in Supplementary Text 1 and Text 2.

The MARINA-Global-L model accounts for eleven livestock species following the GloWPa-Crypto L1 model (Vermeulen et al., 2017). These are buffaloes, camels, cattle, chickens, ducks, donkeys, goats, horses, mules, pigs, and sheep. The model distinguishes the two main systems of livestock manure management: grazing and storage. In the grazing systems, animals drop their manure directly on land. In the storage systems, manure is collected and stored before application on land. We account for losses of nutrients and oocysts during storage before manure application. These are, for example, denitrification of N and decay of oocysts during manure storage (Strokal et al., 2016; Vermeulen et al., 2017). We account for removals and losses in soils as a function of runoff.

Generally, the model distinguishes between (losses to rivers via surface runoff from soils after manure application) and point (direct discharges of manure to rivers) sources of nutrients and oocysts in rivers from livestock production. Direct discharges of manure to rivers are only the point source for Chinese rivers (Strokal et al., 2016; Chadwick et al., 2015). The model considers the crop-livestock relation. The model includes the application of synthetic fertilizers and animal manure on land in calculating inputs of manure-relevant pollutants from land to rivers.

The main model inputs include livestock numbers, the excretion rates of *Cryptosporidium* and nutrients, the prevalence of *Cryptosporidiosis*, manure production per animal, manure management, and runoff (Fig. 1, details in Tables S1-S7, Figs. S1-S4). Most of the model inputs for

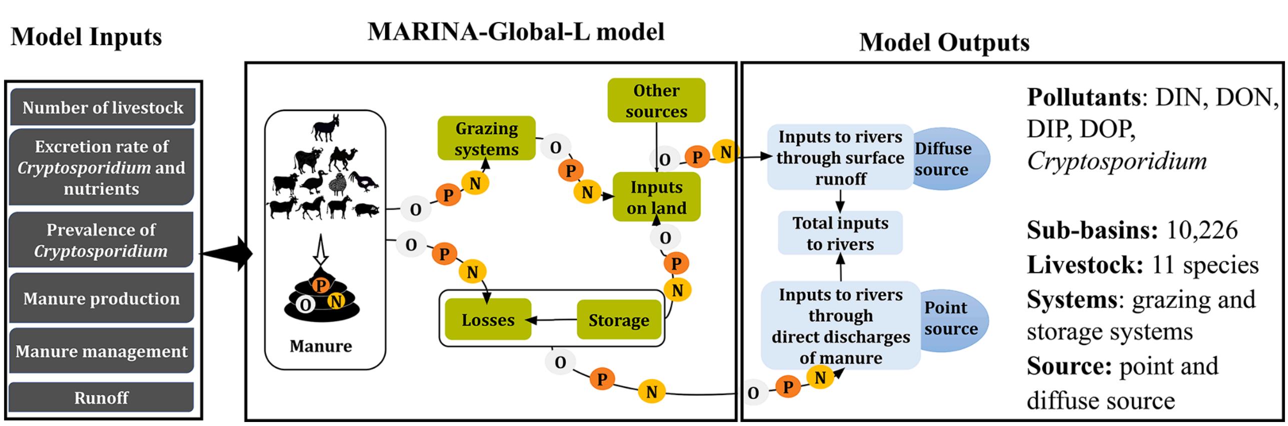


Fig. 1. An Overview of the MARINA-Global-L model with a focus on livestock manure. MARINA-Global-L is short for **M**odel to **A**ssess **R**iver **I**nputs of pollut**N**ts to se**A** for **L**ivestock. Livestock icons indicate the eleven livestock species in the model: buffaloes, camels, cattle, chickens, ducks, donkeys, goats, horses, mules, pigs, and sheep. N, P, and O are short for Nitrogen (N), Phosphorus (P), and Oocysts (O). For details and a complete list of model inputs see Tables S1-S7 and Figs. S1-S2. Inputs to rivers from direct discharges of manure are only relevant for China.

calculating manure-associated nutrients and oocysts on land are derived from the GloWPa-Crypto L1 model. Vermeulen et al. (2017) collected data from various sources (Bouwman et al., 2013; Robinson et al., 2014; IPCC, 2006; Safley et al., 1992; Weedon et al., 2011). Therefore, here, we take the data for oocysts from Vermeulen et al. (2017) at 0.5° grid or country scales (details in Fig. S1). In addition, some model inputs such as N, and P excretion rates, fractions of N volatilization, and direct discharges of manure are from literature (Bouwman et al., 1997; Bouwman and Van Der Hoek, 1997, 2017; IPCC, 2006; Sheldrick et al., 2003; Smith, 1991) and NUFER (**NU**trient flows in **F**ood Chains, **E**nvironment and **R**esources use) datasets at a country scale (Ma et al., 2012). Nutrient inputs to cropland from the application of synthetic fertilizers, atmospheric N deposition, and biological N fixation are derived from the updated IMAGE-GNM (**I**ntegrated **M**odel to **A**ssess the **G**lobal **E**nvironment-**G**lobal **N**utrient **M**odel) model (Beusen et al., 2015). For *Cryptosporidium*, the GloWPa-Crypto C1 model provided model inputs such as runoff fractions for oocysts and their decay (Vermeulen et al., 2019). We process country or grid model inputs to sub-basins as Table S4 describes.

The final model outputs are annual inputs of DIN, DIP, DON, DOP, and *Cryptosporidium* (as oocysts) to rivers Eqs. (1)–(5), Fig. 1). These outputs are by sub-basin, eleven livestock species, grazing and storage systems, and by point and diffuse sources (Figs. 2–6). Calculations are done in two steps (Fig. 1). First, inputs of the pollutants to land are calculated from animal manure (N, P, and oocysts, Eq. (1)). This is done as a function of manure management, livestock number, and removal during storage systems. For nutrients, inputs of total N and P to land are calculated. Second, inputs of the pollutants to rivers are calculated as a function of inputs to land corrected for crop uptake (N and P) and soil-associated retention rates (for N, P, and oocysts, Eqs. (2)–(5). Soil-associated retentions reflect the removal of pollutants during export from land to rivers. This includes, for instance, oocysts decay and N denitrification. For nutrients, inputs of dissolved inorganic and organic N and P to rivers are calculated. The sum of inorganic and organic forms equals to the total dissolved N (TDN) and total dissolved P (TDP). For N and P, we accounted for crop production receiving nutrients from not only manure but also from synthetic fertilizers, atmospheric N deposition, and biological N fixation (Eq. (3)). We corrected the amount of N and P in soils for crop uptake ($G_{p,j}$ in Eq. (3)). The main equations to calculate inputs of nutrients and oocysts to land and rivers are as follows (details are in Tables S1-S7):

$$\mathbf{WSdif_{t,\ i,\ j}} = \mathrm{WSdif_{t,\ i.sg,\ j}} + \mathrm{WSdif_{t,\ i,\ ss,\ j}} \quad (\mathit{Nutrients, oocysts}) \tag{1}$$

$$\mathbf{RS_{t,\ i,\ j}} = \mathrm{RSdif_{t.i.j}} + \mathrm{RSpnt_{t,\ i,\ j}} \quad (\mathit{Nutrients, oocysts}) \tag{2}$$

$$\mathbf{RSdif_{t.i.j}} = \mathrm{WSdif_{t.i.j}} \times \mathrm{G_{t.j}} \times \mathrm{FE_{ws.t.j}} \quad (\mathit{Nutrients}) \tag{3}$$

$$\mathbf{RSdif_{t,\ i,\ j}} = \sum\nolimits_{m=12}^{m} \mathrm{WSdif_{t,\ i,\ m.j}} \times \mathrm{FE_{ws.t.i.m.j}} \quad (\mathit{Oocysts}) \tag{4}$$

$$\mathbf{RSpnt_{t,\ i,\ j}} = \mathrm{S_{t.i.ss,\ j}} \times \mathrm{fr_{\ i,\ d.j}} \quad (\mathit{Nutrients, oocysts}) \tag{5}$$

Where, $\mathbf{WSdif_{t.i.j}}$ is the input of pollutants (t: N, P, and oocysts) to watershed land (WSdif) from the manure of livestock species (i) in sub-basin (j) (kg/yr for N and P, oocyst/yr for *Cryptosporidium*).

