Groundwater nutrient inputs into an urbanized tropical estuary system in Indonesia

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Submission date: 07-Jul-2021 12:36PM (UTC+0700)
Submission ID: 1616653119
File name: nputs_into_an_urbanized_tropical_estuary_system_in_Indonesia.pdf (3.39M)
Word count: 11338
Character count: 58419

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Article *in* Science of The Total Environment · February 2018
DOI: 10.1016/j.scitotenv.2018.01.281

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Science of the Total Environment 627 (2018) 1066–1079

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Groundwater nutrient inputs into an urbanized tropical estuary system in Indonesia

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HIGHLIGHTS

- Groundwater discharges 106,000 mol d^{-1} of DIN and 5000 mol d^{-1} of DIP to the river.
- Land use and environmental infrastructure correlates with nutrient dynamics.
- Medium populated cities can contribute to nutrient flux comparable with megacities.

GRAPHICAL ABSTRACT

ARTICLE INFO

Article history:
Received 5 December 2017
Received in revised form 27 January 2018
Accepted 27 January 2018
Available online xxxx

Editor: D. Barcelo

Keywords:
Nutrient
Water quality
Groundwater discharge
Estuary
Radon
Medium cities
Indonesia

ABSTRACT

Groundwater discharge is known to transport nutrients into estuaries at several locations around the world. However, few studies report groundwater-associated nutrient fluxes from tropical developing regions such as Southeast Asia, even though this area shows the strongest human modifications in the coastal zone worldwide. We investigated groundwater nutrient flux into two streams and estuaries (Awur and Sekumbu Bay) in the urban area of Jepara, Indonesia, and its relation with the land usage surrounding the estuaries.

We found that average concentrations of NO_3, NH_4, and PO_4 in Jepara's aquifer reached 145 µM, 68 µM, and 14 µM, respectively, and our results indicated that these were mainly originated from untreated sewage, agriculture, and manure input. Approximately 2200 ton N year^{-1} and 380 ton P year^{-1} were removed in the soil and aquifer before the nutrients were discharged into the river. The total groundwater discharge into the river and estuary was estimated to 461 × 10^3 m^3 d^{-1}, or up to 42% of the river discharge. Discharge of groundwater-associated NO_3 (72 × 10^3 mol d^{-1}), NH_4 (34 × 10^3 mol d^{-1}), PO_4 (5 × 10^3 mol d^{-1}), and additional surface runoff may contribute to eutrophication and a decrease of nearshore surface water quality. Nutrient concentrations in groundwater, river, and coastal seawater in the Jepara region are similar to those found in major urban areas in Southeast Asia, e.g. Manila and Bangkok, even though Jepara has smaller size and population. Thus, our results indicate that medium populated cities with highly modified regional land use can contribute a significant amount of nutrient discharge in the coastal area and should be included in global assessments of nutrient budget calculation.

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https://doi.org/10.1016/j.scitotenv.2018.01.281
0048-9697/

1. Introduction

Nutrient transport via groundwater discharge has been identified as a significant pathway for land-derived contaminants to estuaries and coastal seawaters at multiple locations, especially in regions with high anthropogenic activities (Burnett et al., 2007; Hwang et al., 2010; Lee et al., 2009; Taniguchi et al., 2008; Wu et al., 2013). Groundwater could be discharged across the sea floor as submarine groundwater discharge, or it could penetrate the subaerial aquifer close to the coast (e.g. into an estuary) and discharge as terrestrial surface water (Burnett et al., 2006). Here we use the term "coastal groundwater discharge" to address both phenomena simultaneously. Groundwater-derived nutrient fluxes to the coastal ocean may have an ecological impact such as eutrophication and algal blooms (Lee et al., 2009; Valiela and Costa, 1988). In some areas, nutrient loading via groundwater to the coastal area can be comparable with contributions from river water, as groundwater percentage in river discharge was overall estimated to 3–40% (Taniguchi et al., 2002), but examples of very low contributions (between 5 and 10%, Charette and Buesseler (2004); Dulaiova et al. (2006); Makings et al. (2014)) or very high contributions (up to 80%, Peterson et al. (2010)) underscore the wide range of local groundwater discharge to rivers.

