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Increasing dissolved nitrogen and phosphorus export by the Pearl River (Zhujiang): a modeling approach at the sub-basin scale to assess effective nutrient management

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Abstract The Pearl River (Zhujiang in Chinese) has been exporting excess of nitrogen (N) and phosphorus (P), causing eutrophication in the coastal waters of southern China for decades. However, sources of these nutrients and their locations are not well studied for the Pearl River basin. As a consequence, it is difficult to formulate effective management options to reduce these nutrients in the river and to prevent further eutrophication. We developed a sub-basin model based onto the Global *NEWS*-2 (Nutrient Export from WaterSheds) model for the period of 1970–2050 to analyze trends in dissolved inorganic N and P (DIN and DIP) and to identify the main sources of these nutrients and their locations. We validated our model by comparing modeled nutrient fluxes with observed. Future analyses are based on Millennium Ecosystem Assessment scenario that assumes a globalized world with a reactive environmental management. DIN and DIP inputs to the coastal waters are calculated to increase by a factor of 2–2.5 between 1970 and 2050. Over two-thirds of the DIN and DIP inputs to the coastal waters stem from two down-stream basins (Zhujiang delta and Dongjiang), where agriculture and sewage are important drivers of this increase. Agriculture accounts for over 40 % of DIN inputs to coastal waters. Sewage and agriculture account for over 90 % of DIP inputs. Thus nutrient management in agriculture and sewage in down-stream areas is more effective in reducing coastal eutrophication than nutrient management in up- and middle-stream areas of the Pearl River basin.

Keywords Nitrogen · Phosphorus · The Pearl River · Sub-basin scale modeling

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Introduction

China is a country with a rapidly growing population and economy (Hou et al. 2013; Liu et al. 2012; Ma 2012; Ma et al. 2013; Vitousek et al. 2009; Zhou et al. 2012). An unwanted side-effect of this is increased environmental pollution. For instance, coastal eutrophication problems with harmful algal blooms have been observed since the 1970s in Chinese seas (Diaz and Rosenberg 2008; Liu and Qiu 2007; Selman et al. 2008; Wang et al. 2007; WRI 2010). This is a result of increased nitrogen (N) and phosphorus (P) inputs to rivers from human activities on the land such as agriculture, industry, and sewage (Aregay and Minjuan 2012; Ermolieva et al. 2009; Maimaitiming et al. 2013; Qu and Kroeze 2010, 2012; Vitousek et al. 2009).

In this study we focus on the Pearl River basin, which is the third largest river in China (around 440 thousands km^2; Fig. 1). The drainage basin of the river covers Yunnan, Guizhou, Guangxi and Guangdong provinces. It serves as a major water supplier for human consumption, agriculture (e.g., irrigation), hydropower generation and navigation in southern China (Cui et al. 2007; Weng 2007). However, the Pearl River estuary has been eutrophic since the 1970s leading to harmful algal blooms, and environmental and economic losses (Huang et al. 2003; Wang et al. 2007). The contribution of human activities (e.g., agriculture and urbanization) to eutrophication may increase in the coming years because of a rapidly growing population and economy (Hou et al. 2013; Liu et al. 2012; Ma 2012; Ma et al. 2013; Vitousek et al. 2009; Zhou et al. 2012).

The sources of nutrients and their locations in the basin have not been studied to a large extent because of the large basin size that complicates a quantitative assessment. This holds for both empirical and model-based studies. Empirical studies are scarce and often limited to concentration measurements at specific locations, for selected nutrient forms at specific times (Chen et al. 2008; Huang et al. 2003; Liu et al. 2009; Yan et al. 1999, 2011; Zhang 2002; Zhang et al. 1999). Modeling studies (Alexander et al. 2002; Kroeze et al. 2013; Ongley et al. 2010) exist but their spatial resolution is often ether too coarse (Mayorga et al. 2010; Ti et al. 2012) or too detailed (Arnold et al. 1998; Schwarz et al. 2006; Smith et al. 1997) for such large data-poor basins. Examples are the Soil and Water Assessment Tool (SWAT) model and a coupled physical–biological model applied to the Dongjiang (Wu and Chen 2013; Zhou et al. 2012) and to the Pearl River delta (Hu and Li 2009), respectively. An advantage of the SWAT model is that it can simulate nutrients in surface waters and their sources (e.g., agriculture) on a monthly basis for small watersheds, making it possible to analyze spatial and seasonal variability of nutrient fluxes (Gassman et al. 2007). Implementing this model to the entire Pearl River basin, however, requires detailed input data (e.g., daily hydrology, soil properties of different depths), which are often not available in good quality for such large basins. A coupled physical–biological model of Hu and Li (2009) quantifies nutrient fluxes and their transformations, but only for the Pearl River delta and only for the contemporary period.

Global *NEWS*-2 (Nutrient Export from WaterSheds) (Mayorga et al. 2010; Seitzinger et al. 2010) is a global spatially explicit basin scale model. It quantifies past (1970, 2000) and future (2030, 2050) trends in river export of nutrients and their sources (e.g., agriculture, sewage). The model has been applied at the global scale (Mayorga et al. 2010) and at regional scales such as the Black Sea (Strokal and Kroeze 2013; Strokal et al. 2014c), the Bay of Bengal (Sattar et al. 2014; Zinia and Kroeze 2014), Indonesia (Suwarno et al. 2013, 2014), Africa (Yasin et al. 2010). The model was applied to the Chinese rivers including the Pearl River (Qu and Kroeze 2010, 2012; Strokal et al. 2014b) and the Yangtze (Yan et al. 2010). The main strength of this model is to analyze causes of coastal eutrophication in large data-poor regions such as the Pearl River basin (see "Methodology" section for more details about the model). However, for rivers as large as the Pearl modeling at the basin scale may not help us in prioritizing pollution management strategies at the provincial or county level to manage coastal eutrophication in an efficient way. The Pearl River Water Resources Commission (PRWRC) stresses the need to perform sub-basin evaluation of water resources (Cui et al. 2007). So far, sub-basin studies do not exist for the entire Pearl basin.

Our main research objective is to quantify trends in dissolved inorganic N (DIN) and P (DIP) export by the Pearl River to coastal waters by source, and to identify locations of the nutrient sources. These dissolved nutrients are the most reactive forms of N and P, and thus important in coastal eutrophication (Dumont et al. 2005; Garnier et al. 2010). To this end, we applied Global *NEWS*-2 at the sub-basin scale for the period of 1970-2050. In the following, we describe the sub-basin scale modeling approach applied to Global *NEWS*-2 ("Methodology" section), present model results on trends in DIN and DIP export by the Pearl River and their sources ("Results and discussion" section), and conclude main findings ("Conclusions" section). Our

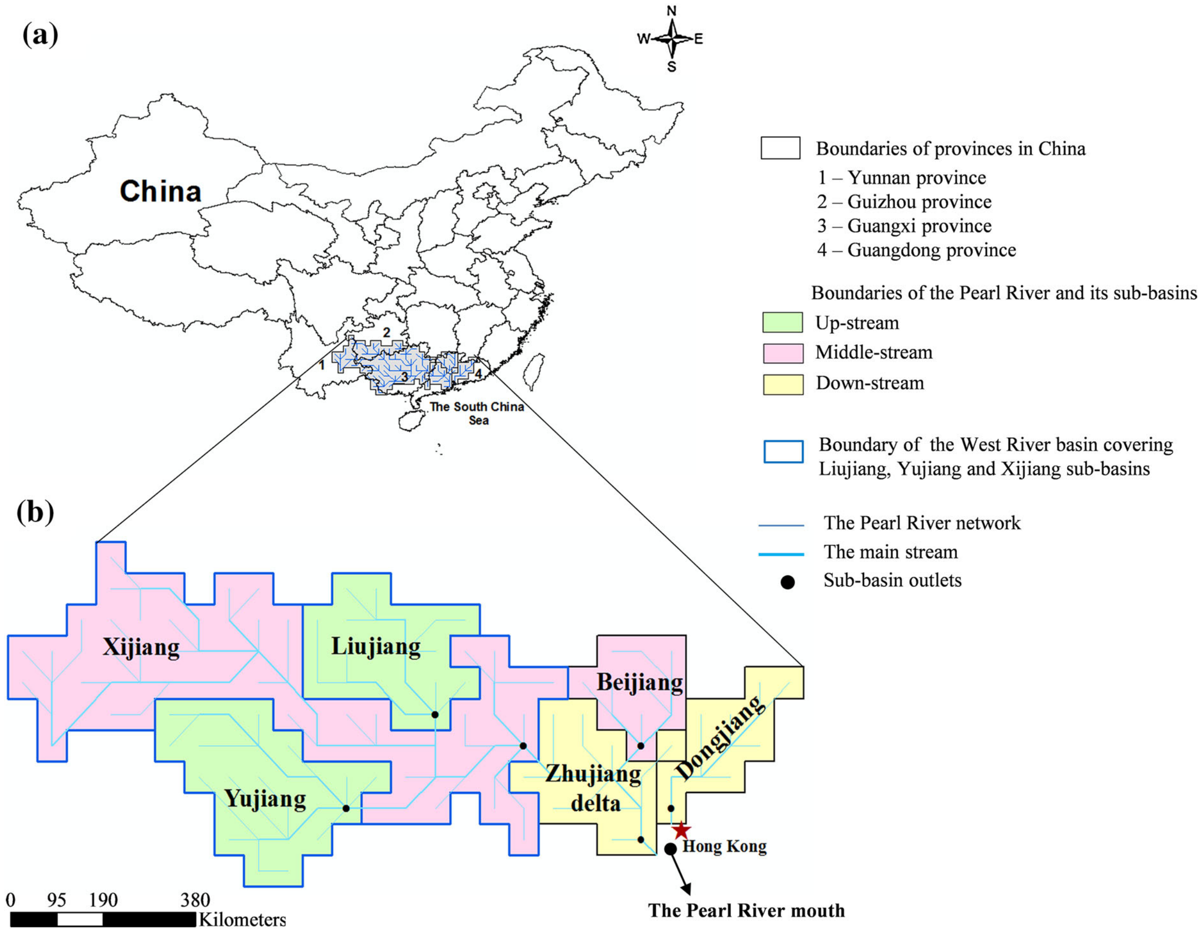

Fig. 1 **a** The Pearl River basin located in China, **b** the Pearl River sub-basins. Provincial boundaries are from the global administrative areas (GADM 2012). Sub-basins are delineated based on the topological simulated network (STN-30 v6.01) (Mayorga et al. 2010; Vörösmarty et al. 2000b)

study provides new insights in effective sub-basin management to reduce eutrophication in aquatic systems.