$\mathbf{WSdif_{t,\ i,\ sg,\ j}}$ is the input of pollutants (t: N, P, and oocysts) to

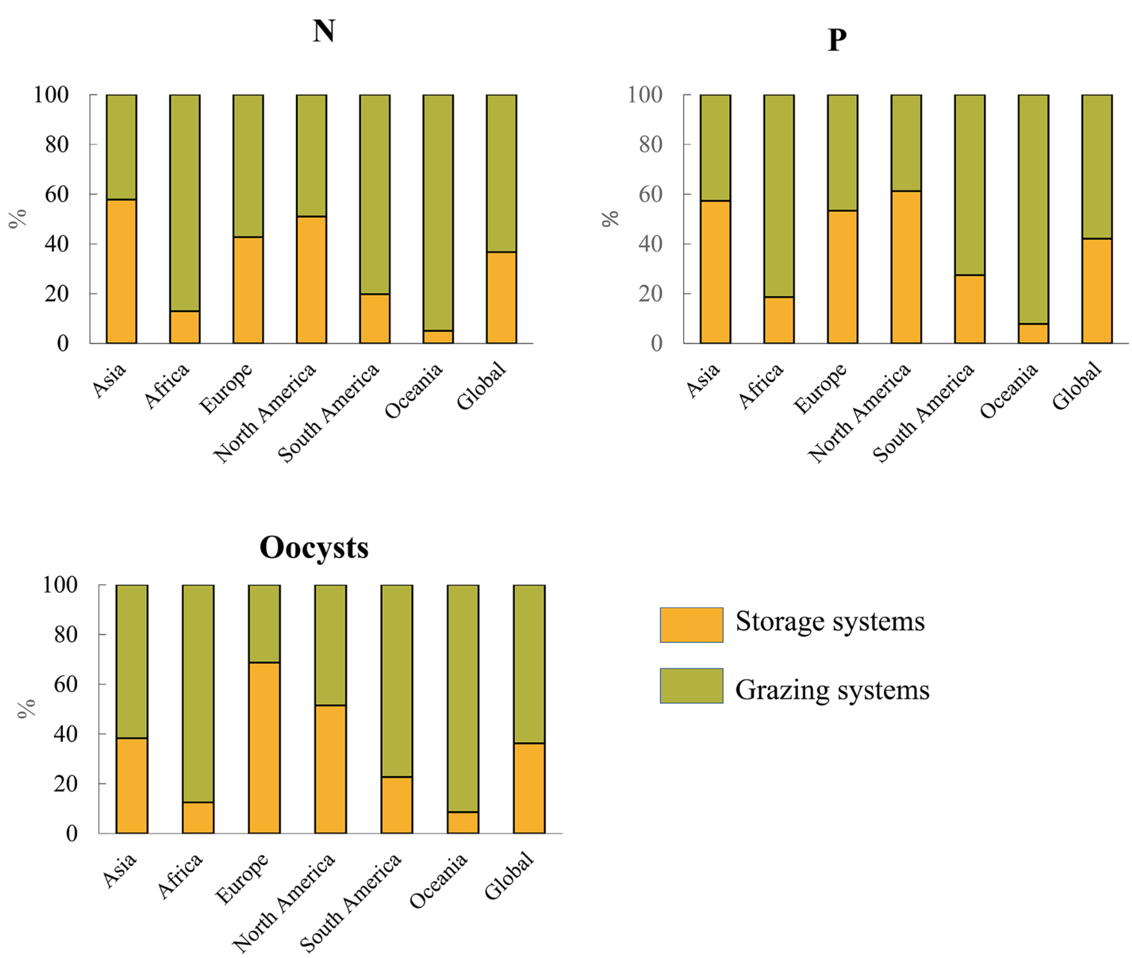


Fig. 2. The shares of grazing and storage systems in manure-inputs of nutrients and oocysts of *Cryptosporidium* to land by continent (%). N and P are short for nitrogen and phosphorus, respectively. Amounts of N, P, and oocysts on land from storage systems are calculated as excretion minus losses for N or decay for oocysts. Amounts of N, P, and oocysts on land from grazing systems are considered direct inputs to land. See Section 2 for the model description and model inputs.