Even though groundwater discharge has been studied by a lot of local studies around the world, studies in tropical regions are scarce (cf. Moosdorf et al., 2015). Groundwater-associated nutrient fluxes in tropical regions, in this case Southeast Asia, could be important due to wet humid climate, long coastlines, and favorable hydrogeological conditions, such as a high permeability of the aquifer and a high groundwater recharge (Burnett et al., 2007; Moosdorf et al., 2015). Tropical streams and estuaries are generally more sensitive to ecosystem modification than temperate ones, particularly from terrestrial based nitrogen loading, due to tropical vegetative capacity in retaining N (Downing et al., 1999). Southeast Asia also has high population density and is listed among the regions with strongest human modifications of the coastal zone worldwide (Elvidge et al., 1997). Previous studies in this region point to significant nutrient fluxes through groundwater into bays in Bangkok and Manila, two of the most populated capitals in Southeast Asia (Burnett et al., 2007; Taniguchi et al., 2008). Indonesia has a population of 255 million people and the second longest coastline in the world. Groundwater nutrient concentrations in some major Indonesian cities exceed the Indonesian standard limit for drinking water (Arthana, 2007; Delinom et al., 2009). Indonesia is also a hotspot of surficial nitrogen and phosphorus yields due to high population density and runoff per area (Seitzinger et al., 2005).Thus groundwater flux into Indonesian coastal seawaters (marine and estuarine) could be a pathway of nutrient transport that alters the environmental condition in the adjacent coastal areas. Previous groundwater discharge investigations in Indonesia identified individual locations of coastal groundwater discharge, but its quality and environmental effect in the coastal zones have not yet been evaluated (Bakti et al., 2012; Bakti et al., 2014; Lubis et al., 2011; Umezawa et al., 2009).

The city of Jepara is located in northern central Java, which is the most populated island in Indonesia. It is characterized by urban areas, intensive agriculture, aquaculture, and medium-sized industries. We hypothesize that anthropogenic activities affect groundwater and surface water nutrient fluxes in the Jepara area, and that the groundwater significantly contributes to coastal pollution. Two bays and their respective tributaries in Jepara were studied to identify the amount of groundwater discharge and its environmental implications in the adjacent coastal areas.

1.1. Study site

Jepara has a population of 1.2 million inhabitants (2014) with agriculture, fisheries, and small furniture industry as main source of income (Jepara Bureau of Statistics, 2016). Its geological setting is dominated by a strato-volcano (Mt. Muria, 1602 m) occupying the east part of Jepara. Most of the rivers flowing westward through the city of Jepara originate from this mountain. Since the coastal plains on which the city is built are bounded by Quaternary volcanoes, their alluvial products are mostly derived from redeposited volcanoclastic materials and caused the presence of extensive aquifers (Said and Sukrisno, 1988).

The sampling locations, Sekumbu and Awur Bay, are the closest bays to the city center of Jepara (Fig. 1). Sekumbu Bay coastline is 5.4 km long and it is fed by two rivers and two small channels. In Sekumbu Bay we conducted research on the main and biggest river in Jepara, the Wiso River. Awur Bay is slightly larger than Sekumbu Bay with 5.7 km coastline and it is located in the south of Sekumbu Bay. Two rivers and three channels enter Awur Bay, the biggest of which is the Kanal River where we implemented our study. Wiso and Kanal River originate in the same reservoir (Bapengan Dam), which is located 3 and 2.8 km upstream from the river mouth of Kanal and Wiso River, respectively (Fig. 1). Based on Central Java Water Resources Agency, average water discharge for Wiso River is 8.7 $m^3 s^{-1}$, while Kanal River is 5.4 $m^3 s^{-1}$ during the rainy season. The average depth of Wiso and Kanal River is 0.9 m and ~1.2 m, respectively. Coastal areas of Awur and Sekumbu Bay are mostly comprised of residential areas, agriculture fields, and pasture for livestock. Karimunjawa National Park, a Marine Protected Area that contains abundant and diverse wildlife, is located 25 km north of Jepara in the Java Sea.

Previous studies of Jepara's coastal seawater quality show nutrient concentrations exceeding the Indonesian standard limit for coastal seawater (Ayuningsih et al., 2014; Maslukah et al., 2014; Ministry of Environment and Forestry, 2004). In addition, substantial coral reef degradation was reported in the coastal area close to Sekumbu Bay due to a combination of sewage and sedimentation (Edinger et al., 1998). As the city's population is projected to grow by 1.5% per year until at least 2030, there has been extensive land use expansion to cater the economic and population development, e.g. new residential areas, new irrigation system for agricultural sector, and a brand new coal-based power plant (Jepara Regional Planning Agency, 2011). However, the economic infrastructure trend could not be followed by proper environmental and sanitation arrangements. As of 2017, there is no central wastewater treatment plant to cater the sewage loading, even though the city's long term plan suggests to build one by the end of 2030 (Jepara Regional Planning Agency, 2011).