Methodology

A sub-basin scale modeling approach for the Pearl River

We developed a sub-basin scale approach to quantify dissolved inorganic N and P export by the Pearl River to the coastal waters by source during 1970–2050 (Fig. 2). We applied this approach to the Global *NEWS*-2 model. The original Global *NEWS*-2 model is described by Mayorga et al. (2010). Below we briefly describe the sub-basin scale approach for DIN and DIP export by the Pearl River. First, we identified sub-basins of the Pearl River and then we quantified nutrient export at the sub-basin scale.

We identified six sub-basins: Yujiang, Liujiang, Xijiang, (they form the West River basin), Beijiang, Dongjiang and Zhujiang delta (Fig. 1b). Their names refer to the names of the rivers at their outlets. The Beijiang, Dongjiang and Zhujiang delta sub-basins are defined on the basis of earlier studies (Cui et al. 2007; Niu and Chen 2010; Weng 2007; Zhang et al. 2007). We added to these the Yujiang and Liujiang rivers, which are two main tributaries of the Xijiang river (the West River) according to Niu and Chen (2010), Cui et al. (2007) and Zhang et al. (2007). We classify these six sub-basins into up-stream (Liujiang and Yujiang), middle-stream (Xijiang and Beijiang) and down-

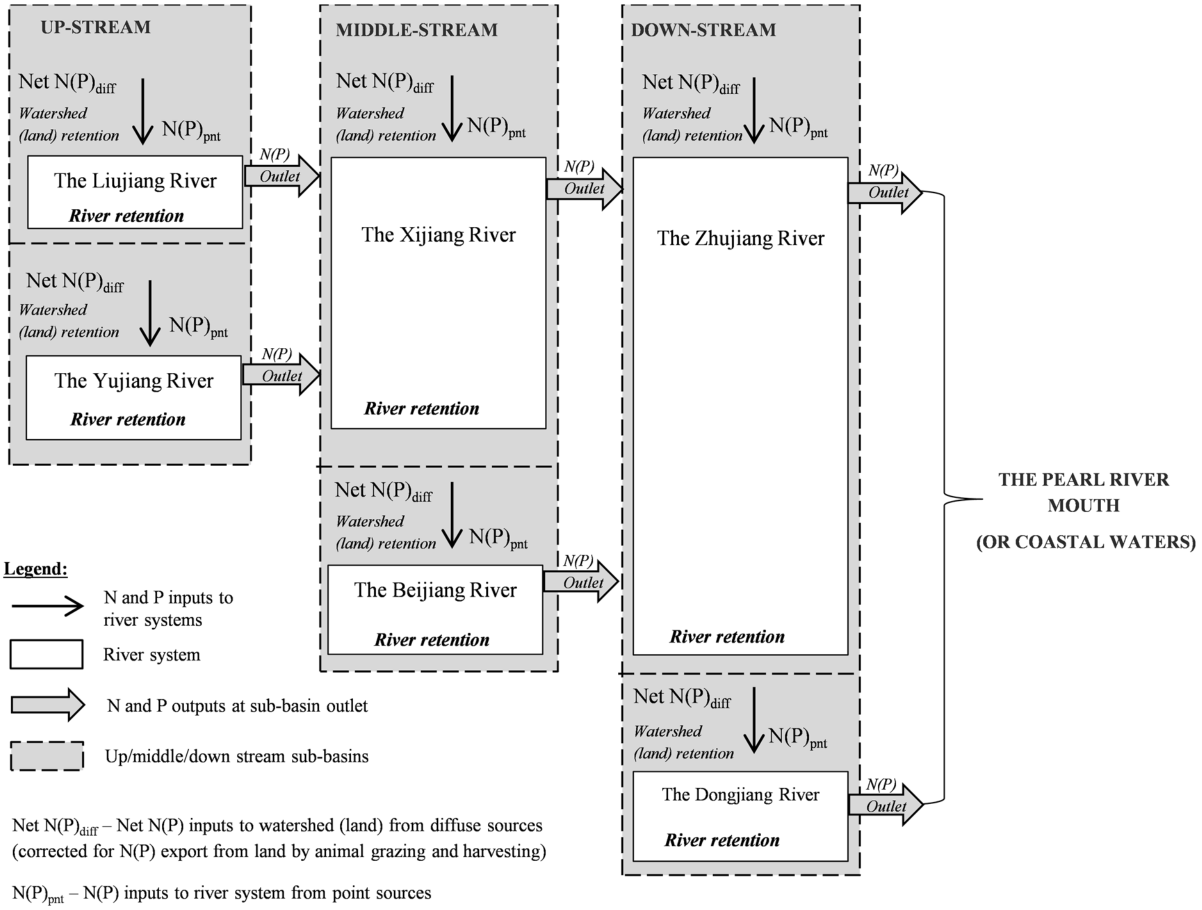

Fig. 2 Sub-basin scale modeling framework for the Pearl River

stream (Zhujiang delta and Dongjiang), which is needed for further calculations (Fig. 2).

Our sub-basin modeling approach for the Pearl River (Fig. 2) allows for analyzing the contribution of sources at sub-basin scale to DIN and DIP inputs to the coastal waters, for past and future years. This has not been done before for the Pearl River. DIN and DIP export by the Pearl River is modeled as a function of N and P inputs to surface waters in each sub-basin. We account for diffuse and point sources (see below for a description), corrected for nutrient retention on land. Some of these nutrients are lost (e.g., N denitrification, water withdrawal for irrigation) or/and retained (e.g., N and P sedimentation in reservoirs) in surface waters before reaching the outlets of the sub-basins (Fig. 2). The amount of nutrients exported to the coastal waters of the Pearl River from each sub-basin outlet is determined by transport of nutrients through river systems in the more down-steam sub-basins. Thus DIN and DIP exported to the outlet of an up-stream sub-basin is either transported to the coastal waters by the rivers in the middle- and down-stream sub-basins, or lost or/and retained before reaching the coastal waters. Likewise, middle-stream sub-basins transport nutrients that are not lost or retained to more down-stream sub-basins. The down-stream sub-basins include the mouth of the Pearl River, and discharge into the coastal waters (Fig. 2).

Our sub-basin model quantifies DIN and DIP transport to the mouth of the Pearl River (indicated as M) from each sub-basin j (ju. jm and jd for up-stream, middle-stream and down-stream sub-basins, respectively) and from each source y as follows:

$$M_{F.y.ju} = \left(RS_{F.y.ju} \times FE_{riv.F.ju}\right) \times FE_{riv.F.jm} \times FE_{riv.F.jd} \quad (1)$$

$$M_{F.y.jm} = \left(RS_{F.y.jm} \times FE_{riv.F.jm}\right) \times FE_{riv.F.jd} \quad (2)$$

$$M_{F.y.jd} = RS_{F.y.jd} \times FE_{riv.F.jd} \quad (3)$$

where $M_{F.y.ju}$, $M_{F.y.jm}$, $M_{F.y.jd}$ are the transport of nutrients by form (F: DIN, DIP) to the mouth of the Pearl River from source y and from up-stream (ju), middle-stream (jm) and down-stream (jd) sub-basins (kg year^{-1}), respectively. $RS_{F.y.ju}$, $RS_{F.y.jm}$, $RS_{F.y.jd}$ are nutrient inputs (F: DIN, DIP) to the river system (RS) from source y in up-stream (ju), middle-stream (jm) and down-stream (jd) sub-basins (kg year^{-1}). $FE_{riv.F.ju}$, $FE_{riv.F.jm}$, $FE_{riv.F.jd}$ are fractions of nutrient (F: DIN, DIP) inputs to the rivers that are exported at the outlet of up-stream (ju), middle-stream (jm) and down-stream (jd) sub-basins (0–1).

We applied Global *NEWS*-2 (Mayorga et al. 2010) to model $RS_{F.y.ju}$, $RS_{F.y.jm}$, $RS_{F.y.jd}$ and $FE_{riv.F.ju}$, $FE_{riv.F.jm}$, $FE_{riv.F.jd}$. Box A.1 and Table A.1 in Supplementary Materials summarize the main equations to quantify these variables. These variables are modeled similarly for each sub-basin and thus they are indicated as $RS_{F.y.j}$ and $FEriv_{F.j}$ without specifying up-, middle- and down-stream sub-basins in Box A.1.

Nutrient inputs to the river system include diffuse ($RSdif_{F.y.j}$) and point ($RSpnt_{F.y.j}$) source inputs of nutrients from land. *Diffuse sources* include synthetic N and P fertilizer use (for DIN, DIP), animal manure excretion (for DIN, DIP), atmospheric N deposition on agricultural areas and non-agricultural areas, and biological N_2-fixation (for DIN) by agricultural crops and natural vegetation, and weathering of P-contained minerals in agricultural and non-agricultural areas. In this study agricultural areas combine five land use types: grassland in pastoral systems (e.g., dominated by grazing, limited manure storage and application), grassland in mixed systems (e.g., areas close to rivers, manure storage and application can take place), wetland rice, legumes (e.g., soybean and pulses) and cropland (e.g., maize, cereals) (Bouwman et al. 2009). Non-agricultural areas include any other land cover.

Nutrient inputs to the river system in sub-basin j from diffuse source y, except for P weathering, are calculated as (see Box A.1 in Supplementary Materials) (Bouwman et al. 2009; Mayorga et al. 2010):

$$RSdif_{F.y.j} = WSdif_{E.y.j} \times G_{F.j} \times FE_{ws.F.j} \quad (4)$$

where $RSdif_{F.y.j}$ is the nutrient input to the river system (RS) from diffuse (dif) source y in sub-basin j (kg year^{-1}). $WSdif_{E.y.j}$ is the inputs of nutrient by element (E: N, P) to land from diffuse source y in sub-basin j (kg year^{-1}). $G_{F.j}$ is the fraction of land-surface diffuse F (F: DIN, DIP) sources remaining after animal grazing and crop harvesting in sub-basin j (0–1), applicable for agricultural areas only. It is calculated as nutrient export from land by animal grazing and crop harvesting divided by the total nutrient inputs to agricultural land from all diffuse sources (see Box A.1 and Table A.1 in Supplementary Materials). $FE_{ws.F.j}$ is the fraction of nutrient form (F: DIN, DIP) that is exported from land (watershed) to the river of sub-basin j (0–1). This fraction is calculated as a function of annual runoff from land to surface waters (see Box A.1 and Table A.1 in Supplementary Materials).