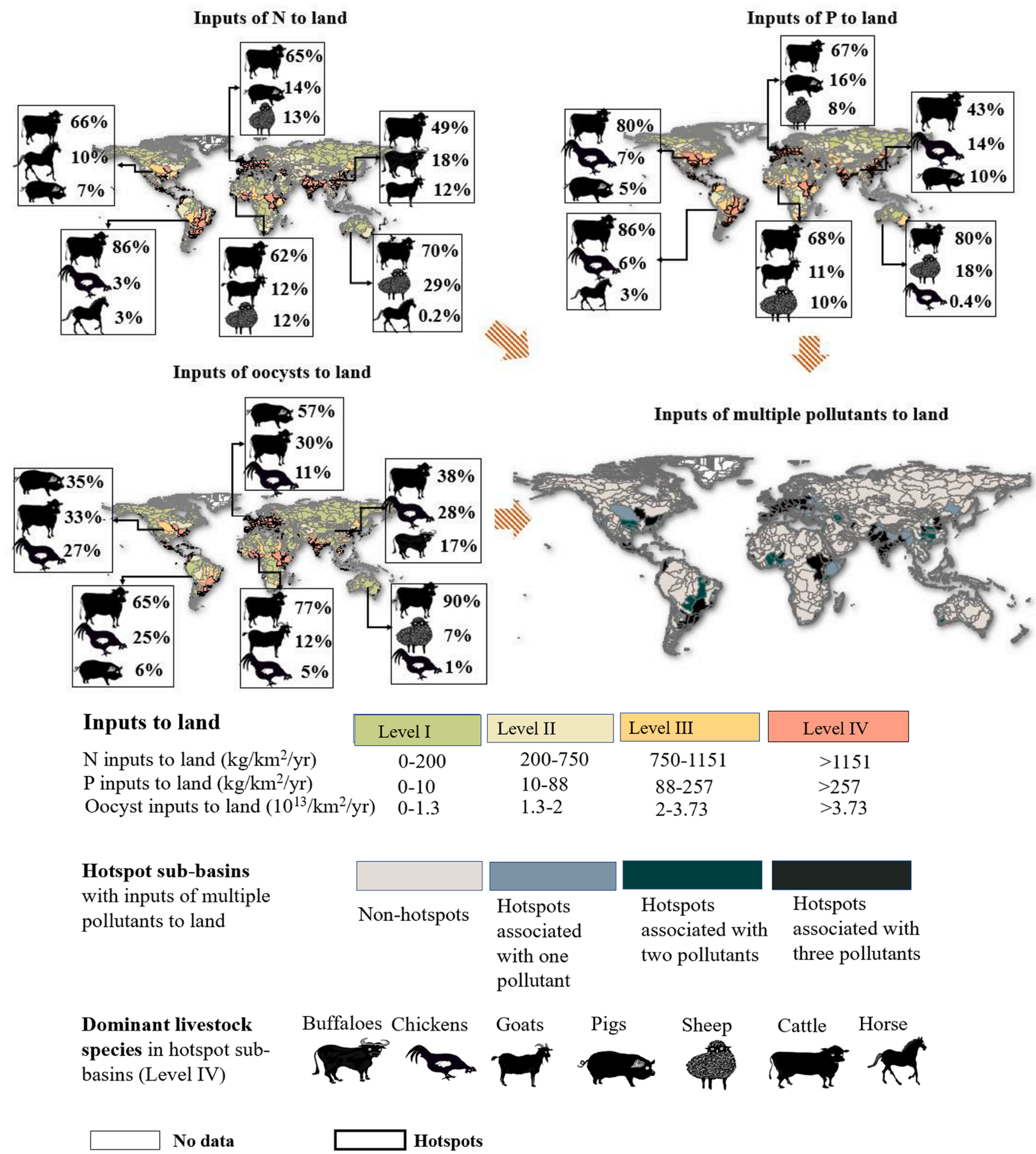


Fig. 3. Inputs of nitrogen (N), phosphorus (P), and oocysts to land from livestock manure by sub-basin in 2010 (kg/km^2/yr for N and P, 10^{13} oocysts/ km^2/yr). Inputs are presented at the sub-basin scale and range from Level I to Level IV. Sub-basins in the range of Level IV are considered as pollution hotspots (see Section 2.3 for the definition). Black lines delineate sub-basins that are considered pollution hotspots. Shares of dominant livestock species are shown for sub-basins with pollution hotspots (Level IV). Sources: the MARINA-Global-L model (see Section 2 for the model description).

watershed land (WSdif) from the manure of livestock species (i) in grazing systems (sg) in sub-basin (j) (kg/yr for N and P, oocyst/yr for *Cryptosporidium*). It is calculated using the manure excretion of nutrients and oocysts for grazed animals (details in Tables S1-S4). $\mathbf{WSdif}_{t.\ i.\ ss.\ j}$ is the input of pollutants (t: N, P, and oocysts) to watershed land (WSdif) from the manure of livestock species (i) in storage systems (ss) in sub-basin (j) (kg/yr for N and P, oocyst/yr for *Cryptosporidium*). It is calculated using the manure excretion of nutrients and oocysts for animals in storage systems ($\mathbf{S}_{t.i.ss.\ j}$, see its explanation below) that is corrected for losses of N to air and decay of oocysts during storage.

$\mathbf{RS}_{t.\ i.\ j}$ is the total input of pollutants (t: DIN, DIP, DON, DOP, oocysts) to rivers (RS) from the manure of livestock species (i) in sub-basin (j) (kg/yr for nutrients, oocyst/yr for *Cryptosporidium*).

$\mathbf{RSdif}_{t.\ i.\ j}$ is the input of pollutants (t: DIN, DIP, DON, DOP, oocysts) to rivers (RS) from the manure of livestock species (i) as diffuse source (dif) in sub-basin (j) (kg/yr for nutrients, oocyst/yr for *Cryptosporidium*).

$\mathbf{G}_{t.\ j}$ is the fraction of pollutants (t: N and P) that is remained in soil after correcting for crop harvesting and livestock grazing in sub-basin (j) (0–1). $\mathbf{G}_{t.\ j}$ is calculated as 1 – (N or P export from land via crop harvesting and animal grazing) / (the sum of N or P inputs to land). For N, inputs to land are from synthetic fertilizers, animal manure, atmospheric N deposition, and biological N fixation. For P, inputs to land are from synthetic fertilizers and animal manure.

$\mathbf{FE}_{ws.t.\ j}$ is the export fraction of pollutants (t: N and P) that is

