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Response of water quality to land use and sewage outfalls in different seasons

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1	<b>Response of water quality to land use and sewage</b>
2	outfalls in different seasons, considering
3	oxygen-demanding contaminants
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### 28 Abstract

29 To better manage water environment in highly polluted rivers, the impacts of land use, sewage outfalls and seasons on water quality should be investigated. When 30 considering the effects of oxygen-demanding contaminants, 31 the complex interdependencies were hard to describe by conventional methods. The Bayesian 32 33 Networks (BNs), in which each variable only depends on its immediate parent variables, can solve this problem. In this study, the BNs were developed to assess the 34 35 impacts of land use and sewage outfalls on Ammonia Nitrogen (AN) and Dissolved Oxygen (DO) concentration in the Huaihe River Basin (HRB) for different seasons 36 and spatial scales, where AN was selected as a typical oxygen-demanding 37 contaminant. The BNs made good agreements between observed and predicted values. 38 AN negatively affected DO concentration, which was more significant in dry seasons. 39 40 Land use and sewage outfalls data at local scale (less than 20km radii around monitor stations) gave the best explanations to variations in AN and DO concentration, which 41 reveals that controlling water contaminants sources at the local scale can improve 42 water quality efficiently. Wastewater from sewage outfalls was the strongest 43 contributor to AN pollution in dry seasons, which was weakened in wet seasons by 44 intensive dilution process. Farmland acted as "sink" for its storage capacity of 45 contaminants in dry seasons and as "source" in wet seasons. The transformations 46 between two processes were caused by the huge variations between surface runoff in 47 dry and wet seasons. Woodland and grassland positively influenced water quality, 48 therefore, these could be used as pollution buffers around rivers to protect the water 49 environment. Urban made a disproportionately strong contribution to water pollution, 50 which revealed that intensive anthropogenic activities exacerbate water quality 51

- 52 degradation. These results can enhance understanding in influence factors on water
- 53 quality and contribute to effective water environment management.

# 54 Keywords

55	Land use, Sewage outfalls, Ammonia nitrogen, Dissolved oxygen, Bayesian Networks,
56	Spatial scales
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#### 89 **1 Introduction**

The degradation of surface water quality has become a global environment issue, 90 which can be affected by many factors, such as vegetation characteristics, climate 91 92 change, rivers topography and land use in catchments (Ai et al., 2015; Wang et al., 2015; Williams et al., 2015; Bocaniov and Scavia, 2016; Chen et al., 2016; Julian et 93 al., 2016; Zieliński et al., 2016; Rodrigues et al., 2018; Shukla et al., 2018). Due to 94 95 huge amount of influence factors and complex processes involved (Carey et al., 2013; Selle et al., 2013), it is a challenge to assess the relationship between land use, sewage 96 outfalls and water quality as contaminants come from both point (discharged into 97 rivers by sewage outfalls) and non-point sources (transported by surface runoff from 98 land). However, scientific assessments of influence factors on water quality are 99 100 essential to implement effective strategies for better water environment.

101 Many previous studies analyzed the influence of land use on water quality. Some of them focused on the catchment scale (Keesstra et al., 2014; Ai et al., 2015; 102 103 Meneses et al., 2015; Julian et al., 2016; Van Eck et al., 2016; Rodrigues et al., 2018), while other researches compared the multiple spatial scales from local to catchment 104 (Dodds and Oakes, 2008; Tran et al., 2010; Monteagudo et al., 2012; Tudesque et al., 105 2014; Ward and Kaczan, 2014; Wang et al., 2015; Martin et al., 2017; Vrebos et al., 106 107 2017; Shukla et al., 2018). Some researches pointed out that land use at the catchment 108 scale had the most significant correlation to water quality while others found land use at local scales could give better explanations to variations in water quality indicators. 109 Moreover, other studies reported that different kinds of water quality indicators tend 110 to be affected by land use at different spatial scales (Chang, 2008; Hurley and 111 Mazumder, 2013; Delpla and Rodriguez, 2014; Chen et al., 2016; Ding et al., 2016). 112

113 Therefore, in order to get scientific evaluations of land use impacts on water quality, it114 is necessary to take different spatial scales from local to catchment into consideration.