2. Material and methods

2.1. Field measurements

The field survey was conducted from November–December 2016, during the rainy season in Indonesia. Salinity, temperature, pH-values, and dissolved oxygen (DO) were measured directly in the field using handheld probes: conductivity measuring cell (WTW™ TetraCon 925-P), pH (WTW™ Sentix 940), and DO (WTW™ FDO 925). Sensors were calibrated each day before the start of sampling using the manufacturer's protocol. DO results were corrected based on salinity and temperature as described in Weiss (1970).

We used radon (^{222}Rn) as tracer for coastal groundwater discharge. Usage of ^{222}Rn as groundwater tracer in surface water has been recognized due to its conservative behavior in nature and its much higher concentration in groundwater compared to surface water (Cable et al., 1996; Ellins et al., 1990). Radon-in-water measurement has been utilized extensively to quantify groundwater discharge to streams or coastal areas (Burnett and Dulaiova, 2003). ^{222}Rn grab samples were taken from groundwater ($n = 7$) to quantify groundwater end-member activity needed for discharge calculation in Eqs. (1) and (2). There were also ^{222}Rn grab samples collected from Wiso River ($n = 9$), Kanal River ($n = 7$), and Bapengan Dam ($n = 1$), in the same place with nutrient sampling points, to determine which part of the stream had the highest groundwater discharge. All grab samples were collected in 250 ml glass