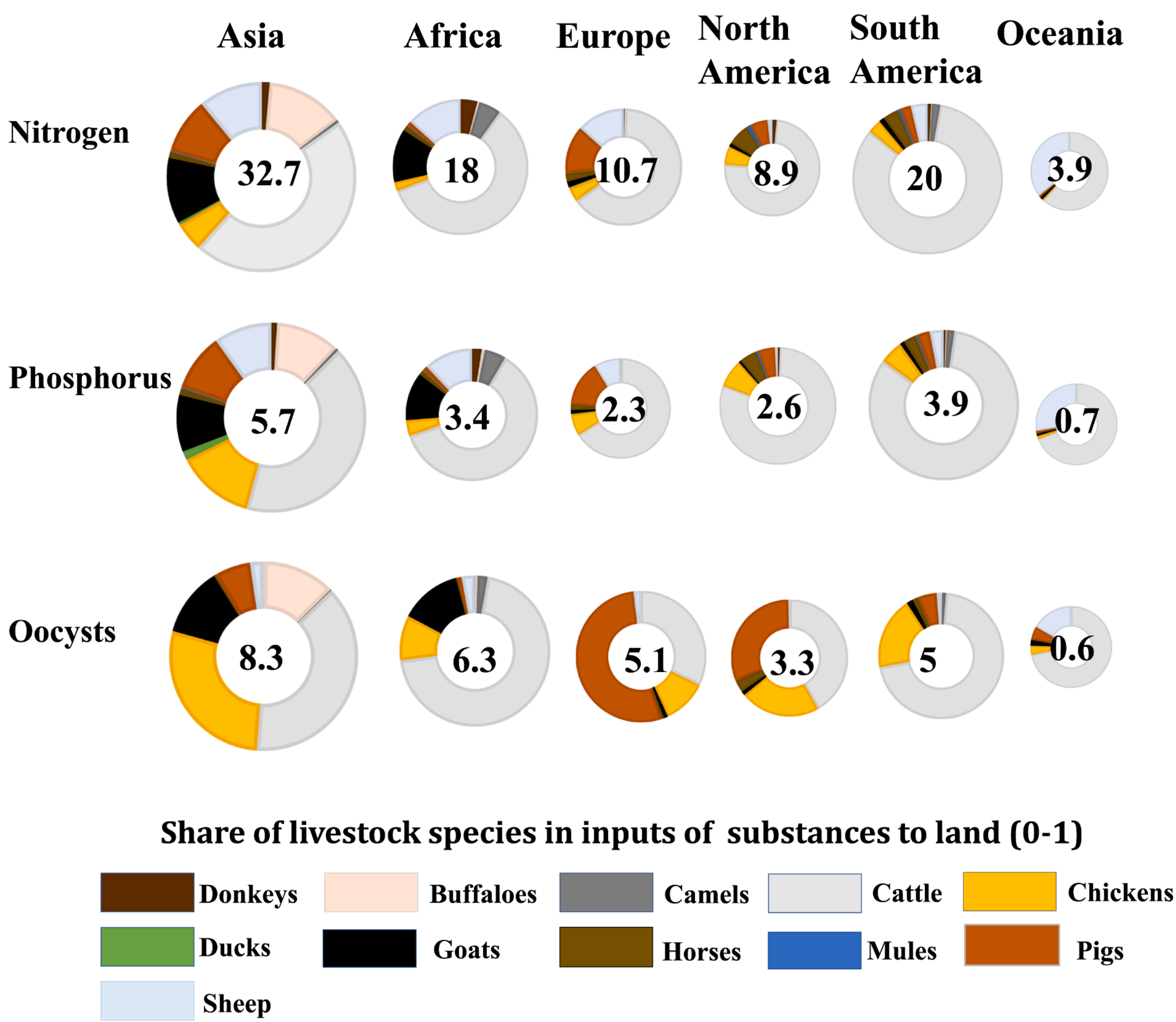


Fig. 4. Inputs of nitrogen (N), phosphorus (P), and oocysts to land (Tg for N and P, 10^{20} for oocysts) and their shares (0–1) by continent in 2010. Pie charts indicate the shares of individual livestock species in inputs of N, P, and oocysts for continents (0–1). Different colors in the pie chart show the shares of individual livestock species in inputs of N, P, and oocysts to land. The sizes of pie charts and the numbers inside of the pie charts indicate the total inputs of N, P, and oocysts to land per continent (unit for N and P is Tg/yr, for oocysts is 10^{20} oocysts/yr). We distinguish six continents: Asia, North America, South America, Africa, Europe, Oceania. Sources: the MARINA-Global-L model (Section 2 for the model description).

exported to rivers as a form (DIN, DON, DIP, DOP) in sub-basin (j) (0–1). It is calculated as a function of surface runoff (details in Tables S1-S4). For oocysts, we calculate $\mathbf{FE}_{ws.t.i.j}$ taking a slightly different approach (see below). $\mathbf{FE}_{ws.t.i.m.j}$ is the fraction of pollutant (t: oocysts) that is exported to rivers from land in month (m) and sub-basin (j) (0–1).

$\mathbf{WSdif}_{t.\ i.\ m.j}$ is the input of pollutants (t: oocysts) to watershed land (WSdif) from the manure of livestock species (i) in month (m) and sub-basin (j) (oocyst/month for *Cryptosporidium*).

$\mathbf{RSpnt}_{t.i.\ j}$ is the input of pollutants (t: DIN, DIP, DON, DOP, oocysts) to rivers (RS) from the manure of livestock species (i) as point source (pnt) in sub-basin (j) (kg/yr for nutrients, oocyst/yr for *Cryptosporidium*).

$\mathbf{S}_{t.i.ss.\ j}$ is the excretion of pollutants (t: N, P, and oocysts) in the manure of livestock species (i) in storage systems (ss) (kg/yr for N and P, oocyst/yr for *Cryptosporidium*).

$\mathbf{fr}_{i.\ d.j}$ is the fraction of direct discharges (d) of pollutants (t: N, P, and oocysts) in manure to rivers from livestock species (i) in sub-basin (j) (0–1).

The model evaluation is presented in Section 4.1.

2.2. Defining pollution hotspots

We define "pollution hotspots" for the inputs of nutrients and oocysts to land and rivers inspired by the approach of Wang et al. (2018). We first rank the inputs of nutrients and oocysts to land and rivers per km^2 of sub-basin area from the lowest to the highest values: from Level I (lowest) to Level IV (highest). Level IV includes the highest values, top 10% most polluted sub-basins. The other three levels reflect the lower values. The ranges, to some extent arbitrary, are described below.

For inputs to land, Levels I, II, and III have the following ranges. For Level I, inputs of N to land range from 0 to 200 kg/km^2/yr. This is from 0 to 10 kg/km^2/yr for P, and from 0 to 1.3 × 10^{13} oocysts/km^2/yr for *Cryptosporidium*. Ranges of Level II are as follows: 200–750 kg/km^2/yr for N, 10–88 kg/km^2/yr for P and 1.3 × 10^{13}–2 × 10^{13} oocysts/km^2/yr for *Cryptosporidium*. Ranges of Level III are as follow: 750–1151 kg/km^2/yr for N, 88–257 kg/km^2/yr for P, and 2 × 10^{13} - 3.73 × 10^{13} oocysts/km^2/yr for *Cryptosporidium*.

For inputs to rivers, Levels I, II, and III have the following ranges. For Level I, inputs of TDN to rivers range from 0 to 30 kg/km^2/yr. This is from 0 to 1 kg/km^2/yr for TDP, and from 0 to 0.4 × 10^{13} oocysts/km^2/yr for *Cryptosporidium*. Ranges of Level II are 30–150 kg/km^2/yr for TDN,