Dissolved oxygen (DO) is one of the most important parameters in assessing 115 water quality and sustainability of ecosystems (Khan and Valeo, 2016; Heddam and 116 Kisi, 2018). It reflects the balance between oxygen-producing processes and 117 oxygen-consuming processes in rivers (e.g., chemical oxidation) (Fijani et al., 2019), 118 119 while a certain level of DO is essential for aquatic life to survive (Singh et al., 2009). DO concentration could be affected by many factors, such as land use, water 120 121 temperature and some water quality variables, which had been analyzed by many previous researches (Tran et al., 2010; Liu et al., 2012; Ai et al., 2015; Wang et al., 122 2015; Ding et al., 2016; McGrane et al., 2017; Vrebos et al., 2017; Missaghi et al., 123 2018; Shukla et al., 2018). Previous researches had revealed that Ammonia Nitrogen 124 (AN) is an important oxygen-demanding contaminant that had significant negative 125 influence on DO concentration through redox process and nitrification process (Singh 126 et al., 2009; Najah et al., 2011; Antanasijević et al., 2014; Zabed et al., 2014; 127 Rosecrans et al., 2017). Chen et al. (2016) also pointed out that nitrogen pollution, 128 especially ammonium, contributed to low DO concentration in rivers. However, the 129 potential effects of oxygen-demanding contaminants on DO level were neglected in 130 previous researches when analyzing the influence factors on DO. 131

Both of AN and DO can be affected by land use and seasons, besides, AN has effects on DO level as an oxygen-demanding contaminant. Moreover, point sources (sewage outfalls) also make strong contributions to AN pollution in receiving water, thus, they also should be taken into account. Describing these complex interdependencies between influence factors and water quality is a challenge, however, it is essential in better river basin management (Ward and Kaczan, 2014; Ai et al.,

2015). The conventional regression models, such as linear or non-linear models, are 138 hard to describe these complicated dependencies. The Bayesian Networks (BNs), 139 140 which haven't been fully understood and extensively applied in water environment researches (Korb K, 2004; Aguilera et al., 2011; Li et al., 2018; Wijesiri et al., 2018b), 141 can solve the problem by factorizing global probability distribution into local 142 143 probability distribution for each variable by the directed acyclic graph (DAG). In this 144 way, the particular variable can be modeled only depending on the information from its direct influence variables (parents variables). Moreover, the BN can provide an 145 146 approach to incorporate both effects of quantitative and qualitative variables into one model, and the season scenario (wet and dry seasons) is a qualitative variable in our 147 study. 148

In this paper, we analyzed the impacts of land use and sewage outfalls on water 149 quality in the Huaihe River Basin (HRB) in different seasons (dry and wet) and spatial 150 scales (from local to catchment). The HRB is a highly polluted river basin, where 151 many large-scale water pollution incidents occurred as a result of rapid social and 152 economic development and intensively anthropogenic activities. It affected the safety 153 of drinking water for about 10 million local residents (Zhao et al., 2012; Wang et al., 154 2014; Zhai et al., 2017; Xu et al., 2018). Based on the former researches, AN is the 155 most serious contaminant in this area (Zhai et al., 2014; Xu et al., 2018). Therefore, 156 157 AN and DO were selected as two typical water quality indicators to analyze in this paper and AN is the typical oxygen-demanding contaminant which has a significant 158 influence on DO level in rivers. Moreover, as longitudinal data (data collected over a 159 period in time), which includes information in changed influence factors and water 160 quality, can increase the reliability in model results when comparing to cross-sectional 161 data (data collected at a single point in time) (Wijesiri et al., 2018a), the longitudinal 162

163 datasets (from 2000 to 2013) in the HRB were applied in our study.

The main objectives of this paper are to (1) develop BNs model to describe 164 complex interdependencies between influence factors and water quality in the HRB; 165 (2) find out the spatial scale that land use and sewage outfalls can explain the 166 variations in water quality indicators best in the HRB; (3) assess the influence factors 167 on water quality considering the oxygen-demanding contaminant (AN) in both dry 168 169 and wet seasons. This study will enhance understanding of the effects of land use, sewage outfalls and seasons on water quality, which is essential and meaningful for 170 171 effective water environment management.