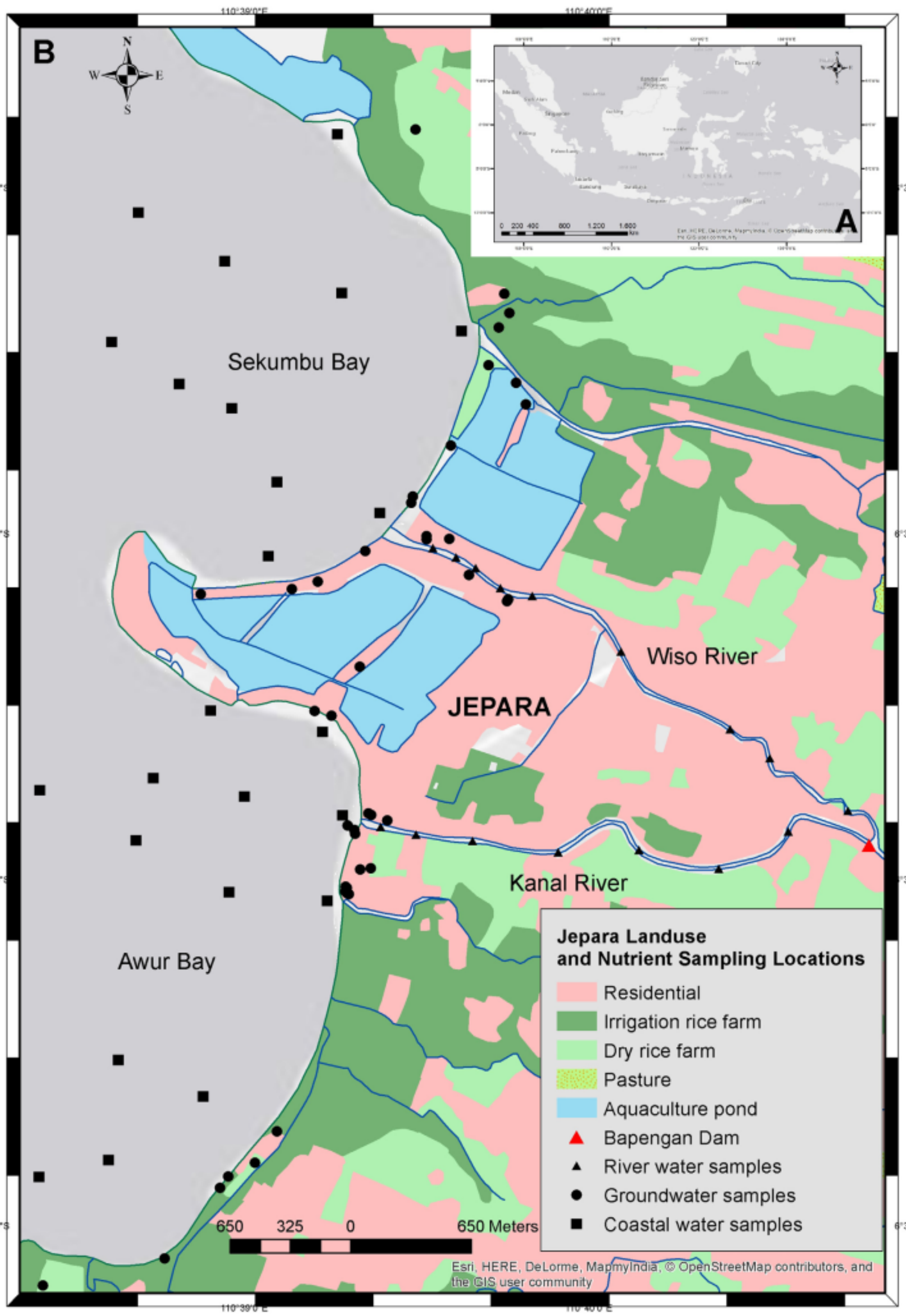

Fig. 1. Map of Indonesia (A), land use, and sampling points in Jepara (B). Landuse data was taken from Central Java Water Resources Management Agency (2016).

bottles by inserting a tube into the bottom of the bottle and filling it until it overflowed. The samples were analyzed within 6 h after sampling time using a RAD7 radon detector (Durridge Company Inc.) with the RAD H_2O accessory and WAT-250 protocol (Durridge, 2015). A 6 h decay correction was applied to the result based on RAD7 manual (Durridge, 2015).

To estimate the total amount of groundwater discharging into the bays, a stationary time series mooring was fixed in each of the river mouth over 24 h. ^{222}Rn was measured with 10 min counting interval using RAD7 with the Radon-in-Water accessory and Rad AQUA (Durridge, 2015). ^{222}Rn activities were later integrated into 30 min intervals. In parallel with the continuous ^{222}Rn measurements, salinity, water level, and temperature were measured every 30 min. A calibrated HOBO™ conductivity meter (model: U24–002) was deployed next to the RAD7 pump, while a HOBO™ water level logger (model: U20 L) was fixed to the river bed. ^{222}Rn in ambient air was measured at the end of each time series with the RAD7 detector. Wind speed was measured on site with an anemometer in order to correct for ^{222}Rn atmospheric evasion losses. In addition, a ^{222}Rn river spatial survey was implemented with shorter 5 min counting interval of the RAD7 detector. This high resolution river survey was conducted in the last 1.5 km downstream of each river because preliminary ^{222}Rn grab sample results suggested that ^{222}Rn concentrations downstream exceeded those upstream. ^{222}Rn concentrations from estuary and downstream spatial survey were used to calculate groundwater flux using equation stated in Eqs. (1) and (2) below.

A coastal ^{222}Rn spatial survey was also conducted in both Sekumbu and Awur Bay from a boat during low tide. It went from the southernmost part of Awur Bay and followed the coastal line until the northernmost part of Sekumbu Bay to identify major points of groundwater discharge signal.

Four water samples of 20 l each were collected from Wiso River and Kanal River to account for ^{226}Ra decay correction in the groundwater model in Section 2.1.2 below. ^{226}Ra was measured by Radium Delayed Coincidence Counter (Radecc) described in Waska et al. (2008).

Nutrient samples were taken from coastal groundwater (n = 44), river water (n = 17), estuarine water (n = 18), and coastal seawater (n = 24) of Awur and Sekumbu Bay. Coastal groundwater samples were taken from existing wells close to the sea or close to the rivers, i.e. 11 bore wells and 35 dug wells. Dug well water was pumped through plastic tubing by submersible pump and the samples were collected after letting the water from the well flow for a few minutes. Bore well water was obtained by 10–15 min of purging until standing water in the plumbing and piping were removed. River and seawater samples were collected from ~10 cm water depth. Nutrient samples were filtered through a Millipore cellulose acetate cartridge filter (0.45 μm pore size), collected into 40 ml polyethylene sample bottles, and stored on ice until analysis. NH_4 and PO_4 samples were measured directly in the field within 6 h, while NO_3 samples were poisoned by 50 μl saturated $HgCl_2$ solution and analyzed at the laboratory of ZMT in Bremen.

500 ml of estuarine and coastal seawater from Sekumbu Bay (n = 11) and Awur Bay (n = 12) were collected in dark polyethylene bottles to measure chlorophyll-*a* concentration. These samples were stored on ice in the dark until they were filtered with Whatman GF/C (47 mm diameter) filters within one day. To account for organic concentration, COD samples (n = 63 from all groundwater and river sampling points) were collected in 100 ml polyethylene bottles and acidified by sulfuric acid to pH < 2. COD sampling was only implemented in freshwater due to possible interference with chloride when using the reactor digestion method.