Weathering of P-containing minerals in soils of agricultural and non-agricultural areas is modeled as a function of annual runoff from land to streams (an export-coefficient approach), following Global *NEWS*-2. Details are presented in Box A.1 and Table A.1 in Supplementary Materials and in Mayorga et al. (2010).

Point sources include human waste (for DIN and DIP) and detergents (for DIP). Nutrient inputs to the rivers of each sub-basin j from each point source y are calculated as (see Box A.1 and Table A.1 in Supplementary Materials) (Mayorga et al. 2010; Van Drecht et al. 2009):

$$RSpnt_{F.y.j} = \left[\left(1 - hw_{frem.E.j}\right) \times I_j \times WShw_{E.y.j}\right] \times FEpnt_{F.j} \quad (5)$$

where $RSpnt_{F.y.j}$ is the nutrient input to the river system (RS) from point (pnt) source y in sub-basin j (kg year^{-1}). $hw_{frem.E.j}$ is the fraction of nutrient element (E: N, P) removed during wastewater treatment in sewage facilities of sub-basin j (0–1). I_j is the fraction of population connected to sewage facilities (0–1). $WShw_{E.y.j}$ is the input of nutrient element (E: N, P) in human waste (y) and detergents (y) in watersheds (land) of sub-basin j (kg year^{-1}). $FE_{pnt.F.j}$ is the fraction of nutrient element (E: N, P) in sewage effluents that is entering the river of sub-basin j as a form (F: DIN, DIP) (0–1).

The river export fractions ($FE_{riv.F.j}$ in Eqs. 1–3) reflect the retention of nutrient form (F: DIN, DIP) in the river system ($L_{F.j}$, 0–1), in dammed reservoirs ($D_{F.j}$, 0–1), and nutrient losses by water consumption for purposes such as irrigation and hydropower ($FQrem_j$, 0–1) (Mayorga et al. 2010):

$$FE_{riv.F.j} = \left(1 - L_{F.j}\right) \times \left(1 - D_{F.j}\right) \times \left(1 - FQrem_j\right) \tag{6}$$

River retention ($L_{F.j}$, 0–1) is considered for DIN only in Global *NEWS*-2 (Mayorga et al. 2010), and reflects mainly the denitrification. The fraction of DIN retention in rivers is calculated as a function of sub-basin area. In this study we also consider river retentions of DIP (e.g., by sedimentation processes). For DIP we assume a 50 % retention for all sub-basins and years based on Strokal and de Vries (2012). This reflects the net effect of different retention processes (e.g., accumulation of P in sediments due to its binding by iron). We discuss this assumption in "Strengths and weaknesses of the modeling approach at the sub-basin scale" section.

Nutrient retentions in dammed reservoirs of each sub-basin ($D_{F.j}$) are calculated following Global *NEWS*-2 (Mayorga et al. 2010): first, nutrient retention is quantified for each reservoir and then averaged over the sub-basin using actual water discharge (after water is removed for consumption) at the outlet of the sub-basin (see Box A.1 and Table A.1 in Supplementary Materials for details).

Nutrient removal from the river system by water consumption ($FQrem_j$, for DIN and DIP) is a function of actual and natural (before water is removed for consumption) water discharges at the outlet of each sub-basin (Box A.1 and Table A.1 in Supplementary Materials) (Mayorga et al. 2010).

Model inputs

In this study, most of the model inputs are from existing gridded global datasets developed for the Global *NEWS*-2 model (0.5 × 0.5° longitude by latitude). Also model parameters are from Global *NEWS*-2, except for reservoir characteristics, which are from the Global Reservoir and Dam (GRanD) database. Below we describe the sources of model inputs and parameters in more detail.

Gridded datasets of the Global *NEWS*-2 model were developed in earlier studies (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009). Bouwman et al. (2009) and Van Drecht et al. (2009) prepared model inputs for diffuse (e.g., nutrient inputs to land from synthetic fertilizers, animal manure applications) and point (e.g., human waste production) sources of nutrients using the Integrated Model for the Assessment of the Global Environment (IMAGE) model. Fekete et al. (2010) prepared hydrological model inputs (e.g., runoff, water discharge) using the water balance model (WBM) model. These datasets are available for 1970, 2000 and 2050. And, they were implemented to the original Global *NEWS*-2 model (Mayorga et al. 2010; Seitzinger et al. 2010). For the year 2050, the storylines of the four Millennium Ecosystem Assessment (MEA) scenarios (Alcamo et al. 2005) were quantitatively interpreted by the IMAGE model to produce the gridded inputs (Seitzinger et al. 2010). These scenarios are Global Orchestration (GO), Adapting Mosaic (AM), TechnoGarden (TG) and Order from Strength (OS) (Alcamo et al. 2005; Carpenter et al. 2006; Seitzinger et al. 2010). These storylines address future trends in climate and hydrology (Fekete et al. 2010), and in nutrient management for agriculture (Bouwman et al. 2009) and sewage (Van Drecht et al. 2009). Detailed descriptions of the scenarios can be found in various sources (Alcamo et al. 2005; Bouwman et al. 2009; Carpenter et al. 2006; Fekete et al. 2010; Seitzinger et al. 2010; Van Drecht et al. 2009).

We used these existing gridded datasets to derive the following variables for Pearl River sub-basins for 1970, 2000 and 2050 (Box A.1 in Supplementary Materials; Table 1): sub-basin areas, land use (agricultural areas and non-agricultural areas), nutrient inputs to watersheds (land) from diffuse ($WSdif_{E.y.j}$) and point sources ($WShw_{E.y.j}$), total population and population with a sewage connection (I_j), annual runoff from land to streams and water discharges at the sub-basin outlets. We applied ArcGIS functions to aggregate gridded information to the sub-basin scale (for details see Table A.1 in Supplementary Materials). For 2050 we used the gridded dataset prepared based on the GO scenario because it assumes a globalized trends in socio-economic development with a reactive approach towards environmental management. Economic growth is driven by globally connected markets. Population growth is moderate because GO assumes low fertility (e.g., two births per woman in China), and mortality as a result of better education (Alcamo et al. 2005). Investments in education and infrastructure are assumed to be high in this world (Alcamo et al. 2005; Carpenter et al. 2006). People can migrate across national borders because of the globally connected society, resulting in higher migration rates (Alcamo et al. 2005). Also the

Table 1 An overview of model variables and parameters

Variable/parameter	Varying among			Source
	Sub-basins	Nutrients	Years	
Total area	X			A
For diffuse sources				
Agricultural area	X		X	A
Watershed diffuse sources and watershed export	X	X	X	A
Watershed export constant		X		B
Diffuse export coefficient	X			B
For point sources				
Watershed point sources	X	X	X	Q
Nutrient removal during sewage treatment	X[a]	X	X[a]	B
Total population and population with sewage connection	X		X	A
For hydrology and reservoirs				
Runoff, actual and natural water discharges	X		X	A
Data for each reservoir/dam: volume, depth and water discharge	X	X	X	C
Retention in reservoirs	X	X	X[b]	Q
Retention in the river	X	X[c]		Q
Runoff shape constants		X		B

X indicates whether variables/parameters vary among sub-basins, nutrient elements (N, P) or years

Blanks indicate that model variables/parameters do not vary among sub-basins, nutrient elements (N, P) or years

Sources A is global datasets at 0.5 by 0.5° (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009); B is basin scale information from Global *NEWS*-2 (Mayorga et al. 2010); C is Global Reservoir and Dam database (GRanD) (Lehner et al. 2011a, b). Q means calculated as indicated in the text. For details see Table A.2

[a] Nutrient removal is the same for the Zhujiang River sub-basins (Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta). Values for 1970 are from Global *NEWS*-2 (Mayorga et al. 2010). Values for the Dongjiang sub-basin are assumed for N to be 0.8 in 2000 and 2050, and for P to be 0.8 in 2000 and 0.9 in 2050 based on expert knowledge (see text)

[b] Nutrient retention in reservoirs of each sub-basin ($D_{F,j}$) is calculated for each reservoir based on input data from GRanD database, and then averaged for sub-basins using actual water discharge at the outlet of the sub-basin for 1970 and 2000. For 2050, reservoir-specific input data are not available. We assumed a fixed ratio between 2000 and 2050 for D_{DIN} and D_{DIP} for sub-basins derived from Global *NEWS*-2 (Mayorga et al. 2010) for the Zhujiang and Dongjiang basins (see Fig. 1). The factor was calculated for these basins as: (value of 2050 minus value of 2000)/value of 2000. This way, we calculated a factor of 1.7 increase for the sub-basins of the Zhujiang basin (the Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta) and 3.0 for the Dongjiang sub-basin

[c] Nutrient retention in the river (e.g., via denitrification for DIN, sedimentation processes for DIP) is calculated as a function of sub-basin area (see Box A.1) for DIN. For DIP, we assumed 0.5 based on Strokal and de Vries (2012)

urban population is assumed to increase as a result of migration of people from rural to urban areas for better jobs (Alcamo et al. 2005; Van Drecht et al. 2009). Society will have access to better sanitation and improved sewage wastewater treatment. Thus more people are assumed to be connected to sewage systems (Van Drecht et al. 2009). This may increase nutrient inputs to rivers from sewage unless sewage treatment is effective enough to reduce nutrients. Food production will be diversified to meet the food demand of a growing world population (Bouwman et al. 2009). This is associated with increased nutrient inputs to agriculture. The demand for energy and irrigation will drive construction of dams (Fekete et al. 2010), influencing nutrient retention in the river network. In this GO world, the economy is generally considered more important than the environment. Environmental problems will be addressed only when they occur. Local environmental problems will be difficult to solve due to globalization trends (Alcamo et al. 2005; Carpenter et al. 2006).