#### 172 2 Material and methods

### 173 **2.1 Study area and monitor stations**

174 The Huaihe River Basin (HRB) is one of the most important basins in eastern China (Fig. 1 (a)), with a drainage area of 270,000 km<sup>2</sup>. It locates between latitudes 175 30°~36°N and longitudes 111°~121°E. The Main Reaches of Huaihe River (MRHR) 176 originates from the Tongbai Mountain in the Henan province, and runs through the 177 Anhui and Jiangsu province from west to east before flowing into the Hongze Lake 178 (Fig. 1 (b)) (Zhai et al., 2017; Xu et al., 2018). The population density in the HRB is 179 614 persons per square kilometer (Zhai et al., 2014), which is 5 times higher than the 180 national average population density. The HRB is intensively influenced by 181 anthropogenic activities (Zhai et al., 2017), especially in the MRHR and main 182 tributaries, such as the Sha Ying River (SYR) and Guo River (GR). 183

184 Twenty monitor stations were in the study area (Table 1), six of them (S1-S6) in 185 the MRHR, eight of them (S9-S16) in the SYR, and three of them (S18-S20) in the

- 186 GR. The other three stations (S7, S8, S17) lie in the Shi River (SR), Hong River (HR)
- 187 and Jia Lu River (JLR), respectively. Water quality samples were collected weekly or
- 188 monthly, while discharge data were measured daily at all monitor stations.

## *Table 1*

*Details of twenty monitor stations in the HRB.* 

Station code	Station name	Location	Longitude (°E)	Latitude(°N)
S1	Changtaiguan	MRHR	114.07	32.32
S2	Xixian	MRHR	114.73	32.33
S3	Huaibin	MRHR	115.42	32.43
S4	Wangjiaba	MRHR	115.60	32.43
S5	Wujiadu	MRHR	117.37	32.95
<b>S</b> 6	Xiaoliuxiang	MRHR	118.13	33.17
<b>S</b> 7	Tanjiahe	SR	113.97	31.90
<b>S</b> 8	Bantai	HR	115.07	32.72
S9	Gaocheng	SYR	113.13	34.40
S10	Huaxing	SYR	113.67	33.92
S11	Huangqiao	SYR	114.45	33.77
S12	Zhoukou	SYR	114.65	33.63
S13	Huaidian	SYR	115.08	33.38
S14	Jieshou	SYR	115.35	33.27
S15	Fuyang	SYR	115.83	32.90
S16	Yingshang	SYR	116.28	32.65
S17	Fugou	JLR	114.40	34.07
S18	Boxian	GR	115.87	33.80
S19	Guoyang	GR	116.22	33.52
S20	Mengcheng	GR	116.55	33.28



*Fig. 1.* (a) The map of China. (b) The map of the Huaihe River Basin (HRB) with locations of twenty monitor stations.

#### 196 **2.2 Data sources and processing**

The water quality datasets (including AN concentration, DO concentration and 197 198 water temperature) and amount of AN contaminants from sewage outfalls from 2000 to 2013 were provided by the Monitoring Center of Huai River Water Resource 199 Protection Bureau. All water quality variables were measured following the national 200 standard methods of water quality testing (Water quality-Determination of 201 202 ammonia-Distillation and titration method, 1987; Water quality-Determination of dissolved oxygen-Electrochemical probe method, 1987; Water quality-Determination 203 204 of water temperature-Thermometer or reversing thermometer method, 1991; Water quality-Determination of ammonia nitrogen-Distillation-neutralization titration, 2010; 205 Water quality-Determination of dissolved oxygen-Electrochemical probe method, 206 2010). The discharge dataset was collected from the hydrographic office of Huaihe 207 River Commission of the Ministry of Water Resources, P. R. C. In order to assess 208 influences of land use and sewage outfalls on water quality in different seasons, the 209 dataset from October to next March were set to be the dry season and from April to 210 September were set to be the wet season according to climate conditions in the HRB 211 (Chen et al., 2016). Subsequently, average AN concentration, DO concentration, water 212 temperature and amount of AN contaminant from sewage outfalls in dry and wet 213 seasons were calculated over the research period. 214