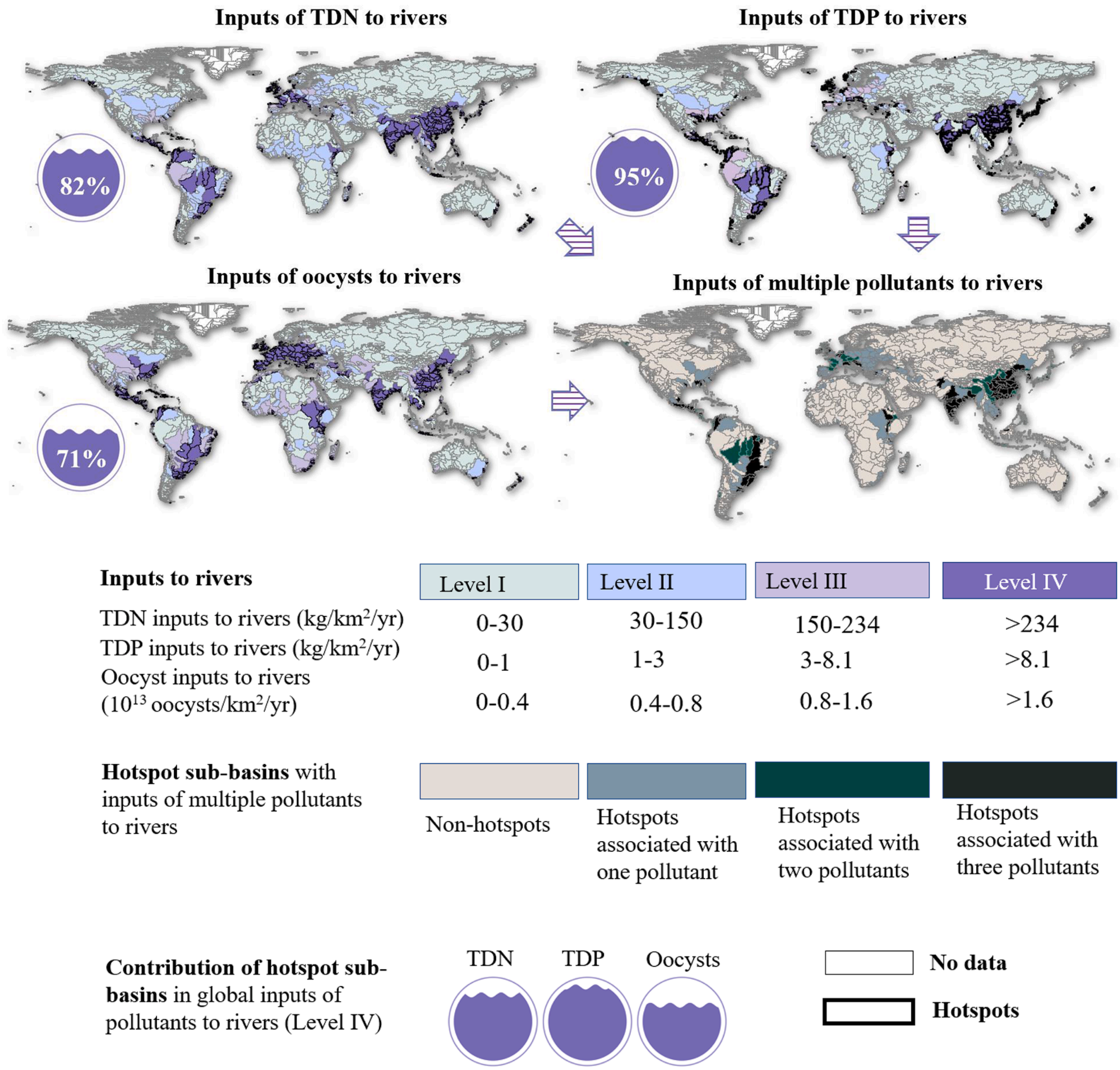


Fig. 5. Inputs of total dissolved nitrogen (TDN), total dissolved phosphorus (TDP), and oocysts to rivers from livestock manure in 2010 (kg/km^2/yr for TDN and TDP, 10^{13} oocyst/km^2/yr). Inputs are presented at the sub-basin scale and range from Level I to Level IV. Sub-basins under Level IV are considered river pollution hotspots (see Section 2.3 for the definition). Sources: the MARINA-Global-L model (see Section 2.2 for the model description).

1–3 kg/km^2/yr for TDP, and 0.4×10^{13}–0.8×10^{13} oocysts/km^2/yr for *Cryptosporidium*. For Level III, the ranges are 150,234 kg/km^2/yr for TDN, 3–8.1 kg/km^2/yr for TDP, and 0.8×10^{13} - 1.6×10^{13}oocysts/km^2/yr for *Cryptosporidium*.

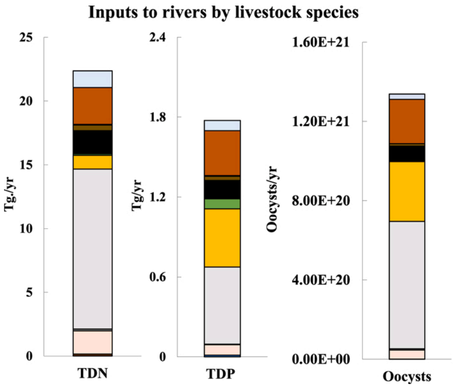
Level IV is considered a pollution hotspot. Level IV has sub-basins in the top 10% with the highest inputs of nutrients and oocysts on land and in rivers. We also define multi-pollutant hotspots. For this, sub-basins are further classified based on the number of pollutants for which the inputs of at least two of the pollutants to land or rivers are in the top highest 10% (under Level IV).

3. Results

3.1. Nutrients and oocysts on land from livestock

Globally, 123 Tg N, 23 Tg P, and 1.0×10^{22} oocysts are excreted in livestock manure in 2010 (Fig. S4). Not all of this reaches the land. Some may be retained in storage or lost to water or air. As a net effect, 94 Tg N, 19 Tg P, and 2.9×10^{21} oocysts are applied on land (Fig. S5). Globally, grazing systems contribute 58% of P, 63% of N, and 64% of oocysts to land (Fig. 2). The share of the grazing and storage systems in manure inputs to land differ among continents (Fig. 2). In many continents, the grazing systems are dominant contributors to N, P, and oocysts on land. This is different in Europe (P, oocysts), North America (N, P, oocysts), and Asia (N and P) where more than half of manure inputs to land are from storage systems (Fig. 2) and largely from chickens, ducks and pigs (Fig. S5).

Asia, South America, and Africa are responsible for over 68% of global N, P, and oocysts on land in 2010 (Fig. 3). For example, livestock in these three continents contributes 70 Tg N, 13 Tg P, and 2.0×10^{21} oocysts on land (Fig. 3). The largest share is for Asia. This is largely associated with a large number of animals in this continent (Fig. S3, Table S8). The contribution of Europe is 12%, 12%, and 18% to global inputs of N, P, and oocysts to land, respectively (Fig. 4). This



Share of livestock species in inputs to rivers (%)

Inputs to rivers		Cattle	Chickens	Pigs	Others	Total
TDN	Total inputs	56	5	13	26	100
	Diffuse sources	63	4	7	26	100
	Point sources	25	8	38	29	100
TDP	Total inputs	32	25	19	24	100
	Diffuse sources	61	10	7	22	100
	Point sources	19	32	25	24	100
Oocysts	Total inputs	48	23	17	12	100
	Diffuse sources	51	19	16	14	100
	Point sources	15	58	21	6	100

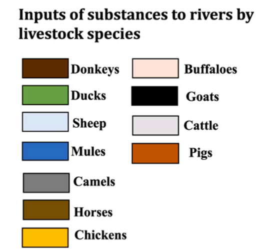


Fig. 6. Global inputs of pollutants to rivers (Tg/yr for TDN and TDP, oocysts/yr for oocysts) and the share of individual livestock species to these inputs in 2010 (%). TDN and TDP are short for total dissolved nitrogen (TDN) and total dissolved phosphorus (TDP). Oocysts belong to *Cryptosporidium*. We consider eleven types of livestock species. "Others" include buffaloes, camels, ducks, donkeys, goats, horses, mules, sheep. Direct discharges of manure and surface runoff are point and diffuse sources of river pollution, respectively. Sources: the MARINA-Global-L model (see Section 2.2 for the model description).

contribution is smaller than the contribution of Asia because the livestock numbers in Asia are five times higher than that in Europe (Fig. S3, Table S8).

Inputs of N, P, and oocysts to land differ largely among sub-basins (Fig. 3). Hotspots of N, P, and oocysts on land are mainly concentrated in sub-basins in Europe, India, the middle parts of Africa and South America, and the eastern part of China (Fig. 3). For example, many sub-basins in Europe, Asia, North, and South America receive more N, P, and oocysts (>1151 kgN/km^2, 257 kgP/km^2, and 3.73×10^{13} oocysts/km^2/yr) than other sub-basins in 2010. This also holds for some sub-basins in Africa (Fig. 3). As a result, 1302 sub-basins can be considered as pollution hotspots of nutrients and oocysts with inputs of N, P, and oocysts to land exceeding 1151 kg N/km^2, 257 kg P/km^2 and 3.73×10^{13} oocyst/km^2 (Level IV). 12% of the global sub-basin areas (1.6×10^7 km^2) is highly polluted by three types of substances simultaneously. Almost all sub-basins in Europe are influenced by three types of substances. In addition, the main Chinese sub-basins contribute a considerable amount (4.4 Tg/yr for N, 1 Tg/yr for P, and 1×10^{20} oocysts/yr for *Cryptosporidium*) to the global inputs of nutrients and oocysts to land as a result of intensive livestock production and direct discharge of manure to rivers..