Basin scale information from Global *NEWS*-2 (Mayorga et al. 2010) was taken for the following model parameters: watershed export constant (needed to calculate $FE_{ws.F}$), diffuse export coefficient (needed to calculate DIP from weathering), constants determining runoff curve (needed for $FE_{ws.F}$) and nutrient removal during sewage treatment ($hw_{frem.F}$, needed to calculate $RSpnt_{F.y.j}$) (see Eqs. 4, 5; Table 1 and Box A.1 in Supplementary Materials). These are basin scale parameters. Global *NEWS*-2 (Mayorga et al. 2010) divides the Pearl River into two basins: the Zhujiang (covering the Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta sub-basins in our study) and Dongjiang (Fig. 1b). We used the model parameters for the Zhujiang basin in Mayorga et al. (2010) for the Liujiang, Yujiang, Xijiang, Beijiang and Zhujiang delta sub-basins for 1970, 2000 and 2050. For the Dongjiang sub-basin we also adopted values from Mayorga et al. (2010), except for N and P removal during waste water treatment for 2000 and 2050. For 2000 we assumed an N and P removal fraction of 0.80, and for 2050 this fraction is assumed 0.80 for N and 0.90 for P. These removal fractions are considerably higher than in the other sub-basins (all <0.4 for N and <0.5 for P), reflecting the technologies currently implemented in the Hong Kong area (based on expert judgment).

The GRanD database (Lehner et al. 2011a, b) was used for model inputs for dams and reservoirs. It includes 64 dams constructed during the period of 1957–2000 within the Pearl River sub-basins (Table A.6 in Supplementary Materials). From the GRanD database we derived the following model inputs for each reservoir: volume, depth and water discharge (see Box A.1 and Table A.6 in Supplementary Materials). These inputs are needed to quantify DIN and DIP retentions in dammed reservoirs.

Model inputs for reservoirs for 2050 are not available at the sub-basin scale from the GRanD database. We, therefore, estimated the DIN and DIP retentions in reservoirs for each sub-basin ($D_{DIN.j}$, $D_{DIP.j}$, see Eq. 6) by assuming that the change between 2000 and 2050 at the sub-basin scale is the same as at the basin scale. To this end, we calculated the change in $D_{DIN.j}$ and $D_{DIP.j}$ between 2000 and 2050 from Mayorga et al. (2010) for the Zhujiang and Dongjiang basins (see Table 1).

Model validation

Global *NEWS*-2 was validated for world rivers (Mayorga et al. 2010) and for continents (Strokal and Kroeze 2013; Suwarno et al. 2013; Thieu et al. 2010; Van der Struijk and Kroeze 2010; Yasin et al. 2010) including the Chinese rivers (Qu and Kroeze 2010). Their results confirmed an acceptable performance of the model for DIN and DIP. For example, the Nash–Sutcliffe efficiency (R^2_{NSE}) is 0.54 for DIN and 0.51 for DIP export by large world rivers according to validation results of Mayorga et al. (2010).

In a recent study (Strokal et al. 2014b) we evaluated further the model for DIN and DIP export by Chinese rivers (including the Pearl River). We used the Pearson's coefficient of determination (R^2_P), R^2_{NSE} and Model error (ME) (see Moriasi et al. (2007) and Strokal et al. (2014a) for detailed descriptions). We calculated R^2_P of 0.96, R^2_{NSE} of 0.42 and ME of 18 % for DIN and DIP export by the Chinese rivers. We concluded that the model performance is good for the Chinese rivers. However, the model seems to overestimate yields of dissolved inorganic nutrients for China to some extent (Qu and Kroeze 2010) or/and underestimates retention of nutrients in river systems of large basins like the Pearl.

Here we evaluated the model at the sub-basin scale for the Pearl River. We compared modeled fluxes of DIN and DIP for 2000 with observations. Observations are available for DIN and DIP fluxes at the mouth of the Pearl River and at the outlets of the Dongjiang, Yujiang and Xijiang sub-basins. The observed fluxes (see Table A.3 in Supplementary Materials for the literature sources) were calculated from nutrient concentrations using reported water discharges and areas (see Table A.4 in Supplementary Materials for the literature sources).

Our modeled values are generally in line with the observed DIN and DIP fluxes (Fig. 3; Table A.3 in Supplementary Materials). We calculate about 430 kg km^{-2} $year^{-1}$ of DIN and 10 kg km^{-2} $year^{-1}$ of DIP inputs exported from all sub-basins of the Pearl River to the coastal waters. These values are in reasonable agreement with literature: 523–1148 kg km^{-2} $year^{-1}$ for DIN and 6.3 to 29 kg km^{-2} $year^{-1}$ for DIP (Fig. 3; Table A.3 in Supplementary Materials). For the Dongjiang sub-basin the modeled DIN and DIP fluxes at its outlet fall in the range of the observations (Fig. 3). The observations for DIN, however, vary greatly in the literature that may be associated with differences in the locations of measurement, selected the nutrient forms and time period considered. For example, we calculate around 2000 kg

DIN km^{-2} year^{-1}, while vary between 438 and 2864 kg DIN km^{-2} year^{-1} (Table A.3 in Supplementary Materials). For the Xijiang and Yujiang outlets only concentrations of ammonium (NH_4–N) were available (Table A.4 in Supplementary Materials). We, thus, calculated modeled NH_4–N fluxes for them (see Table A.3 in Supplementary Materials). Modeled values for these sub-basins are within the range of observed values (Fig. 3). For the Xijiang outlet we model 58 kg km^{-2} year^{-1} while observed values are in the range of 41–108 kg km^{-2} year^{-1}. Also for the Yujiang outlet we model fluxes (69 kg km^{-2} year^{-1}) within the observed range (4 and 87 kg km^{-2} year^{-1}) (Table A.3 in Supplementary Materials; Fig. 3).

Our validation results (Fig. 3) also indicate that the sub-basin model intends to underestimate DIN and DIP export by the Pearl River to the coastal waters. This may be caused by missing parameters in the model representing other sources of nutrients in rivers like direct discharges of animal manure to rivers (see discussion in "Strengths and weaknesses of the modeling approach at the sub-basin scale" section). In addition to model validation, we also verified some of our model inputs by comparing them with an independent Chinese county dataset (Fig. A.1; RESDC). This comparison shows a good agreement between two datasets: our gridded dataset and county dataset (R_P^2 is in the range of 0.82–0.99 for compared model inputs). We discuss this comparison in "Strengths and weaknesses of the modeling approach at the sub-basin scale" section.

Based on the validation results we consider the model applicable to the Pearl River. We realize that we rely on a limited number of observations. Some of these observations vary among literature sources (e.g., DIN observed yields for the Dongjiang, see Table A.3 in Supplementary Materials). This indicates that more measurements are needed to test the accuracy of the model. Despite these limitations, the comparison of modeled DIN and DIP values with the few available observations gives us an indication that the model performs in a satisfactory way. This is in line with earlier model validations at the scale of China, with a similar model at the basin scale (Qu and Kroeze 2010, 2012; Strokal et al. 2014b).

Results and discussion

Characteristics of sub-basins

The middle-stream sub-basins include the Xijiang and Beijiang, and cover half of the drainage area of the Pearl River. The remainder of the drainage area includes the up-stream sub-basins Yujiang and Liujiang (30 %) and the down-stream sub-basins Zhujiang delta and Dongjiang (20 %) (see Fig. 1; Table 2).

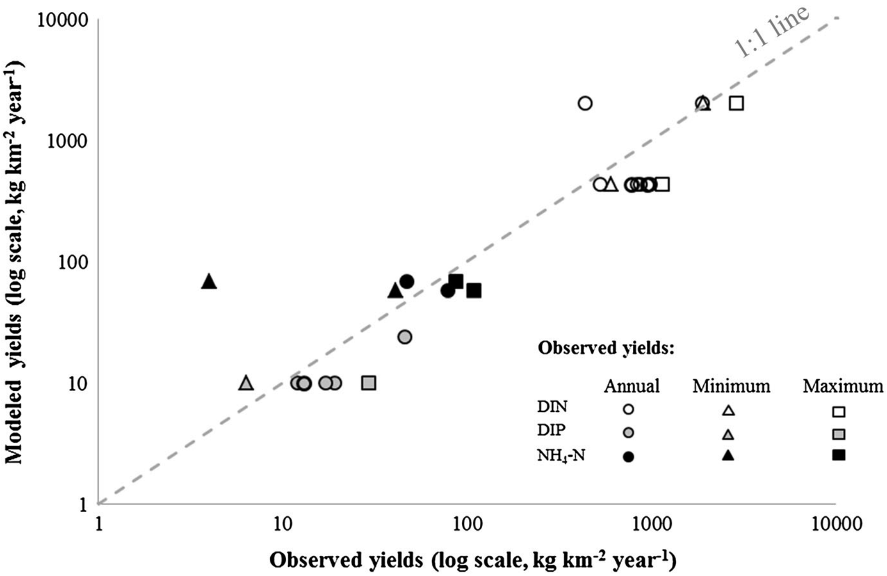

Fig. 3 Modeled versus observed yields of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) export by the Pearl River at the sub-basin scale (log scale, kg km^{-2} year^{-1}). Modeled ammonium (NH_4–N) yields were calculated assuming that 15 % of the total DIN is in ammonium form according to Meybeck (1982). Tables A.3 and A.4 in Supplementary Materials provide detailed information on the sources of observed yields

Agriculture is one of the main suppliers of N and P inputs to land (Fig. 4). In 1970 agriculture was less intensive than in 2000. Agricultural areas for crop and livestock production covered less than 10 % of the sub-basins except for the Xijiang where about half of the basin is covered by agricultural areas (Fig. 4). By 2000 these areas had expanded considerably in all sub-basins especially in the down-stream Dongjiang where almost all land was agricultural. This expansion of agriculture implied increased fertilizer use and manure excretion. The N inputs to land from synthetic N fertilizer use and animal manure increased by a factor of 12 in the Liujiang, Xijiang and Zhujiang delta sub-basins, and by a factor of 50–55 in the Yujiang and Dongjiang sub-basins between 1970 and 2000 (Fig. 4). Likewise, P inputs to land doubled in the Liujiang, Xijiang and Zhujiang delta sub-basins and increased about seven-fold in the Yujiang and Dongjiang sub-basins.

By 2050 the calculated agricultural areas are smaller than in 2000 in most sub-basins. This may seem surprising given the increasing population, but it can be explained by the assumed expansion of urban areas (Qu and Kroeze 2010, 2012). Nevertheless, N and P inputs to land may still increase (Fig. 4), indicating the intensity of agricultural practices in the future.