The digital elevation model (DEM) at 90 m  $\times$  90 m resolution and land use 215 map in 2000, 2005 and 2010 were collected from the Data Centre for Resources and 216 217 Environmental Science, Chinese Academy of Sciences (RESDC, http://www.resdc.cn/). The locations of monitor stations, land use, stream networks 218 and DEM data were transformed to GIS layers by ArcGIS 10.5 (ESRI Company, 219

Redlands, California, USA) for the HRB under the Gauss-Kruger projected coordinate system. Based on stream networks and topographical features extracted from the DEM, the HRB was delineated into twenty sub-catchments. Each monitor station is outlet point in the corresponding sub-catchment.

To obtain the spatial scales at which land use and sewage outfalls data could give 224 225 the best explanations to variations in AN and DO concentration, we took seven spatial 226 scales (from local to catchment) into consideration. The six local scales are 10km, 15km, 20km, 30km, 40km and 50km radii around each monitor station in 227 228 corresponding sub-catchment, and the catchment scale is entire upstream catchment (EUC) for each station. The demonstration of EUC, 50km and 20km scale were 229 shown in Fig. 2 (a), (b) and (c), respectively (Hurley and Mazumder, 2013; Delpla and 230 Rodriguez, 2014). Six categories of land use were considered in our study: woodland, 231 grassland, water, urban, rural resident land and farmland (Fig. 2). The land use types 232 and inclusions were shown in Table S1. The percentage of land use area in 2000, 2005 233 and 2010 and the amount of AN contaminants from sewage outfalls were extracted in 234 a cumulative manner at seven spatial scales (Bostanmaneshrad et al., 2018). As few 235 changes in the percentage of land use had happened in all spatial scales and land use 236 data were available in only three years, land use in 2000, 2005 and 2010 are used to 237 match the dataset from 2000 to 2003, from 2004 to 2008 and from 2009 to 2013, 238 respectively. Because the data used were from different sources and had a huge 239 difference in ranges and magnitudes, we scaled all dataset before feeding into the 240 models following the standardized method recommended by Fijani et al. (2019). 241

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Fig. 2. Three spatial scales, (a) the entire upstream catchment (EUC), (b) 50 km radii around monitor stations and (c) 20km radii around monitor stations, used for land use and sewage outfalls data extraction. Different colors represent different land use types. The data in 10km, 15km, 30km and 40km radii scales were extracted

*similarly*.

#### 250 **2.3 Methods**

251 The Bayesian Network (BN) is a graphical approach including nodes and arrows which represent random variables (continuous and/or discrete) and probabilistically 252 conditional dependencies between variables, respectively. The structure of the BN can 253 be revealed by a directed acyclic graph (DAG) which defines a factorization of global 254 probability distribution into a set of local probability distributions for each variable 255 following the Markov Property. Therefore, each variable only depends on its 256 257 immediate parent variables, which could describe the complex system in a simple way. The BN model is a two-step approach, which first learns the model structure using 258 Structure Learning Algorithms. Then based on local conditional dependencies, it 259 estimates the conditional regression coefficients or conditional probabilities for 260 continuous variables or discrete variables, respectively. The advantage of BN is that 261 262 each local conditional function could be considered without explicit information in global probability distribution (Korb K, 2004; Scutari, 2010; Li et al., 2018; Wijesiri 263 264 et al., 2018a).

In this study, the BNs were developed to describe the complex interdependencies between land use, sewage outfalls and water quality indicators considering effects of oxygen-demanding contaminants at seven spatial scales in the HRB (Fig. 3). Accordingly, six land use categories, seasons and sewage outfalls were factors that influenced AN concentration, while land use, water temperature, seasons and AN (a typical oxygen-demanding contaminant) concentration were factors that affected DO concentration.