2.1.1. Sample analysis

Nutrient samples were analyzed using standard photometrical methods for NO_3 (Grasshoff et al., 2009), Hach salycilate method for NH_4, and Hach PhosVer3 ascorbic acid method for PO_4. Quality control was carried out using Hach check standards for each type of measurement before and after all measurements were implemented. Every mention of dissolved inorganic nitrogen (DIN) in this study refers to NO_3 and NH_4. Initially, NO_2 was also measured for all samples but the results were not included here due to negligible concentrations.

COD was measured using Hach USEPA reactor digestion methods based on Standard Methods 5220 D (Rice et al., 2012). Chlorophyll-*a* was quantified using Standard Methods 10200H (Rice et al., 2012). Symbol ± in numerical results refers to one standard deviation. Based on nutrient and chlorophyll-*a* concentration, eutrophication index (EI) in Jepara coastal area is determined using TRIX index, where scale of 4–6 means "moderate eutrophication" and scale >6 means "severe eutrophication" (Vollenweider et al., 1998).

The software suite R was used for all statistical analyses. Linear regression analysis was used to evaluate correlations between nutrients with physical parameters. Similarity between sample pools was analyzed by one-way ANOVA. If assumptions for parametric tests were violated, data was root or log transformed.

2.1.2. Groundwater discharge model

Groundwater discharge calculation in the river section below is based on model by Burnett et al. (2010) and Peterson et al. (2010). Eq. (1) shows the measured ^{222}Rn activity which is corrected for all possible sources and sinks such as ^{222}Rn ingrowth from parents ^{226}Ra, atmospheric evasion, and decay losses, and divided by the ^{222}Rn activity of the groundwater endmember, before it is multiplied by river flux to obtain the amount of groundwater discharge into the river. This equation refers to maximum range estimate of groundwater discharge, assuming that all groundwater input happens in the upstream part of the river. Q_{Gw} is groundwater flux (m^3 d^{-1}), C_{Rn} is ^{222}Rn concentration from river spatial survey and 24 h time series measurement (Bq m^{-3}), F_{atm} is atmospheric evasion (Bq m^{-2} d^{-1}), R is residence time (d), h is average depth of river (m), C_{Ra} is ^{226}Ra concentration (Bq m^{-3}), λ is decay constant of ^{222}Rn (0,181 d^{-1}), C_{Rne} is average ^{222}Rn concentration in shallow groundwater endmember (Bq m^{-3}), and Q_R is river flux (m^3 d^{-1}). F_{atm} or atmospheric loss is defined based on MacIntyre et al. (1995).

$$Q_{Gw}\left(m^3\ d^{-1}\right) = \left[\frac{\left[C_{Rn}\left(Bq\ m^{-3}\right) + \left[\frac{F_{atm}\left(Bq\ m^{-2}\ d^{-1}\right) \times R\,(d)}{h\,(m)}\right] - C_{Ra}\left(Bq\ m^{-3}\right)\right] e^{\lambda R}}{C_{Rne}\left(Bq\ m^{-3}\right)}\right] \times Q_R\left(m^3\ d^{-1}\right) \quad (1)$$

Eq. (2) below shows a minimum estimate of groundwater discharge assuming that groundwater input to the system occurs directly in the point of measurement or in the downstream part, hence there is no correction for atmospheric losses and radioactive decay.

$$Q_{Gw}\left(m^3\ d^{-1}\right) = \left[\frac{C_{Rn}\left(Bq\ m^{-3}\right) - C_{Ra}\left(Bq\ m^{-3}\right)}{C_{Rne}\left(Bq\ m^{-3}\right)}\right] \times Q_R\left(m^3\ d^{-1}\right) \quad (2)$$

C_{Rne} is obtained from average ^{222}Rn concentration in all shallow groundwater samples, as groundwater from the shallow aquifer most likely feeds the rivers while deep groundwater from >20 m depth is unlikely to discharge into the shallow rivers. All values were measured directly except for river flux and residence time. River flux used for this calculation was taken from Jepara Water Resources Agency and Jati (2014) data for rainy season (November–January), while residence time was calculated by dividing river volume (total upstream area multiplied by average depth based on field measurements) by river flux data. Total upstream area of Wiso and Kanal River were estimated by examination of satellite photos from online sources, i.e. Google Earth. Upstream area was also used to calculate nutrient flux per discharge area in Section 4.2.