Inputs of N and P to rivers from agricultural sources (fertilizers and manure) depend on nutrient retentions in watersheds (land). In this study the watershed export fractions ($FE_{ws.F}$ in Table 2) account for these retentions. This fraction is lower for the up- and middle-stream sub-basins than for the down-stream sub-basins (Table 2). In other words, lower fractions of N and P inputs to land enter rivers in up-stream and middle-stream sub-basins. This can in part be explained by lower annual runoff from land to streams in those sub-basins (for details on hydrology see

Table 2 Characteristics of the sub-basins of the Pearl River: sub-basin area, population density, watershed ($FE_{ws.F}$) and river ($FE_{riv.F}$) export fractions for dissolved inorganic nitrogen (DIN) and phosphorus (DIP) in 1970, 2000 and 2050

Sub-basins	Area (1000 km^2)	Year	Population (inh km^{-2})[a]	Watershed export fraction ($FE_{ws.F}$, 0–1)[b]		River export fraction ($FE_{riv.F}$, 0–1)[b]	
				$FE_{ws.DIN}$	$FE_{ws.DIP}$	$FE_{riv.DIN}$	$FE_{riv.DIP}$
Liujiang	58	1970	89	0.38	0.05	0.32	0.42
		2000	136	0.38	0.05	0.30	0.22
		2050	137	0.47	0.07	0.28	0.07
Yujiang	77	1970	95	0.32	0.04	0.32	0.42
		2000	148	0.24	0.02	0.29	0.37
		2050	154	0.27	0.03	0.29	0.35
Xijiang	194	1970	121	0.38	0.05	0.30	0.24
		2000	186	0.38	0.05	0.23	0.06
		2050	187	0.47	0.07	0.18	0.07
Beijiang	31	1970	133	0.55	0.09	0.38	0.45
		2000	205	0.54	0.09	0.28	0.07
		2050	205	0.69	0.12	0.22	0.07
Zhujiang delta	48	1970	308	0.62	0.11	0.31	0.39
		2000	473	0.59	0.10	0.28	0.29
		2050	474	0.67	0.12	0.28	0.25
Dongjiang	34	1970	167	0.69	0.12	0.34	0.35
		2000	257	0.61	0.11	0.30	0.22
		2050	258	0.69	0.12	0.18	0.06

See "Model inputs" section for information on model inputs

[a] Average values were calculated as the total population of the sub-basin divided by the area of this sub-basin

[b] Values were calculated based on equations in Box A.1

Table A.5). We do not calculate large differences in watershed export fractions between past and future years. These export fractions, however, are much higher for DIN than for DIP. This is because P has generally stronger ability for accumulation is soils than N (Bouwman et al. 2009; Busman 1997; Schoumans and Groenendijk 2000).

Sewage is another important source of N and P in the sub-basins (Fig. 4). In this study sewage sources include wastewater from human excreta (for N and P) and detergents (for P: from laundry and dishwashers). In 1970 the sewage production of N and P in the watersheds of the Zhujiang delta sub-basin was higher (0.9 ton km^{-2} for N, 0.2 ton km^{-2} for P) than in the other sub-basins (about 0–0.4 ton km^{-2} for N, 0–0.08 ton km^{-2} for P). Between 1970 and 2000 N and P production doubled in all sub-basins. From 2000 onwards nutrient production from sewage may increase further under a globalized word with reactive environmental management (Fig. 4). An important reason of these increases is the growing population and economy (Qu and Kroeze 2010, 2012). The Zhujiang delta sub-basin is the most populated (around 300 inh km^{-2}) than the up-stream (about 90–95 inh km^{-2}), middle-stream (120–130 inh km^{-2}) and Dongjiang (about 170 inh km^{-2}) sub-basins in 1970 (Table 2). Between 1970 and 2000, the population in the Pearl River basin increased by around 55 % and may continue increasing in the future (Table 2).

The amount of N and P entering rivers from sewage sources depends on the number of people connected to sewage systems and the effectiveness of nutrient

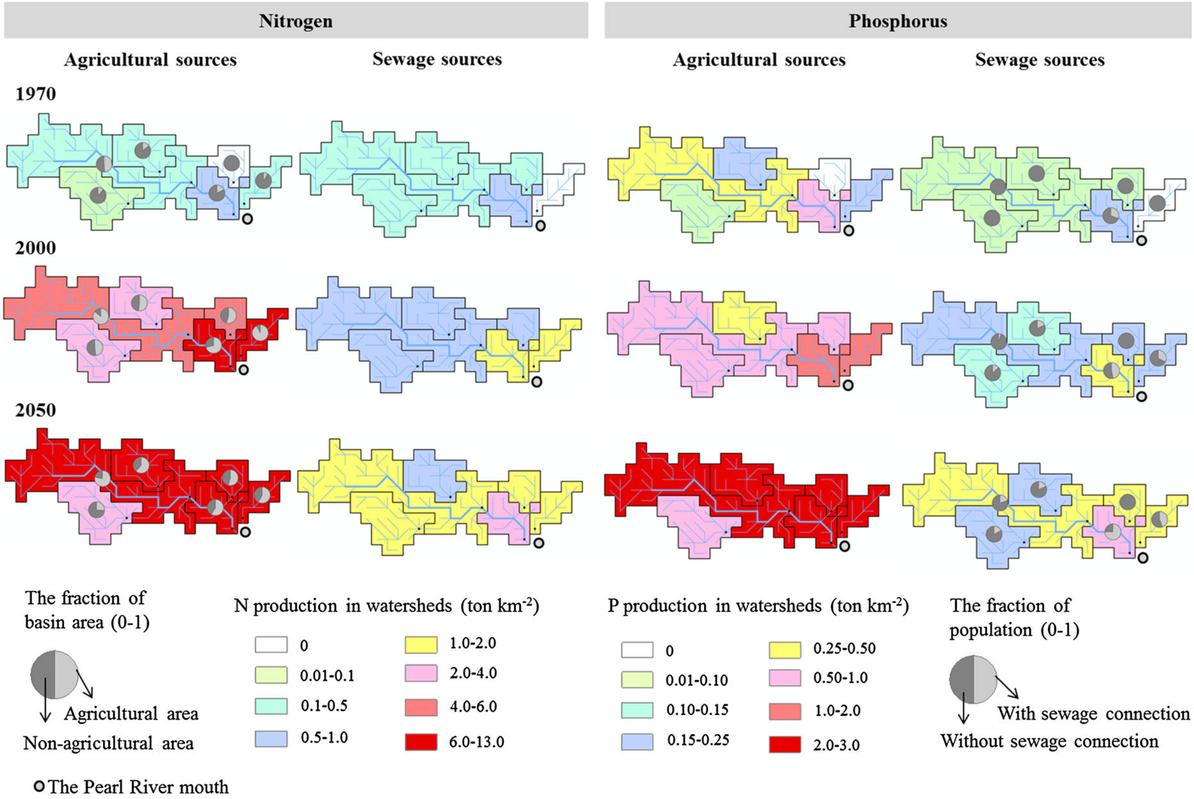

Fig. 4 Nitrogen (N) and phosphorus (P) production (as element, ton km^{-2} year^{-1}) in watersheds (land) of the Pearl River sub-basins from agricultural (WSdif$_{E.y.j}$ in Eq. 4) and sewage (WShw$_{E.y.j}$ in Eq. 5) sources in 1970, 2000 and 2050. Agricultural sources include animal manure excretion and synthetic fertilizer use. Sewage sources include wastewater from human excreta (for N and P) and detergents (for P). *Pie charts* represent the fractions of basin areas that are agricultural and non-agricultural (*left hand side*), and the fractions of population with (I$_j$ in Eq. 5) and without sewage connection (*right hand side*). Nutrient inputs for 2050 are based on the Global Orchestration scenario of the Millennium Ecosystem Assessment that assumes a globalized world with reactive management towards environment. "Model inputs" section and Table A.2 provide information on model inputs

treatment in these sewage systems (Van Drecht et al. 2009). In 1970 the number of people connected to sewage systems was very low, except for the Zhujiang delta sub-basin (Fig. 4). As a result N and P inputs from sewage systems were not discharged to rivers of those sub-basins. These nutrients were assumed to be lost or retained in the terrestrial system in line with Van Drecht et al. (2009). In contrast, about 15 % of the population in the Zhujiang delta sub-basin was with sewage connection in 1970 (Fig. 4), however, sewage treatment was absent. By 2000 more people became connected to sewage systems especially in the down-stream sub-basins (Fig. 4). N and P removal via sewage treatment was about 10 % for the sub-basins except for the Dongjiang (80 %). By 2050 almost two-thirds of the Zhujiang delta population and half of the Dongjiang population might be connected to sewage systems (Fig. 4). In other sub-basins, however, sewage connection is lower, especially in the Beijiang sub-basin (Table 2; Fig. 4). The efficiency of nutrient treatment is expected to increase in the coming years (see "Model inputs" section for model inputs).

The river network can retain considerable amounts of N and P, and this is influencing nutrient export to coastal waters. In this study the river export fraction ($FE_{riv.F}$ in Table 2) accounts for nutrient retentions within (via dam construction and in river retentions) and losses from (via water consumption) the river network (see Tables A.5 and A.6 in Supplementary Materials for details). This fraction ranges from 0.25 to 0.45 in 1970, depending on the sub-basin (Table 2). Between 1970 and 2000 this fraction decreased slightly for DIN for all sub-basins and decreased largely for DIP with larger decreases for the Liujiang, Xijiang and Beijiang (Table 2). The main reason of these decreases is the increasing number of dams along the river network between 1970 and 2000 (Table A.6). Between 2000 and 2050 the river export fraction may decrease further for the majority sub-basins because of envisaged damming of rivers (Table 2).

Another important factor influencing nutrient export to coastal waters is the distance that nutrients travel through a sub-basin to the coastal waters. For instance, DIN and DIP inputs to rivers of the up-stream sub-basins have to travel across middle- and down-stream sub-basins. The longer the transport takes, the more nutrients are retained within the river systems before reaching the coastal waters. Ye et al. (2012) studied the effects of nuttrient retentions in rivers and their transport to the outlet of the Vermilion Basin in east-central Illinois. They also reported that the traveling disctance of dissolved nutrients from up-stream to down-stream areas can affect their transport to the outlets.

Riverine inputs of dissolved inorganic N from sub-basins to coastal waters 1970–2050

We calculate that about 120 kton of DIN was exported to the coastal waters by the Pearl River in 1970 (Fig. 5, left column). By 2000 the total DIN inputs (from all sources) to coastal waters had increased by about 60 %. Between 2000 and 2050 the DIN inputs to coastal waters may further increase, reaching almost 250 kton in 2050 (Fig. 5, left column). Agricultural activities such as synthetic N fertilizer use and animal manure excretion are the main causes of these increases. The down-stream sub-basins, Zhujiang delta and Dongjiang, are dominant contributors of DIN inputs to the coastal waters (Fig. 5, right column). However, their contribution changes between past and future years (Fig. 5).