In order to conform the conditional Gaussian distribution, the two water quality indicators (AN and DO) were taken by log-transformation. The log-transformed concentration of AN and DO, water temperature, the proportion of six land use and AN amount from sewage outfalls were fed into the BNs as quantitative (continuous) data, while seasons were fed as qualitative (discrete) data, namely, "dry season" and "wet season" scenarios. Then, conditional regression coefficients can be estimated from the BNs and then impacts of all factors can be evaluated. The BNs were developed by "bnlearn" package (Scutari, 2010) in the R statistical computing platform (Team, 2016), which is a common program in statistical analysis.

281 In order to find the spatial scales that at which land use and sewage outfalls can give the best explanations to variations in AN and DO concentration, the 282 283 goodness-of-fit of models were evaluated at seven spatial scales by Pearson's correlation coefficients (Cor, Eq. 1) and Nash-Sutcliffe efficiency coefficients (NSE, 284 Eq. 2) (Nash and Sutcliffe, 1970). According to the recommendation from Moriasi et 285 al. (2007), when the NSE of a model is higher than 0.5, the model can be viewed as 286 acceptable. Accordingly, the best fitted BN model and the most correlated spatial 287 scale can be selected with the highest Cor and NSE. 288

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$$Cor = \frac{\sum_{i=1}^{N} [(obs_i - \overline{pred}) \times (pred_i - \overline{pred})]}{\sqrt{\sum_{i=1}^{N} (obs_i - \overline{obs})^2} \times \sqrt{\sum_{i=1}^{N} (pred_i - \overline{pred})^2}}$$
(1)

290

291 
$$NSE = 1 - \frac{\sum_{i=1}^{N} (obs_i - pred_i)^2}{\sum_{i=1}^{N} (obs_i - \overline{obs})^2}$$
 (2)

where N is the number of data points;  $obs_i$  and  $pred_i$  are the *i*th observed and predicted value;  $\overline{obs}$  and  $\overline{pred}$  are the mean of observed and predicted value, respectively.



**Fig. 3.** Structure of the Bayesian Network (BN) for modelling AN concentration as a function of land use, seasons and sewage outfalls, while modelling DO

296 concentration as a function of land use, seasons, water temperature and AN concentration, where AN is a typical oxygen-demanding contaminant.

### 297 3 Results and discussion

#### 298 **3.1 Spatial patterns of AN and DO concentration in dry and wet seasons**

The two water quality indicators had different patterns in different seasons in the 299 HRB (Table S2). AN concentration at all stations were higher in dry seasons than in 300 wet seasons, because increasing discharge in wet seasons contributed to dilute AN 301 302 pollution. The lowest AN concentration was observed at S1, which is the headwater station in the MRHR. AN concentration at the stations in the MRHR (S1-S6) and the 303 304 HR (S8) was relatively lower than stations in other tributaries, and JLR (S17) station 305 had the highest AN concentration in both dry and wet seasons. It is corresponding to 306 results from Xu et al. (2018) that water quality in the MRHR was better than that in tributaries. 307

308 DO concentration at all stations were higher in dry seasons than in wet seasons. As higher DO level means healthier ecosystem, this result implies that there are more 309 challenges existing in managing water environments in wet seasons in the HRB. DO 310 concentration at the headwater station (S1) was the highest in both dry and wet 311 312 seasons, while at S11 in the SYR was the lowest among all stations. Stations in the 313 MRHR had slightly higher DO concentration than the other stations. Based on these two water quality indicators, monitor stations in the MRHR, especially the headwater 314 station, had the best water environment condition in the HBR. 315

**316 3.2 Model performances at different spatial scales** 

# 317 The Cor and NSE of BNs at seven spatial scales were used to select the most

318 significantly influenced spatial scales (Fig. 4). Proportions of land use types (Table S3)

and amounts of AN pollution from sewage outfalls (Table S4) were different across

the seven spatial scales from local to catchment. Land use and sewage outfalls in 320 relatively small local scales (10km, 15km, 20km radii around monitor stations) can 321 give better explanations to variations in AN concentration than in relatively larger 322 scales (30km, 40km, 50km radii around monitor stations and EUC), while the best 323 fitness of observed AN concentration is in 20km scale with the highest Cor and NSE. 324 Similar results had been reported by several previous studies. Dosskey et al. (2010) 325 326 pointed out that the river buffer zone is the most effective scale in reducing water contaminants from point and non-point sources. Tran et al. (2010) found that land use 327 328 in long distance from receiving water had low possibility to make strong effects on water quality than in shorter distance. Ding et al. (2016) reported that most of the 329 water quality parameters were explained better by land use in the catchment scale, 330 however, nutrients tend to achieve the best explanations in riparian scale. Besides, the 331 long residence time of groundwater associated with nitrification and denitrification 332 processes in riparian zone is also a potential reason for why land use and sewage 333 outfalls at small scales were more correlated to AN concentration (Meynendonckx et 334 al., 2006). 335