2.1.3. Nutrient mass balance in groundwater and river water

Nutrient input from agriculture in Table 3 was estimated based on total agriculture area, cultivation index, and fertilizer usage from Katam (2017) for regencies surrounding Wiso River/Sekumbu Bay and Kanal River/Awur Bay, i.e. Batealit, Tahunan, Jepara, and Pakisaji. Potential nutrient excess from human sewage was calculated based on total population in the above regencies (Jepara Bureau of Statistics, 2016), percentage of households without access to sanitation system (Jepara Regional Planning Agency, 2010), and average of nitrogen (N) and phosphorus (P) content in human sewage (Henze et al., 2008). Manure waste contribution into groundwater calculation were derived from average N and P content in livestock waste (Ruddy et al., 2006) and total livestock population in the above regencies (Katam, 2017).

Nutrient flux mass balance in upstream, middle, and downstream in Fig. 6 was estimated by multiplying river discharge with nutrient concentration in river section, assuming constant river discharge from upstream to downstream. Nutrient transformation in the river was calculated without contribution of surface runoff, due to unavailable runoff data.

3. Results

3.1. Physical parameters

Physical parameter results for temperature, salinity, and DO in coastal groundwater, river water, estuarine water, and seawater are shown in Table 1. Temperature was similar in groundwater and surface water during the sampling period. Wiso and Kanal Rivers both had zero salinity for the most of their stream section except the last 1 km downstream. Most of the groundwater and river segments also appeared to be oxic (DO > 1 mg L^{-1}). There was no consistent trend between groundwater salinity compared to distance from coastline/riverbank. Between the two locations, data for each physical parameter are statistically similar ($p > 0.05$, one way ANOVA).

3.2. Nutrient and ^{222}Rn spatial distribution

Fig. 2 shows nutrient and ^{222}Rn distribution in study sites. NO_3 was the dominant form of nutrients in groundwater, even though the concentration is very variable even at a spatial scale of tens of meters (Fig. 2a). Groundwater NO_3, NH_4, and PO_4 concentrations were all statistically similar in both study sites ($p > 0.05$, one-way ANOVA), with concentration averages of 120–160 μM, 50–70 μM, and 8–19 μM for NO_3, NH_4, and PO_4, respectively (Table 1).

Mean nutrient concentrations between two studied rivers were also similar, except for NH_4 concentrations close to the Wiso River estuary (Table 1, Figs. 2b, and 3), which were unusually high. Fluvial NH_4 concentrations also demonstrated a linear correlation with organic matter as COD ($p < 0.05$), suggesting a common source, e.g. sewage or waste discharge.

Nutrient concentrations in coastal seawater were mostly lower than in groundwater or river water, except for two NH_4 values close to aquaculture ponds (Fig. 2b) and high PO_4 concentration in the coastal seawater of Awur Bay (Fig. 2c). Eutrophication index (EI) in the estuarine and coastal seawater, which was derived from nutrient and chlorophyll-*a* concentrations, indicated moderate eutrophication in Sekumbu Bay (EI between 4 and 6) and severe eutrophication in Awur Bay (EI > 6).

Nutrient data from this expedition are stored and accessible via https://doi.pangaea.de/10.1594/PANGAEA.884292.

^{222}Rn activity in shallow coastal groundwater in Sekumbu and Awur Bay area varied from 3000 to 16,000 Bq m^{-3} ($n = 7$). As seen in Table 1, Wiso River had higher fluvial ^{222}Rn activity than Kanal River, even though the opposite happened in the estuarine activity. Wiso River had ^{222}Rn concentration variation between 3000 and 5000 Bq m^{-3} in the downstream river section and 100–800 Bq m^{-3} in the estuary, while in Kanal River the range was 900–2500 Bq m^{-3} in the downstream river section and 100–1600 Bq m^{-3} in the estuary. Higher ^{222}Rn activities were measured approximately 1.5 km close to the estuaries in both rivers (Figs. 2d and 3) than those upstream, which coincided with higher NO_3 concentrations. NH_4 displays sharp increase closer to the estuary of Wiso River, but not in Kanal River, while PO_4 displays constant concentrations throughout both river sections.

Continuous ^{222}Rn sampling was conducted downstream (red dots in Fig. 2d) to find out whether there was a ^{222}Rn local hotspots in the rivers. However, there was no specific peak of ^{222}Rn activity observed during the spatial measurement and the detected ^{222}Rn range was distributed between 3000 and 5000 Bq m^{-3} for downstream Wiso River and 900–2500 Bq m^{-3} for downstream Kanal River without major spikes/outliers.