Cattle production is an important contributor to inputs of N, P, and oocysts to land in many sub-basins and continents (Fig. 3 and 4). In sub-basins that we consider pollution hotspots, the share of cattle production in the inputs of N, P, and oocysts per km^2 ranges from 33% to 90% depending on sub-basins and substance (Fig. 3). For the European sub-basin hotspots, the contribution of pig production to oocysts on land is larger (57%) than the contribution of the cattle (30%) and chicken (11%) production. For the North American sub-basin hotspots (mainly in the Mississippi River basin), the contribution of pig manure to oocysts on land is larger (35%) than the contribution of cattle (33%) and chicken manure (27%) (Fig. 3). On a continental scale (Fig. 4), the contribution of cattle manure is also larger compared to the other livestock. This holds for most continents and substances. Chicken production plays an important role in continents such as Asia, North America, and South America mainly for P and oocysts. Pig production is important in Asia (for N and P), Europe (for N, P, and oocysts), and North America (for oocysts) (Fig. 4).

3.2. Nutrients and oocysts in rivers from livestock

Approximately 22 Tg TDN, 1.8 Tg TDP, and 1.3×10^{21} oocysts enter rivers in 2010. Around 20 Tg N enter rivers as DIN and 2 Tg N enter rivers as DON. For P, this is 1.3 Tg for DIP, and 0.5 Tg for DOP (Fig. S4). However, there is a large spatial variability in river pollution among continents (Fig. 5). At the continental scale, the majority of nutrients and oocysts input to rivers are calculated for Asia and South America. Europe also accounts for large amounts of TDN and oocysts in rivers. This can be explained by the spatial variability in livestock production and manure management (Fig. 3, Table S4). Sub-basins with river inputs exceeding 234 kg N/km^2/yr, 8.1 kg P/km^2/yr, and 1.6×10^{13} oocysts/km^2/yr are considered as pollution hotspots (Level IV). One-fourth of the sub-basin areas (1592 sub-basins) are identified as river pollution hotspots with nutrients and *Cryptosporidium*. These pollution hotspots are responsible for 82% of TDN, 95% of TDP, and 71% of oocysts inputs to rivers globally (Fig. 5). These hotspots are mainly found in Asia, Europe, and South America (Fig. 5).

Cattle, pigs, and chickens are responsible for 74–88% of nutrients and oocysts in rivers in 2010, globally (Fig. 6). Cattle production takes the largest share in global river pollution, which is similar on land (Fig. 6, Section 3.1). Global inputs to rivers from cattle production are 12 Tg of DIN, 0.6 Tg of DON, 0.5 Tg of DIP, 0.1 Tg of DOP, and 6.4×10^{20} oocysts in 2010 (Fig. S6). The substances from livestock species enter rivers via diffuse and point sources (Chadwick et al., 2015; Strokal et al., 2016; Wang et al., 2020).

The share of diffuse and point sources in river pollution differs among the eleven types of livestock species (Fig. 6). Globally, the share of diffuse sources (surface runoff) in river pollution from cattle production ranges from 51% to 63% depending on pollutants (Fig. 6). It is much higher than the contribution from other livestock species. However, point sources of cattle production account for 15–25% of river pollution globally (Fig. 6). This means that compared to other livestock species, diffuse sources of cattle production are more important for river pollution than point sources. This is different for pigs and chickens where point sources of manure play an important role compared to other livestock species globally. For example, pig production accounts for 21% of oocysts, 25% of TDP, and 38% of TDN in rivers globally from point sources (Fig. 5). Point sources from chicken production account for 8% of TDN, 32% of TDP, and 58% of oocysts in rivers globally. Point sources are particularly important for the sub-basins in China.

4. Discussion

4.1. Model uncertainties and comparisons

Our model integrates the GloWPa and MARINA modeling approaches that were previously evaluated (Strokal et al., 2021a; Strokal et al., 2016, 2019; Vermeulen et al., 2017, 2019). The GloWPa models have been validated using available observations for a set of rivers for *Cryptosporidium* (Vermeulen et al., 2017). For nutrients, our model builds on the family of the MARINA models that were evaluated globally, nationally, and seasonally in several earlier studies (X. Chen et al., 2019a, 2019b; Wang et al., 2020; Strokal et al., 2016; Yang et al., 2019; Goshu et al., 2020; Strokal et al., 2021a) (e.g., $R^2 = 0.90$ for the Yangtze basin, $R^2 = 0.84$ for China).

We further evaluate our model using the "building trust" approach (Strokal, 2016; Strokal et al., 2021a; Wang et al., 2020). This includes comparisons with other studies, sensitivity analyses, and discussion of limitations and uncertainties.

◦ Comparisons of model inputs with other studies

Model inputs are, for instance, manure excretion and inputs of pollutants to land (see Section 2). Our model inputs are generally in line with other studies (Supplementary Table S11–16, Supplementary Text 4). Globally, our manure excretion is within the range of 80–130 Tg N from the other studies (Bai et al., 2016; Bouwman et al., 2013; FAO., 2018; Sheldrick et al., 2003). Our total inputs of N and P on land coincide with other studies (91–92 Tg N, 17 Tg P) (Beusen et al., 2015; Bodirsky et al., 2012). Our manure excretion for P for 2010 (23 Tg) is somewhat higher than an earlier estimate for 2000 (17 Tg of P) (Bouwman et al., 2013). One of the reasons could be that between 2000 and 2010, livestock production has increased. This increase is estimated at around 20% globally (FAOSTAT 2021). Another reason could be the difference in the methods and scales (details see Supplementary Text 4). We also compare N excretions with FAO datasets (**F**ood and **A**griculture **O**rganization of the United States) for individual species (Table S12). Our result is in line with FAO datasets. For the whole of China, we calculate the manure excretion of 24 Tg N and 4.5 Tg P, which is comparable to the study of Bai et al. (2016) for N (23 Tg N) and slightly lower for P (5.2 Tg P) because of different datasets and calculation methods (Table S13 Supplementary Text 4).

Spatial variabilities in manure excretion are generally in line with existing studies (Potter et al., 2010; Zhang et al., 2017). Potter et al. (2010) presented the total manure production at 0.5° grids. They focused on nutrients and did not distinguish the contribution of individual livestock species. We do this for nutrients and oocysts at the sub-basins scale. Zhang et al. (2017) estimated N excretion from multiple livestock species at 5 arcmin grids. Our study and Zhang et al. (2017) indicate the large share of cattle, pigs, and chickens in the total N excretion.