336 Land use in small local scales can better explain DO concentration than in relatively larger spatial scales in the HRB, which is similar to AN concentration. The 337 reason is that hydrological distances in relatively larger spatial scales (more than 338 20km radii around stations) provide enough contact time for reoxygenation processes, 339 then the water body obtains oxygen equilibrium again at normal conditions. This 340 341 result is consistent with Ding et al. (2016) that the best explanation to DO level was land use at the catchment scale in mountain catchments and at local riparian scale in 342 plain catchments. As the two most mountainous sub-catchments in the HRB are S7 343 and S9 (Table S5), which are headwater stations in the SR and SYR, the areas in 344

20km scale of these two stations (157 km<sup>2</sup> and 447 km<sup>2</sup>) approximate to the areas of the EUC (catchment scale) (157 km<sup>2</sup> and 625 km<sup>2</sup>). Therefore, the land use and sewage outfalls of these two stations in 20km scale are similar to that in the catchment scale.

Land use and sewage outfalls data at 20km scale gave the best explanation to variations in AN and DO concentration, thus, all subsequent analyses in this paper are based on this spatial scale. Our result indicates that the direction to improve water quality more efficiently in the HRB is to pay more attention to pollution at the local scale (less than 20km radii around monitor stations). Moreover, in order to analyze influence factors on water quality comprehensively and scientifically in a river basin, both of local and catchment scales should be taken into account.



356 scale
357 Fig. 4. Pearson's correlation coefficients (Cor) and Nash-Sutcliffe efficiency coefficients (NSE)
358 between observed and predicted AN and DO concentration from BNs at seven spatial scales. The
359 black lines are Cor and red lines are NSE.

The comparisons between observed and predicted values from the BN model at 20km, 50km and EUC scales are shown in Fig. 5. It is evident that the performances of BNs model in AN prediction (Cor=0.91, NSE=0.80) and DO prediction considering AN (Cor=0.88 NSE=0.72) are satisfactory in 20km radii around monitor stations, which are better than that in larger spatial scales. When comparing DO prediction with or without AN (Cor=0.60, NSE=0.36), it shows that AN, as an oxygen-demanding contaminant, had significant influence on DO concentration. AN could explain more than 30% variations in DO concentration, therefore, AN can't be ignored in assessments of influence factors to water quality.



EUC scale. (b) Observed against predicted DO concentration from BN models at 20km, 50km and EUC scale without AN influence. (c) Observed against predicted DO concentration from BN models at 20km, 50km and EUC scale with AN influence.

#### 377 **3.3 Influence factors on AN and DO in dry and wet seasons**

The contribution of each influence factor was calculated by parameters estimation from the BN model at the 20km scale for both dry seasons and wet seasons (Fig.6). The amount of AN from sewage outfalls (point sources) had the strongest contribution (26.2%) in AN concentration in dry seasons. Based on management strategies of sewage outfalls in the HRB, the wastewater was discharged continuously and relatively stable at normal condition, therefore, the effects of sewage outfalls were weakened by dilution process with increasing flows in wet seasons.

As an oxygen-demanding contaminant, AN was the most effective factor in 385 lowering DO level in dry seasons. The negative contribution had declined from 21.1% 386 in dry seasons to 4.1% in wet seasons, which was caused by sever AN pollution in dry 387 seasons and relative lower AN concentration by dilution processes in wet seasons. 388 389 This is consistent with the former researches that AN/nutrients had the significant negative influence on DO concentration from both nutrient-rich and organic 390 391 agriculture flows (Jalali and Kolahchi, 2009; Tran et al., 2010; Antanasijević et al., 2014). 392

Water temperature had significant negative influence on DO concentration with the similar contribution in both dry (-19.8%) and wet (-17.5%) seasons, which is consistent to previous researches (Mahler and Bourgeais, 2013; Zabed et al., 2014; Diamantini et al., 2018; Du et al., 2018; Heddam and Kisi, 2018). Accordingly, higher water temperature at all stations in wet seasons (Table S2) could be a potential reason for relatively lower DO level over that period.