3.3. Estimating groundwater flux using radon concentration

Eqs. (1) and (2) were applied to the results of ^{222}Rn river spatial survey and time series measurements in Sekumbu Bay and Awur Bay to estimate groundwater discharge rates. Results between two equations exhibit small differences, i.e. approximately 0.3% difference between maximum and minimum discharge, so the number that listed in Table 2 is the average between maximum and minimum estimation. The small difference between maximum and minimum result suggests a limited contribution of atmospheric loss and radioactive decay to the discharge calculation. Both rivers have low water residence times due to high discharge (the fieldwork was done during the rainy season) and a short distance between the common source (a dam) and the estuaries (approximately 3 km long). Atmospheric losses are also considered to be low due to the calm weather (the recorded wind speed was 0–1.4 m s^{-1} in Wiso River and 0–2.7 m s^{-1} in Kanal River) and calm waves during our sampling. The ^{222}Rn end-member activity in the groundwater was 8600 Bq m^{-3}. Groundwater nutrient flux was determined by multiplying average groundwater nutrient concentrations with volumetric groundwater discharge rates.

Table 1
Concentration of physical and chemical parameters in individual compartments of the coastal hydrological continuum in Jepara.

Location	Parameter	Groundwater	River	Estuary	Coastal seawater
Sekumbu Bay (Wiso River)	Temperature (°C)	28 ± 1	29 ± 0.4	30 ± 0.7	31 ± 0.7
	Salinity (psu)	0.9 ± 1.5	0.3 ± 1	32 ± 0.4	32 ± 1.6
	Dissolved oxygen (mg L^{-1})	4.2 ± 1.3	4.8 ± 2.3	6.3 ± 0.6	7.4 ± 0.8
	NO_3 (μM)	167 ± 208	89 ± 20	1.7 ± 1.5	2.6 ± 2.9
	NH_4 (μM)	78 ± 119	30 ± 24	22 ± 9.1	5 ± 1.2
	PO_4 (μM)	8.8 ± 7.7	4.6 ± 3.6	2.9 ± 2.7	0.2 ± 0.2
	COD (mg L^{-1})	13 ± 9.9	8.6 ± 11	-	-
	Chl-*a* (μg L^{-1})	-	-	2.1 ± 0	1.2 ± 0.5
	^{222}Rn (Bq m^{-3})	5200 ± 2780	3870 ± 1220	373 ± 197	53 ± 25
	Eutrophication index	-	-	-	5.4
Awur Bay (Kanal River)	Temperature (°C)	28 ± 1	28 ± 0.7	27 ± 5	30 ± 0.6
	Salinity (psu)	0.7 ± 0.5	0.9 ± 1.6	14 ± 6	28 ± 7
	Dissolved oxygen (mg L^{-1})	3.6 ± 1.8	6.8 ± 1.1	6.4 ± 0.7	6.4 ± 0.9
	NO_3 (μM)	123 ± 147	99 ± 35	26 ± 13	12 ± 18
	NH_4 (μM)	58 ± 102	16 ± 7.7	26 ± 14	31 ± 21
	PO_4 (μM)	19 ± 13	4 ± 2.8	10 ± 12	15 ± 9
	COD (mg L^{-1})	6.1 ± 4.6	7.8 ± 12.7	-	-
	Chl-A (μg L^{-1})	-	-	1.7 ± 1.2	0.8 ± 0.5
	^{222}Rn (Bq m^{-3})	11,200 ± 3840	1680 ± 580	570 ± 496	43 ± 38
	Eutrophication index	-	-	-	7.7