◦ Comparisons of model outputs with other studies

Our model outputs are inputs of pollutants to rivers from manure.We also compare model outputs with other models and are able to explain most of the difference. In particular, we compare inputs of different N and P forms in rivers with Global NEWS (Tables S17). The Global NEWS (Global Nutrient Export from WaterSheds model) model calculates inputs of dissolved and organic forms of N and P to rivers from manure (Mayorga et al., 2010). Globally, our model calculated 22 Tg of TDN, and 1.8 Tg of TDP to rivers from manure in 2010. These estimates are higher than that in the Global NEWS model (Table S17). This also holds for our estimates of DIN, DON, DIP, and DOP to rivers (Table S17). The reasons are the differences in time, space, model inputs, and modeling approaches between our model and Global NEWS. For example, we calculate river pollution for 2010 and Global NEWS for 2000. Our model is based on a sub-basin scale modeling approach and Global NEWS is based on a basin-scale modeling approach. The other reason is that we consider direct discharges of manure to Chinese rivers, which is ignored in Global NEWS.

Spatial variabilities in river pollution hotspots are comparable with other studies (Wang et al., 2020; van Vliet et al., 2019; Mayorga et al., 2010; Vermeulen et al., 2019). For example, our river pollution hotspots in many sub-basins of Asia, South America, and Africa coincide with existing studies (Mayorga et al., 2010; Vermeulen et al., 2019). Comparing to studies for nutrients, less data or models are available for pathogen. Generally, our model outputs for *Cryptosporidium* in rivers are comparable with few existing studies (Atwill et al., 2006; Starkey et al., 2007). Our pollution hotspots of *Cryptosporidium* match the pollution hotspots of other microorganisms (Hofstra et al., 2019; Reder et al., 2015). It should be noted that observed values of oocysts in rivers are relatively uncertain. In our study, we focus on both nutrients and oocysts simultaneously, which is not done in existing studies.

Generally, river pollution hotspots (Fig. 5) are calculated for sub-basins that also receive high manure inputs (Fig. 3). This is in line with other studies (e.g., Beusen et al., 2015; Mayorga et al., 2010; Bouwman et al., 2009). Studies show that areas with higher runoff generally receive more pollutants (Yang and Toor, 2018; Chen et al., 2018), which is also shown in our study for sub-basins. Examples are sub-basins located in India, South America, and the south parts of China where the climate is relatively wet with more runoff compared to the North China Plain with relatively dry climate (Zhang and Wang, 2021). In addition, N is more mobile in soils than P (Strokal and de Vries 2012). Thus, more N from land reaches rivers compared to P, which is in line with our study. Oocysts can decay in soils (Vermeulen et al., 2017). Thus, not all oocysts on land can reach rivers. In our model, we account for those factors when calculating inputs to rivers (Section 2).

◦ Sensitivity analysis

We test the sensitivity of the model outputs to changes in model inputs. We ran our model with manure application from the updated IMAGE-GNM model for N and P (Beusen et al., 2015). Results of this ran are comparable with our original model ran for the river pollution with nutrients and their spatial variabilities (Figs. S7-S8). For *Cryptosporidium*, we ran the model with manure application from the GlowPa-Crypto-L1 model and the results are also comparable with our original model ran (Figs. S7-S8).

Furthermore, we compare two methods to aggregate model inputs. We refer to the method used in our model as method 1. Method 2 is an alternative, that we use here for the sensitivity analysis. In method 1 several model inputs (e.g., fractions of grazing systems) are aggregated from country or grids of 0.5° to sub-basins based on livestock population density (details in Figs. S7-S8, Tables S4, S9-S10). This method is transparent but may introduce uncertainties in model outputs. Thus, we use method 2 to test the sensitivity of model outputs to aggregation methods. In method 2, we aggregate some of the model inputs from country or grids of 0.5° by averaging gridded values over the corresponding sub-basins (details in Tables S4, S9-S10). We compare model outputs between these two aggregations methods (Tables S9-S10). This sensitivity analysis confirms that model outputs and our main conclusions are not largely affected by the processing methods of model inputs (Tables S9-S10).

◦ Reflection on model uncertainties

As any integrated models, our model has uncertainties. Nevertheless, we consider our model appropriate for large-scale analyses based on the comparisons with observations, independent datasets and other models, as well as the sensitivity analysis (see above). Model uncertainties are also associated with the modeling approach at the sub-basin scale. Several models (e.g., GLoWPa, IMAGE-GNM) quantify pollution at the 0.5° grid-scale (Vermeulen et al., 2017; Vermeulen et al., 2019; Beusen et al., 2015). Other models do this on the basin scale (e.g., Global NEWS, Mayorga et al., 2010). Our model is in between these two scales. We focus on sub-basins: not too course (basin) and not too detailed (grid). The choice for the sub-basin scale modeling is justified by the research aim. Indeed, some datasets are at grids (e.g., livestock numbers). However, others are national or even regional (e.g., manure management, treatments). We aggregate datasets into the sub-basins and make datasets consistent for nutrients and oocysts (see our Supporting Information for details). We believe that all scales (grids, basins, sub-basins) have pros and cons. We consider sub-basins appropriate in our study considering the availability and uncertainties of the datasets at different scales.

We apply a lumped modeling approach to calculate inputs of pollutants from land to rivers as a function of runoff. In this approach, processes for losses of pollutants in soils (e.g., denitrification, decay of oocysts) are modeled at the sub-basin scale. Other studies modeled nutrients or *Cryptosporidium* for full basins (Mayorga et al., 2010) or 0.5° grids (Beusen et al., 2015; Vermeulen et al., 2019). Our sub-basin scale approach is based on successful earlier model applications (Strokal et al., 2021a; Strokal et al., 2016; Wang et al., 2020; Chen et al., 2019a, 2019b). We improve these earlier studies by harmonizing the model inputs for nutrients and oocysts to the same spatial and temporal level of detail. We use a process-based approach to calculate export fractions of nutrients from land to rivers in sub-basins (Tables S4-S5). Thus, our model does not rely on parameters that were calibrated using a limited set of observations for a certain year or period.

○ Reflection on limitations

Our modeling approach focuses on dissolved nutrients and surface waters (rivers). We do not account for particulates implying that our pollution levels might be underestimated. An important reason is that we focus on dissolved nutrient forms in surface waters that result from manure in livestock production. Particulate nutrients are mainly in rivers from erosion processes (Beusen et al. 2005). On the other hand, we show the impact of livestock production on surface water pollution, which is the main objective of this study.

In addition, existing studies point out the importance of vadose zone (interflow) and groundwater in water pollution assessments. For example, Hartmann et al. (2021) pointed out that up to 50% of infiltrating pollutants may enter groundwater through rapid transport pathways. Some studies indicate that nutrients and oocysts can transport to groundwater through deep percolation (Khan et al., 2018; Balthazard-Accou et al., 2020; Gu et al., 2013). Therefore, the next steps would be to add erosion and explore approaches to account for multiple pollutants in groundwater.

In our study, we account for interactions between nutrients and oocysts at the source site. For example, manure is a common source of both nutrients and pathogens on land and in rivers. Runoff is the common pathway of these pollutants to reach rivers. However, our model does not account for biogeochemical interactions between nutrients and oocysts in rivers. The knowledge in this field is very limited for large-scale water quality modeling. A few studies addressed interactions (Tamis et al., 2021; Kong et al., 2019), but were limited to data and scale. More research is needed to better understand interactions between nutrients and pathogens in river systems.