Farmland had negative and positive relationship to AN concentration in dry seasons and wet seasons, respectively. It implies that farmland experienced different processes in different seasons. As flows were three times lower in dry seasons than

that in wet seasons (Table S2), AN contaminant coming from nitrogen fertilizers 402 possibly can't be transported into receiving water in dry seasons. Accordingly, it 403 would be stored in farmland or even infiltrated into groundwater, therefore, farmland 404 acts as "sink" in dry seasons. It had been reported by Martin et al. (2017) that the 405 legacy effects of land use which caused by groundwater could delay the arrival time 406 of nutrients to the receiving water. Increasing surface runoff in wet seasons had larger 407 408 transportation capacity, therefore, AN contaminant that was reserved in farmland in the former dry seasons and newly applied in wet seasons from nitrogen fertilizers 409 410 were both transported into receiving water. Thus, farmland becomes an important "source" of AN pollution in wet seasons. As the proportion of farmland in the HRB 411 was more than 70%, it had huge storage capacity of AN contaminants in dry seasons 412 and then exported a large amount of contaminants to rivers in wet seasons. 413 Accordingly, 21.7% negative and 23.7% positive contribution to AN concentration 414 was made by farmland in dry and wet seasons, respectively. Similarly, farmland had 415 only 3.7% negative contribution to DO concentration in dry seasons and increased to 416 17.6% in wet seasons. This result is different to the results from many previous 417 studies, while they reported that agricultural land/farmland had negative effects on 418 water quality, which only played a role of "source" to water contaminants 419 (Seeboonruang, 2012; Wan et al., 2014; Wang et al., 2015). However, Wijesiri et al. 420 421 (2018a) found the negative relationship between dryland/irrigated agriculture and nitrates, which reflected the "sink" processes of farmland. All of these previous 422 researches failed to reveal the transformations between "sink" and "source" processes 423 in dry and wet seasons. Some research found that agricultural land influenced 424 nutrients level in rivers and degraded water quality mainly by agricultural surface 425 runoff (Chen et al., 2016), which usually happened over storms or rainy seasons 426

(Miller et al., 2011; Bu et al., 2014). Thus, the potential reason for why previous 427 researches only found the unilateral relationship between farmland and water quality 428 could be that they failed to take seasonal influence (wet/dry) into consideration. 429 Accordingly, meteorological and hydrological conditions have significant differences 430 between different seasons, particularly rainfall and discharge, which often affect water 431 quality strongly (Korb K, 2004; Shrestha and Kazama, 2007). As farmland is the most 432 433 extensive land use type in the HRB and made great contributions to water pollution in wet seasons, it is important for governments to implement strategies in controlling 434 435 contaminants from farmland, such as cut down on the usage of nitrogen fertilizers and encourage farmers to plant crops in more environmental-friendly ways. 436

The rural resident land had positive influence on AN concentration and negative 437 influence on DO concentration in both dry and wet seasons. It is consistent to the 438 results from previous studies that rural resident land was positively correlated with 439 deteriorations in water quality, therefore, it decreased DO concentration in rivers (Cui 440 et al., 2016; Kändler et al., 2017; Shukla et al., 2018). The influences of rural resident 441 land on AN and DO were similar to point source, which were weakened in wet 442 seasons by the dilution process. It was possibly caused by that the contaminants from 443 human living and livestock in rural resident land were discharged directly into rivers 444 by wastewater. It is consistent with results found by Julian et al. (2016) that cattle 445 density, which was correlated to wastewater from livestock, was the primary predictor 446 for nutrients in rivers. 447