In 1970 agricultural sources contributed by 40 % to the total DIN inputs to the coastal waters. About 50–70 % of these agricultural DIN inputs originated from the Zhujiang delta sub-basin, about 20 % from the Dongjiang sub-basin and 10–20 % from the Xijiang sub-basin in 1970 (Fig. 5, middle column). By 2000 agricultural sources were already responsible for almost two-thirds of the riverine inputs of DIN (from all sub-basins) to the coastal waters (Fig. 5, left column). Between 1970 and 2000 the contributions of the Zhujiang delta to the agricultural DIN export decreased (45 %) while the contribution of the Dongjiang sub-basins increased (35–40 %). A decreased contribution of the Zhujiang delta sub-basin resulted from the net effect of increased N inputs to land from agriculture and increased N retentions in land as well as in rivers of this sub-basin ("Characteristics of sub-basins" section). The main reason of the increased Dongjiang contribution is the expanded agricultural areas during the period of 1970–2000 for crop and livestock production that increased N inputs to land ("Characteristics of sub-basins" section).

In 2050 agriculture is calculated to remain the dominant source of DIN in coastal waters (Fig. 5, left column). The Zhujiang delta sub-basin may transport more agricultural DIN to the coastal waters than in

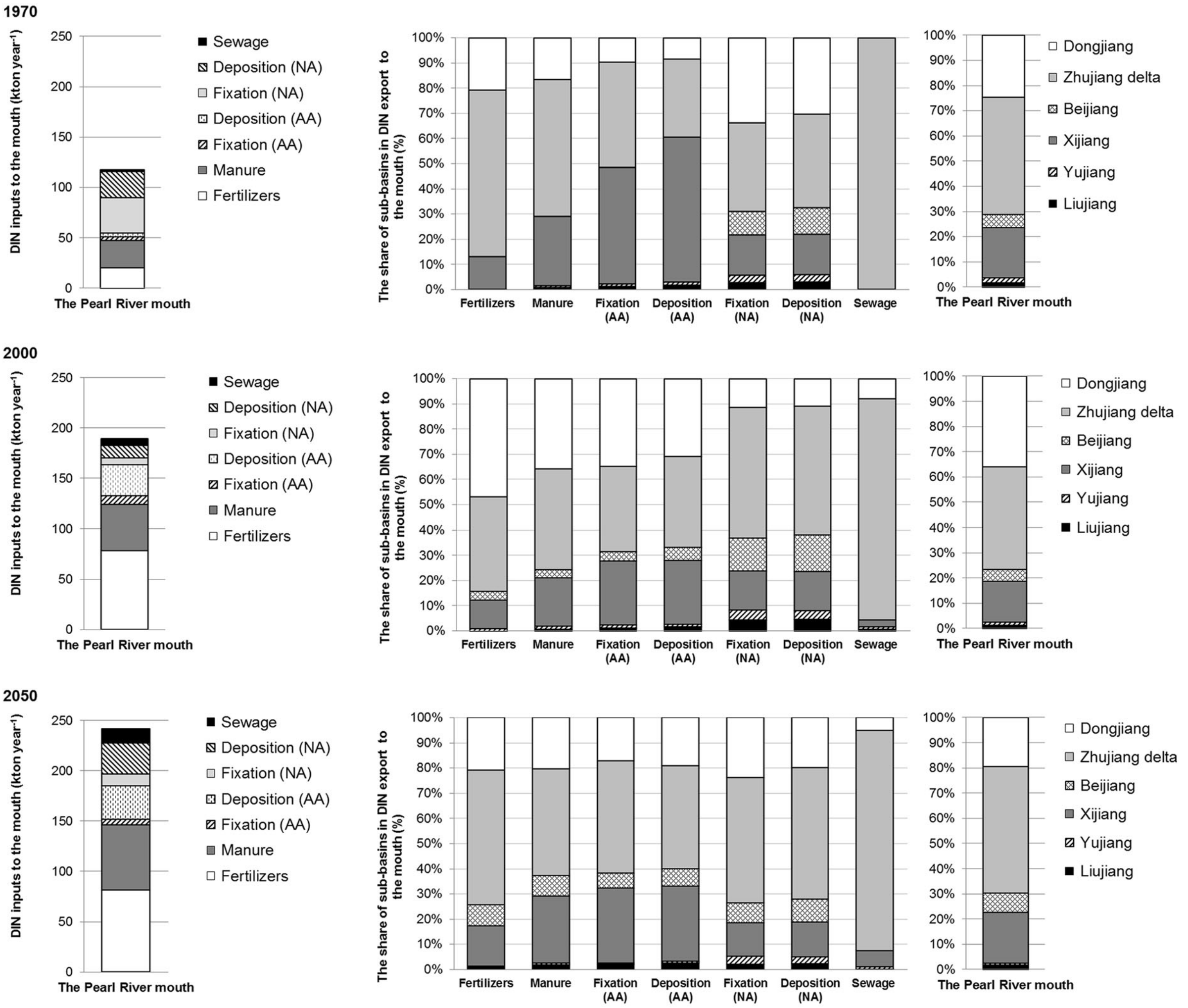

Fig. 5 Modeled inputs of dissolved inorganic nitrogen (DIN) to the coastal waters of the Pearl River (kton year^{-1}) by source categories (*left column*) and the share of Pearl River sub-basins (%) in DIN export from each source category (*middle column*) and in the total DIN export (*right column*) in 1970, 2000 and 2050. Future export is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment that assumes a globalized world with reactive management towards environment. *AA* agricultural area and *NA* non-agricultural area

2000 (Fig. 5) because of the projected further increases in N inputs to land from fertilizers and manure ("Characteristics of sub-basins" section). Inversely, the Dongjiang may export less agricultural DIN in coastal waters in 2050 than in 2000. An important reason for this is the projected decrease in N inputs to agricultural land and increase in DIN retentions in reservoirs of the sub-basins ("Characteristics of sub-basins" section).

Sewage is a relatively small anthropogenic source of DIN in the coastal waters of the Pearl River in 1970. Between 2000 and 2050 the share of sewage to the riverine inputs of DIN to the coastal waters calculated to increase. The Zhujiang delta sub-basin is a major contributor to these DIN inputs (Fig. 5). This might be explained by growing urban population and higher degree of sewage connection that increase DIN export to rivers from sewage systems unless the efficiency of N removal in public wastewater treatment plants is high enough to reduce DIN discharges to rivers of the Zhujiang sub-basin ("Characteristics of sub-basins" section).

In 1970 non-anthropogenic sources such as atmospheric N deposition on non-agricultural areas and biological N_2-fixation by natural vegetation were responsible for almost half of the total DIN inputs to the coastal waters. Between 1970 and 2000, and from 2000 onwards their share is calculated to decrease because of increased contribution of agricultural sources (see Fig. 5).

Our results indicate a minor contribution of the up-stream sub-basins and a relatively small contribution of the middle-stream sub-basins. This is the net effect of (i) spatial variations in land-based N sources, (ii) N retentions in watersheds (land) and in rivers, and (ii) traveling distances of N from sub-basins to the coastal waters. This holds for riverine inputs of DIP to the coastal waters as well (see "Riverine inputs of dissolved inorganic P from sub-basins to coastal waters 1970–2050" section). Details on sub-basin characteristics can be found in "Characteristics of sub-basins" section.

Riverine inputs of dissolved inorganic P from sub-basins to coastal waters 1970–2050

About 3 kton of total DIP inputs were exported to the coastal waters of the Pearl River from all sources and sub-basins in 1970. Between 1970 and 2000, these DIP inputs increased by one-third (4.5 kton in 2000) and may continue to increase from 2000 onwards (see Fig. 6, left column). These increases are the result of anthropogenic sources such as agriculture and sewage. Similar to DIN ("Riverine inputs of dissolved inorganic N from sub-basins to coastal waters 1970–2050" section), the Zhujiang delta sub-basin is the major source of DIP in coastal waters, and this holds for both agriculture and sewage (Fig. 6, right column). In contrast to DIN, about half of the total DIP inputs are from agriculture and another half from sewage (Fig. 6, left column).

In 1970 about 70 % of agricultural DIP inputs to the coastal waters were exported from the Zhujiang delta sub-basin, 20 % from the Dongjiang and 10 % from the Xijiang (Fig. 6, middle column). The share of these sub-basins is different in 2000. The contribution of the Zhujiang delta sub-basin decreased from 70 % (1970) to 50 % (2000) and of the Dongjiang sub-basin increased from 20 % (1970) to 40 % (2000) (Fig. 6, middle column). An important reason of the decreased contribution of the Zhujiang delta sub-basin is an increase in DIP retentions in the river system of this sub-basin because the number of dams doubled during the period of 1970–2000 ("Characteristics of sub-basins" section). An explanation for the increased contribution of the Dongjiang sub-basin is that P inputs to agriculture increased between 1970 and 2000 as a result of increased agricultural area (see "Characteristics of sub-basins" section) and this increased DIP export from land to rivers and coastal waters.

In 2050, two-thirds of the agricultural DIP inputs to the coastal waters are calculated to be from the Zhujiang delta sub-basin and the remainder from the Dongjiang (15 %) and Xijiang (15 %) sub-basins (Fig. 6, middle column). These results for the Dongjiang sub-basin can be explained by the net effect of decreased agricultural areas and increased P retentions in the river system due to damming of the river (see "Characteristics of sub-basins" section). For the Zhujiang delta sub-basin P inputs to agricultural land from P fertilizers and animal manure applications are projected to increase and this increases P export to rivers of this sub-basin and consequently to coastal waters ("Characteristics of sub-basins" section).

Sewage DIP inputs to the coastal waters are mainly from the Zhujiang delta sub-basin for all years (Fig. 6). The main reasons are: (i) this sub-basin is the most populated with higher degree of sewage connection than the others and thus (ii) the P inputs to sewage systems of this sub-basin are higher while the effectiveness of treatment is low compared to the Dongjiang ("Characteristics of sub-basins" section).