Our results are for the year 2010, which is about 10 years from now. We may under- or overestimate pollution levels in sub-basins for today. However, we believe that our conclusions are still relevant. We showed the pollution hotspots as the priority areas for effective manure management. We believe that the locations of those hotspots did not change much between 2010 and today. This is because those places remain densely populated with intensive livestock production (e.g., Asian sub-basins). Another conclusion is the importance of manure as the common source of multiple pollutants in rivers. We also believe that this conclusion is highly relevant today. We show the large contribution of pigs, cattle, and chickens to river pollution. Today, these animals remain important producers of meat for human consumption (FAOSTAT, 2021). This implies that managing the manure of these species is still relevant for reducing future river pollution by multiple pollutants. Moreover, we present our results on the global (e.g., Fig. 6), continental (Fig. 4) and sub-basin scales (e.g., Fig. 5). Thus our insights are relevant for water management at those scales. For management at local scales such as farms, our model needs further validation.

All of the above builds trust in our model results for large-scale analyses. We argue that the inevitable uncertainties in the model inputs and approach do not largely affect our main conclusions. However, for local analyses of livestock impacts on multi-pollutant problems (e.g., within sub-basins, villages), our model needs to be further evaluated and validated.

4.2. Livestock management to reduce river pollution

Our study provides four insights for livestock management. First, we show the priority areas (sub-basins) for pollution reduction in the world (Fig. 5). Sub-baisns allow us to account for the spatial variability especially within the large basins (e.g., Mississippi, Yangtze, Nile, Danube, Amazon). Many studies confirms that water pollution is a worldwide problem. For instance, nutrient cycling and associated pollution are global phenomena. Nutrient-induced eutrophication is found in many surface water such as rivers, estuaries, and lakes (Lapointe et al., 2017; Wynne and Stumpf, 2015; Lehman et al., 2015; Isaacs et al., 2014; Strokal et al., 2016; Mayorga et al., 2010). Pathogen pollution is also a problem in many developing countries, posing health risks (Bouzid et al., 2018). This is a result of livestock production and insufficient manure management leading to high inputs of pollutants to land and thus to rivers (Figs. 3 and 5). Global change may influence the intensity of livestock production and runoff, causing pollution in the coming years (Herrero et al., 2016; Ilea, 2009). Impacts of global change have to be considered in formulating effective management options to reduce pollution hotspots.

Second, our study can help to identify the contributions of livestock systems to river pollution. We show that grazing systems dominate in some regions (e.g. Oceania, Africa) and storage systems in others (e.g. Europe, North America). This indicates where and what efforts are needed to reduce manure-related river pollution. For example, reducing pollutants to rivers from grazing systems is needed in Africa. Reducing pollutants to rivers from storage systems is needed in Europe. This means that effective manure management is important for both systems to avoid future river pollution globally.

Third, we provide a better understanding of the contribution of livestock species to river pollution taking a multi-pollutant perspective. This was not done before for river sub-basins and N, P and oocysts, simultaneously. Our study can be considered an urgent call for better manure management in the production of pigs, cattle, and chickens globally (Figs. 3 and 6). This differs among sub-basins. For example, for Asian sub-basins, river pollution reduction is needed from the production of pigs, sheep, cattle, and chickens. This information could be used to define sub-basin-specific strategies to manage manure effectively. Examples of such technologies are livestock feed management (Ershadi et al., 2020), dietary guidelines (Health Canada, 2019; Caro et al., 2017), or livestock-specific policies.

Fourth, our study indicates the dominant water pollution sources (diffuse and point source) for multiple pollutants. Diffuse sources are most important globally, and point sources are important for China. To our knowledge, this is also the first attempt to quantify the impact of livestock production on multiple pollutants in rivers by source from individual livestock species for sub-basins simultaneously. For example, existing studies also showed the importance of point sources for Chinese rivers (Chadwick et al., 2015; Strokal et al., 2016), but mainly for nutrients. We take a multi-pollutant approach and focus on nutrients and oocysts. Furthermore, existing studies show the importance of diffuse sources, but only for individual pollution types (Tang et al., 2011; van Vliet et al., 2019). Our model could be used for scenario analysis to explore the synergetic actions for reducing multiple pollutants in rivers in the future. Our model could also be used to analyze the impact of upstream activities on downstream water pollution.

4.3. Implications for the achievement of sustainable development goals

Sustainable livestock production supports Sustainable Development Goal (SDG) 2 on food security and SDG 6 on clean water and sanitation

(Mehrabi et al., 2020; Sakadevan and Nguyen, 2017; Tilman and Clark, 2014). Improving livestock production and its manure management (SDG 2) could potentially reduce river pollution with nutrients and oocysts (SDG 6) (Mehrabi et al., 2020; Strokal et al., 2021a). This is a synergetic effect between SDGs 2 and 6. Our results indicate where (sub-basins) and what sources (point or diffuse) we need to focus on in livestock production (SDG 2) to reduce river pollution (SDG 6). This information could support the formulation of effective management options for livestock production that can account for synergies between SDG 2 and SDG 6 and mitigate their trade-offs (e.g., more manure may lead to more multiple pollutants in rivers).

Synergetic solutions are needed to mitigate multi-pollutant problems in many rivers worldwide. Effects of policies on river pollution have been explored in existing studies for many world regions (Yasin et al., 2010; EU Commission, 2019; LNV, 2018). China plays an important role in reducing global river pollution from livestock production. The Chinese government promotes the Agriculture Green Development (AGD) with high agricultural production and low environmental impacts (Shen et al., 2020). AGD facilitates the implementation of environmental policies. For instance, non-livestock production regions and pig-relocation policies have been introduced to controlling surface water pollution (Bai et al., 2019). The Water Pollution Preventions and Control Plan have also been introduced to avoid pollution (Central People's Government of the People's Republic of China, 2015). AGD can serve as an example for other regions. This is because AGD combines efforts in integrating knowledge of crop, animal, food processing, and environmental aspects for effective solutions (Strokal et al., 2021b). Models have been used to quantify the effects of those policies on reducing nutrient pollution in rivers in the future (Wang et al., 2020). AGD highly supports the SDG and can be used for other countries as inspiration towards sustainable agriculture. Our model can be used to explore the effects of the AGD policies on river pollution in China and elsewhere.

5. Conclusions

This study quantified inputs of N, P, and oocysts from *Cryptosporidium* to rivers from livestock production, by animal types and in a spatially explicit way. To this end, we developed and applied the MARINA-Global-L model to 10,226 sub-basins for nutrients and oocysts. Results showed that around 94 Tg N, 19 Tg P, and 2.9×10^{21} oocysts were applied on land from livestock production in 2010. Over 57% of nutrients and oocysts were from the grazed animal systems. Asia, South America, and Africa were responsible for over 68% of these pollutants on land. We calculated that around 22 Tg TDN, 1.8 Tg TDP, and 1.3×10^{21} oocysts entered rivers in 2010. One-fourth of the sub-basin area was responsible for 71–95% of nutrients and oocysts in rivers. Cattle, pigs, and chickens contributed by 74–88% to manure-related inputs of nutrients and oocysts in rivers globally. Our study can support the formulation of effective manure management strategies to achieve SDG 2 (food security) and SDG 6 (clean water).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2021.117906.

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