The influence of urban on AN and DO concentration both became stronger in wet seasons than that in dry seasons, which had the similar pattern with non-point sources. This result corresponds to the previous study which pointed out that urban land use was identified as a primary factor in nitrogen pollution during wet seasons (Chen et al.,

2016). Although urban only covered less than 3% area in the HRB, it was the most 452 significant factor on AN and DO concentration in wet seasons. These results are 453 consistent with Ai et al. (2015) and Wang et al. (2015) who reported that urban land 454 use was small in the percentage of all land use, however, it was identified to make the 455 most significant contribution to water pollution and exerted a significant effect on 456 water quality. The disproportionately strong influence was caused by the high 457 458 percentage of impervious surface coverage in urban, which was related to the human activities in this area. The impervious surface interrupts contaminants infiltrating into 459 460 soil and then mitigates soil retention process. With increasing surface runoff in wet seasons, more contaminants could be transported into receiving water (Cunningham et 461 al., 2010; Tu, 2011; Seo and Schmidt, 2012; Wang et al., 2015; Chen et al., 2016; 462 Meierdiercks et al., 2017). Therefore, the intensive anthropogenic activities in urban 463 could further exacerbate its role in water degradation. Wijesiri et al. (2018a) and 464 Shukla et al. (2018) also pointed out that human activities in specific land use had 465 stronger influence on water quality rather than changes in land use area and natural 466 drivers such as rainfall. 467

Water and woodland had positive influences on water quality in both dry and wet 468 seasons. As grassland didn't have a significant correlation with AN concentration 469  $(\alpha=0.05)$ , it was removed from the influence factors on AN concentration. Many 470 previous studies had found similar results (Bu et al., 2014; Cui et al., 2016; Ding et al., 471 2016; Kändler et al., 2017). The increasing percentage of water area means increasing 472 discharge that can dilute contaminants, thus, it had positive effects on water quality. In 473 addition, the self-purification function of rivers could make contributions to decrease 474 contaminants concentration (Khorsandi, 2015). Woodland and grassland had positive 475 influence on water quality due to its buffer capacity from vegetation for diffuse 476



and grassland around rivers could help to further improve water quality in the HRB.

pollution (Connolly et al., 2015). Therefore, the increasing percentage of woodland

481 *Fig.6. Response of AN and DO to influence factors under different seasons.* 



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To analyze impacts of land use and sewage outfalls on water quality considering oxygen-demanding contaminants in dry and wet seasons from local scale to catchment scale in the HRB, we developed BNs model to describe these complex interdependencies. The results showed that land use and sewage outfalls at local scales (less than 20km radii around monitor stations) explained the variations in AN and DO concentration best, which revealed that paying more attention to controlling water pollution from point and non-point sources within 20km scales can improve water
quality more efficiently. Key results/findings are summarized as follows:

(1) Wastewater from sewage outfalls was the strongest contributor to AN
pollution in dry seasons, whose influence was weakened in wet seasons because of
intensive dilution process by increasing discharge.

494 (2) Farmland acted as "sink" for its storage capacity of contaminants in dry
495 seasons and as "source" in wet seasons. The transformations between "sink" and
496 "source" processes were caused by huge variations between surface runoff in dry and
497 wet seasons.

498 (3) Woodland and Grassland have positive effects on water quality in both dry
499 and wet seasons, which could be used as pollution buffers around rivers to protect the
500 water environment.

(4) Urban and rural resident land played important roles in water quality degradation, especially, urban made a disproportionately strong contribution to water contaminants although it only covers less than 3% area in the HRB. It revealed that intensive anthropogenic activities would exacerbate the negative impacts of urban on water quality.

These results/findings highlight the importance to consider both local and 506 catchment scales in analyzing the impacts of land use comprehensively. Considering 507 508 interactions between water contaminants is also important to analyze influence factors on water quality, which was rarely studied before. In order to better manage water 509 environment, governments should pay more attention to controlling contaminants 510 from farmland and urban, especially in wet seasons. Moreover, as woodland and 511 grassland could be used as pollution buffers, the percentage of these two kinds of land 512 use should be increased around rivers. Although the study presented here was based 513

514	on the HRB, the BN model and approaches can also be applied in other polluted river
515	basins around the world.
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