Strengths and weaknesses of the modeling approach at the sub-basin scale

As any other model, our sub-basin model has its weaknesses. An important source of uncertainty in the model is associated with model parameters and inputs that are based on assumptions and simplifications. For instance, the watershed export constants (eDIN, eDIP in Box A.1 and Table A.1 in Supplementary Materials) and runoff shape constants (aDIN, aDIP, bDIP in Box A.1 and Table A.1 in Supplementary Materials) do not change over time, and are assumed the same for each sub-basin (see Table 1). These constants are used to quantify watershed export fractions ($FE_{ws.F.j}$, Eq. 4) that reflect nutrient retentions in the watershed. We realize that this simplification affects the final result. However, we assume that the associated model error is relatively small. It should be noted that these constants were taken from Global *NEWS*-2 that was calibrated

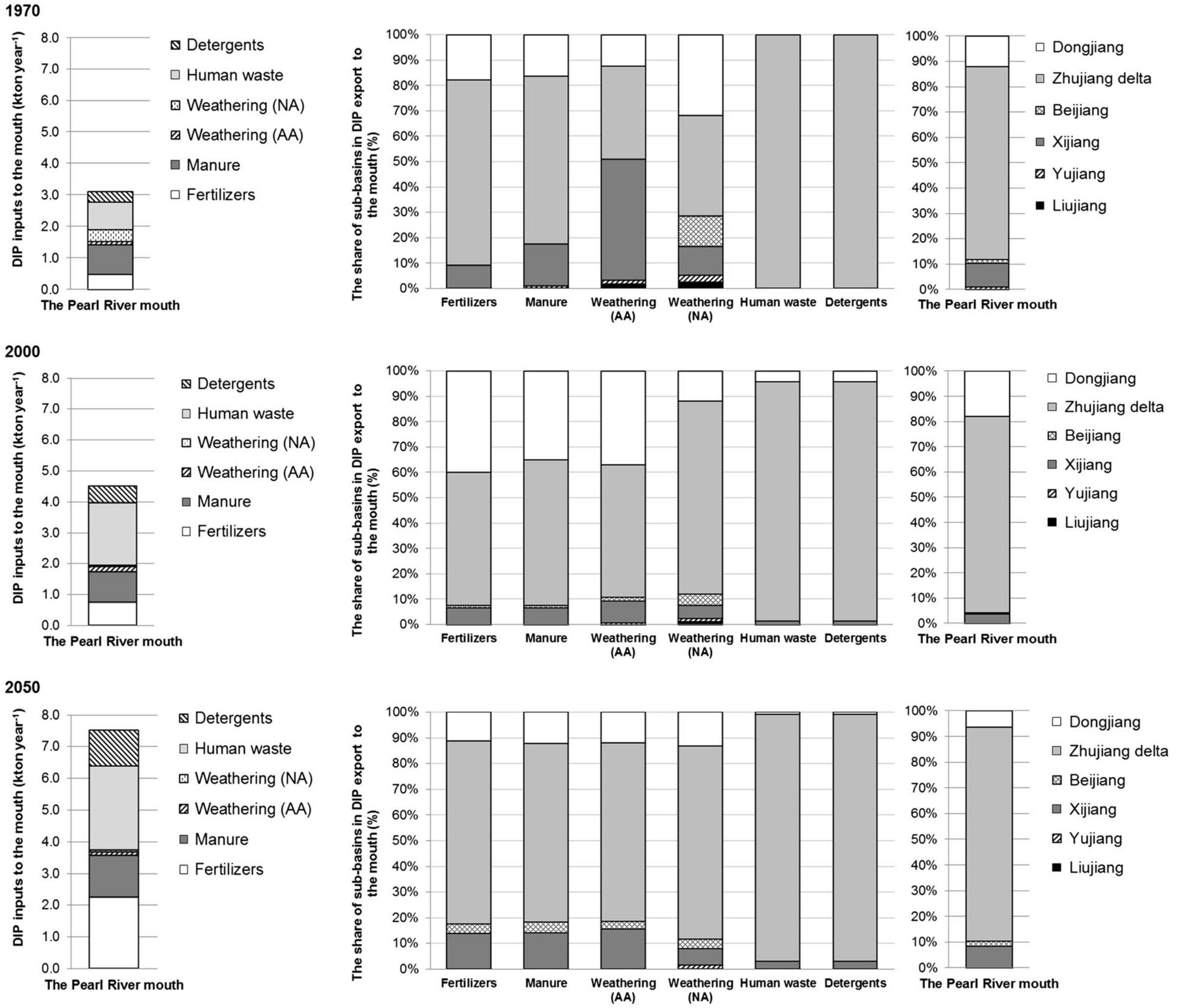

Fig. 6 Modeled inputs of dissolved inorganic phosphorus (DIP) to the coastal waters of the Pearl River (kton year^{-1}) by source categories (*left column*) and the share of Pearl River sub-basins (%) in DIP export from each source category (*middle column*) and in the total DIN export (*right column*) in 1970, 2000 and 2050. Future export is based on the Global Orchestration scenario of the Millennium Ecosystem Assessment that assumes a globalized world with reactive management towards environment. *AA* agricultural area and *NA* non-agricultural area

for large world rivers (Mayorga et al. 2010). No data exist for a calibration for our sub-basins. Therefore, we adopted these global values. Likewise, we used global values for the diffuse export coefficient (EC_{DIP} in Box A.1 and Table A.1 in Supplementary Materials) used to quantify DIP weathering in agricultural and non-agricultural areas.

We used most of the model inputs from the gridded global datasets of Global *NEWS*-2 to quantify nutrient export from sub-basins (Bouwman et al. 2009; Fekete et al. 2010; Van Drecht et al. 2009) (see "Model inputs" section). We, however, realize that these datasets were derived from large-scale (e.g., national) assessments (such as from FAO (2008), Bouwman et al. (2009)) and thus may contribute to uncertainties in the final results. This may be illustrated by the study by McCrackin et al. (2013), who compared predictions of Global *NEWS*-2 with SPARROW for the United States basins. Some disagreements in model predictions were found in particular for smaller basins because of differences in model inputs (e.g., regional US data in SPARROW, and global datasets in Global *NEWS*-2) and modelling approaches. They concluded that model inputs can be one of the uncertainty sources in the model predictions. We compared some of our sub-basin inputs to an independent Chinese county dataset ((RESDC), Wang M. pers. comm.) for the year 2000. We compared data on synthetic fertilizers, animal excretion, biological N_2-fixation by agricultural crops, and total human population. Figure A.1 indicates that there is a good agreement between our sub-basin inputs and sub-basin data derived from the county dataset. For example, R_P^2 is 0.82 (N) and 0.93 (P) for synthetic fertilizers, 0.97 (N) and 0.96 (P) for animal manure. For biological N_2-fixation R_P^2 is 0.98. For the total human population and sub-basin areas this parameter is calculated at 0.91 and 0.99, respectively (Fig. A.1). Furthermore, the drainage areas of the sub-basins delineated based on the global STN-30 network (Vörösmarty et al. 2000a) are in line with various hydrological studies for the Pearl River (Cui et al. 2007; Niu and Chen 2010; Zhang et al. 2007, 2008), and with the county dataset (Fig. A.1). We, thus, consider the gridded datasets acceptable for modeling nutrient export by the Pearl River at the sub-basin scale.

We realize that we used little information from independent regional datasets in our sub-basin model for the Pearl River compared to some other existing studies in the field of nutrient flows in China (e.g., Yan et al. (2010), Ma et al. (2012), Ti et al. (2012)). In contrast to our study, these existing studies analyze nutrient fluxes at a basin or provincial scale, using provincial information (e.g., from China Statistical Yearbooks) as one of the sources for their analyses. For our sub-basin analyses provincial data might not be a good choice because it should be first downscaled to sub-basins under certain assumptions and simplifications (two provinces cover about 80 % of the Pearl basin, see Fig. 1), and the data do not give enough information for 1970, 2000 and 2050. The county-based dataset, which was used for verifying some of our model inputs (see Fig. A.1), may be suitable for sub-scale modelling, but this dataset is not complete yet. Hence, in this study we used the gridded dataset because it provides the required inputs for our sub-basin model for the Pearl River.

Another source of uncertainty is associated with the very fast socio-economic development in parts of our study area, making it difficult to project changes over time. Future projections of nutrient export by the Pearl River depend on storylines of scenarios. In this study we used the globalized GO scenario with reactive environmental management. Details on storylines of GO can be found in Fekete et al. (2010) for hydrology, Van Drecht et al. (2009) for urbanization, and in Bouwman et al. (2009) for agriculture (see also "Model inputs" section). For the Pearl River this GO scenario assumes a moderate population growth (Table 2) with high economic development (e.g., a 10-fold increase in GDP at purchase power parity). Agricultural areas (except for the Beijiang and Liujiang sub-basins) may decrease (up to 46 %) between 2000 and 2050 because of urbanization that requires more land for cities. Nevertheless, agriculture is projected to be intensive for food security reasons. More people are assumed to be connected to sewage systems in 2050 with relatively low efficiencies of nutrient removal during treatment. All these are reflected in the final results, showing increasing trends in nutrient export by the Pearl River ("Riverine inputs of dissolved inorganic N from sub-basins to coastal waters 1970–2050" and "Riverine inputs of dissolved inorganic P from sub-basins to coastal waters 1970–2050" sections).

It should be noted that the storylines of GO were developed from large-scale assessments (e.g., national). This means that this scenario may not account for the fast economic development in large cities of the Pearl basin like Shenzhen (located in the Zhujiang delta sub-basin) and Hong Kong. An example of this is the assumed N and P removal in sewage treatment in the original GO scenario. In most of the Pearl River basin, nutrient removal in sewage treatment is relatively low. However, some urban areas develop relatively fast, for example in the Dongjiang sub-basin draining into Hong Kong. GO projects relatively low efficiencies of N and P removal for the Dongjiang sub-basin. This is not in line with the current developments in technology in, for example, Hong Kong. We, therefore, modified the original GO scenario, and assume a removal fraction for N and P for this sub-basin of 80–90 % in sewage treatment for 2050. This implies that the latest technology in sewage treatment is implemented in our study, based on our knowledge of the fast technical developments in this sub-basin. However, data for these parameters are not available in the literature.

Another model assumption that is relatively uncertain is the retention of DIP in rivers ($L_{DIP.j}$, Eq. 6). Basin-scale models often ignore retention of DIP in rivers, for example Mayorga et al. (2010) and Harrison et al. (2005). In reality, however, not all DIP is exported to the river mouth. We assumed that 50 % of DIP inputs to rivers is retained in rivers (e.g., via accumulation processes between sediment and water). This is based on Strokal and de Vries (2012) who showed that including river retention for DIP in Global *NEWS*-2 improves the model performance for large river basins in the world. They showed that 50 % is a reasonable estimate. We realize that in reality this fraction differs among water bodies and nutrient forms (Chen et al. 2010; Doyle et al. 2003; Reinhardt et al. 2005; Schulz and Köhler 2006; Ti et al. 2012).

In our model, human waste is considered a point source of nutrients in rivers, and animal manure a diffuse source. In reality, however, human wastes can also be a diffuse source (e.g., toilets that are not connected to sewage systems in rural areas) while animal manure can be a point source (e.g., direct discharge into surface waters) in some areas of the Pearl River such as the Beijiang sub-basin (expert judgment). Van Drecht et al. (2009) confirm that human waste can be a diffuse source in areas without sewage systems in China. Recently, Morée et al. (2013) quantified N and P inputs to agriculture from human wastes (as a recycling) on a country scale globally. Their analysis illustrates the importance of human waste as diffuse sources of nutrients in agriculture. Other studies (Bai et al. 2013; Hou et al. 2013; Ma et al. 2010, 2012, 2013) indicated considerable losses of nutrients to water bodies from animal manure systems in Chinese agriculture. Ti et al. (2012) indicated that 25 % of livestock and human waste are directly discharged to Chinese rivers. Even though human waste is a relatively small source of nutrients in rivers and direct inputs of manure to rivers may be exceptions, we suggest to include these sources in our model in the future.

We modeled DIN and DIP export for six sub-basins. We consider this level of detail arbitrary on the one hand, but appropriate on the other, given the scarcity of empirical data needed for validation, and our wish to identify the most important causes of coastal eutrophication in the past and the future. We argue that the chosen scale is probably optimal for the Pearl River, providing results of acceptable reliability that can be used to address the most important environmental issues. Larger scales like river basin-scale applied in the original Global *NEWS*-2 model are not sufficient to identify the sources of polluting nutrients in coastal waters for large river basins (see "Introduction" section). More detailed models such as the SWAT model are powerful tools, but only when they can be accurately applied. For the Pearl River upscaling of such detailed models may result in complex systems that are not transparent and with relatively large uncertainties.

Despite of the abovementioned weaknesses and limitations, we believe that our sub-basin model fits the purpose of our study. We developed the new sub-basin modeling approach that can be applied to large basins to assess effective management of coastal nutrient pollution. In this study we incorporated this approach to Global *NEWS*-2 and applied it to the Pearl River for the first time (see "Methodology" section on the methodology). There are four main strengths of the model.

First, our model is a spatially explicit model. A unique feature of the model is that it can quantify from where nutrients have been transported to the coastal waters (at sub-basin scale) and which human activities (e.g., agriculture, sewage) in these sub-basins are the sources of the nutrients. In other words, the model allows to quantify (1) the contribution of the main sources of DIN and DIP in rivers and coastal waters, and (2) the contribution of sub-basins to nutrient inputs to coastal waters. This was not done before for the Pearl River nor for other large world rivers. Various existing studies (Harrison et al. 2010; Moore et al. 2011; Yan et al. 2010; Yasin et al. 2010) quantify source attributions for nutrient export by large rivers on the basin scale. However, they do not give insights into locations of these nutrient sources within basins (see also "Introduction" section). Our model fills this gap. This information is essential for effective nutrient management. For example, this study provides insights that management of human activities in downstream areas of the Pearl River will likely be more effective to reduce coastal nutrient pollution than management in upstream areas (see conclusions). In addition, the model also addresses past and future trends in DIN and DIP export. This opens the opportunity to explore options to manage nutrients in the coming decades (e.g., via sensitivity and scenario analyses) that will enable to identify effective options.

Second, our sub-basin scale model is transparent and user-friendly. The model is developed for large basins such as the Pearl River. Thus, it is easy to apply this model to other large river basins in China (e.g., Yangtze and Yellow rivers) and worldwide. This is because many large basins are data poor and their large scale makes difficult to conduct empirical studies (see also "Introduction" section). Our model, therefore, offers the possibility to quantify the nutrient export while considering sub-basin characteristics and offers also the opportunity for up scaling to entire China.

Third, the model does not require a lot of input data. Most required inputs can be derived from the existing sources that we mentioned in "Model inputs" section. In addition, we verified some of the model inputs, and the results give us a confidence of using these sources in sub-basin modeling for the Pearl River.

Fourth, taking into account all the above mentioned strengths and weaknesses, we argue that our model is well balanced in terms of the accuracy of model results and uncertainties associated with poor data availability.

From our experience we may draw lessons for modeling nutrient export at the sub-basin scale:

1. Modeling nutrient export at the sub-basin scale for large poorly documented basins is possible;
2. The size of the sub-basins may seem arbitrary, however, it is appropriate for large data-poor river basins;
3. The model has its weaknesses, but model strengths overwhelm these weaknesses, resulting in a proper balance between the accuracy and uncertainty;
4. The model is a useful tool for sub-basin analyses of nutrient flows.

Conclusions

Coastal eutrophication has been increasing fast in the South China Sea since the 1970s. The underlying causes are human activities that increase exports of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) by the Pearl River. The objective of our study was to quantify DIN and DIP export from sub-basins of the Pearl River to coastal waters by source. To this end, we developed a sub-basin scale modeling approach for 1970–2050 to assess the relative shares of sources and sub-basins in the nutrient export at the river mouth. We applied this approach to the Global *NEWS*-2 (Nutrient Export from Watersheds) model. We used datasets of the Global *NEWS*-2 model to derive most of the model inputs (gridded 0.5 by 0.5° cell datasets), and parameters (river basin information) except for inputs needed to model nutrient retentions in reservoirs. For these retentions we used Global Reservoir and Dam databases. Future scenarios for 2050 are based on the Global Orchestration (GO) scenario of the Millennium Ecosystem Assessment (MEA), assuming a globalized world with reactive environmental management. The Pearl River basin consists of six sub-basins including up-stream sub-basins (Liujiang and Yujiang), middle-stream sub-basins (Xijiang and Beijiang) and down-stream sub-basins (Zhujiang delta and Dongjiang). Our study illustrates the importance of applying nutrient management in agriculture and sewage within down-stream sub-basins because of their large contribution to coastal eutrophication.

DIN inputs to the Pearl River mouth are calculated to double between 1970 and 2050 mainly because of agricultural activities in two down-stream sub-basins. In 1970 the down-stream Zhujiang delta contributed by 50–70 % and the down-stream Dongjiang by about 20 % to river export of agricultural DIN. By 2000 this percentage increased to about 35–50 % for the Dongjiang because of expanding agricultural activities to sustain the growing population. We calculate that from 2000 onwards the Dongjiang sub-basin may contribute less to DIN in the Pearl River because of decreasing N inputs to land from agriculture and increasing DIN retentions in reservoirs. This is different for the Zhujiang delta sub-basin with a decreasing contribution between 1970 and 2000, and an increase again by 2050 due to increasing N inputs to agricultural land. The share of the middle-stream Beijiang and Xijiang sub-basins to the total DIN inputs to the river mouth is projected to increase slightly from 2000 to 2050 mainly due to agricultural activities.

DIP inputs to the Pearl River mouth are calculated to increase 2.5-fold between 1970 and 2050. The main cause of this is sewage and agriculture in the down-stream sub-basins. About half of the DIP in the river mouth originates from sewage (human waste and detergents) and the other half from agriculture. The down-stream Zhujiang sub-basin is a major contributor to DIP inputs from sewage because of urbanization. Agriculture is a dominant source of DIP in the Zhujiang and Dongjiang sub-basins in 2000. In 1970 the Zhujiang contributed by about two-thirds to the agricultural DIP inputs to the mouth. The Dongjiang contributed about 20 %. In 2000 the contribution of the Dongjiang was higher than in 1970 while the contribution of the Zhujiang was lower. This is because the Dongjiang sub-basin is dominated by agriculture (90 % of the land was agricultural area in 2000) with an increasing use of P fertilizers and manure. Another reason is that the number of dams in the Zhujiang sub-basin doubled between 1970 and 2000, increasing DIP retention in rivers and reservoirs and thus decreasing DIP export to the river mouth. In 2050 the Zhujiang may contribute by about two-thirds to the total DIP inputs at the river mouth originating from agriculture. The share of the Dongjiang is projected to decrease due to decreasing agricultural areas and high DIP retentions in reservoirs of this sub-basin. The relative share of the middle-stream Xijiang sub-basin in DIP export by the Pearl river may increase in the future as a result of large increases in P inputs to land from agricultural activities.

The large contribution of down-stream sub-basins and very low (or zero) contribution of up- and middle-stream sub-basins to coastal eutrophication can be explained as follows. The first reason is the spatial variability of land-based sources of DIN and DIP. For instance, N and P production in watersheds from agricultural and sewage sources are generally higher in the down-stream sub-basins compared to up- and middle-stream once. Furthermore, down-stream sub-basins are densely populated areas with higher number of people connected to sewage. The number of people connected to sewage systems is another important variable in our calculations. In up- and middle-stream sub-basins the percentage of people connected to sewage systems does not exceed 15 % in 2000 and 2050. This explains partly their low (or zero) contribution to the total DIP export to the Pearl River mouth. The second reason is the retention of nutrients in river systems of the sub-basins and their hydrology. For instance, the up-stream sub-basins are characterized by lower annual runoff than down-stream sub-basins, leading to lower nutrient export from land to rivers. Considerable amounts of nutrients remain in rivers of the sub-basins due to dams, retention processes in waters and water withdrawal for different purposes. This implies that the fractions of nutrient inputs to rivers that are actually transported to coastal seas decreases with distance to the river mouth. Or, in other words, a larger fraction of nutrients entering rivers close to the river mouth reaches the coastal waters than of nutrients entering river more upstream.

Our study is a first attempt to quantify DIN and DIP export from sub-basins of the Pearl River to the coastal waters for 1970, 2000 and 2050. The chosen sub-basin scale is appropriate for the large Pearl River basin to identify the main sources of nutrient pollution and their locations (sub-basins). We identified the most polluting sub-basins that contribute largely to DIN and DIP in coastal waters of the Pearl River. Reduction strategies are most effective in these sub-basins. Clearly, nutrient management in agriculture and sewage in down-stream sub-basins may reduce coastal eutrophication more than this management in up-stream sub-basins of the Pearl River. Our modeling study can, therefore, support decision making on strategies to reduce DIN and DIP inputs to rivers and thus to avoid further eutrophication in the coastal waters of the Pearl River. This study can serve as an example for other large river basins, where allocation of nutrient management areas is required.

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