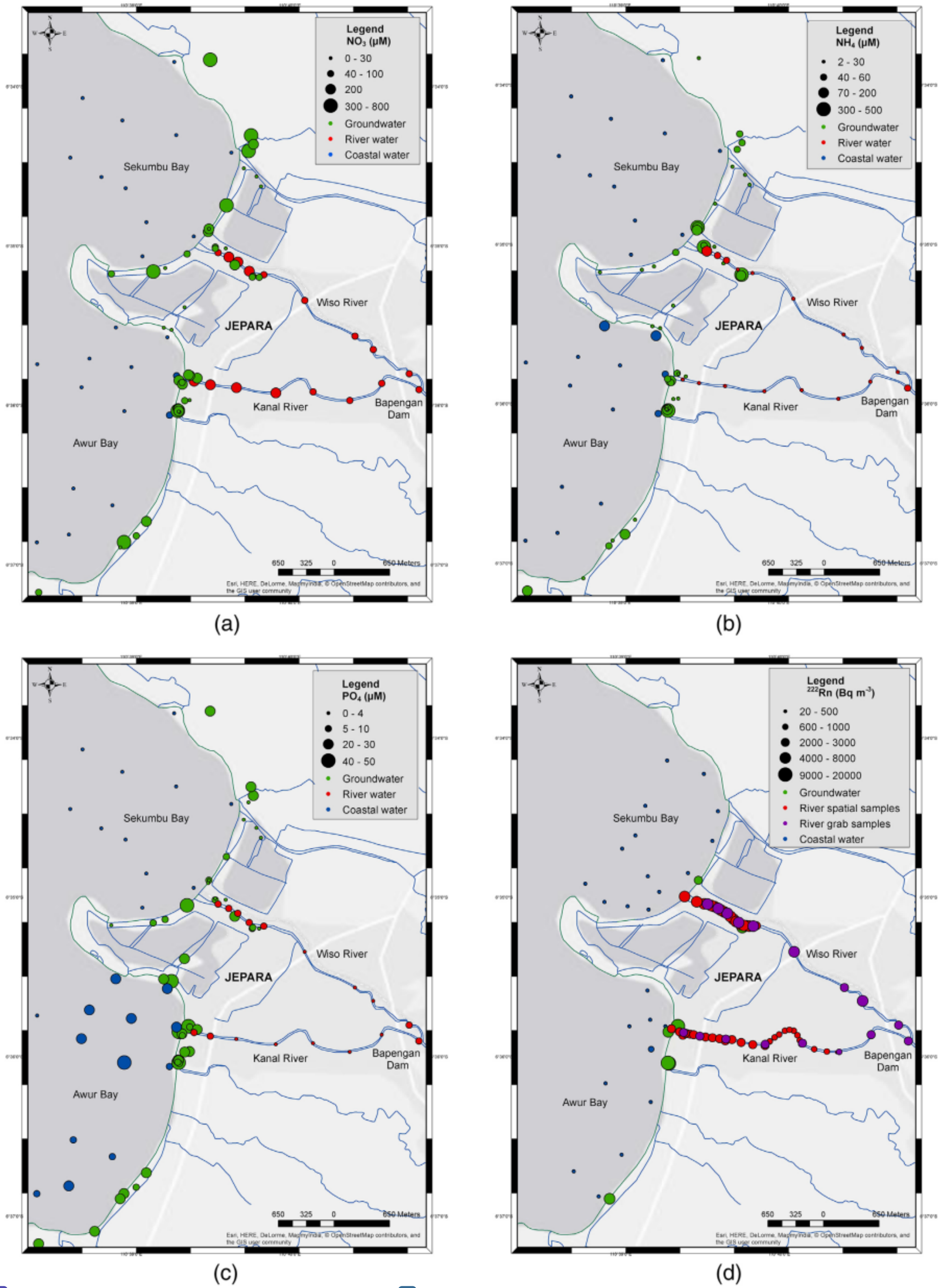

Fig. 2. Spatial distribution of (a) NO_3, (b) NH_4, (c) PO_4, and (d) ^{222}Rn in study sites. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

The estimation of groundwater flux in Table 2 represents groundwater discharging in the downstream part and estuary of each respective river. NO_3 constitutes most of the nutrients that discharged into rivers, followed by NH_4 and PO_4. Based on ^{222}Rn concentration range in downstream and estuaries stated in Section 3.2., averagely $315 \times 10^3\ m^3\ d^{-1}$ or 42% of downstream Wiso River flux was originated from groundwater, and the fraction of groundwater at the estuary mouth decreased to $32 \times 10^3\ m^3\ d^{-1}$ or 4% due to mixing with low ^{222}Rn seawater. In Kanal River, groundwater discharge was averagely $82 \times 10^3\ m^3\ d^{-1}$ or 18% of water in the river downstream and $32 \times 10^3\ m^3\ d^{-1}$ or 7% of estuarine water discharging into Awur Bay. In total, groundwater flux into two river systems in Jepara in rainy season amounted to $461 \times 10^3\ m^3\ d^{-1}$, and they were mostly discharged in low tide (Fig. 4).

Based on estuarine time series measurement (Fig. 4), ^{222}Rn activities were affected heavily by the tidal cycle, where the highest ^{222}Rn signal was detected at low tide. Water level dropped threefold and fourfold during low tide event in Wiso and Kanal River, respectively, and in the same time, ^{222}Rn signal increased three times in Wiso River and four times in Kanal River. At the Kanal River estuary mouth, salinity fell from 20 psu to 0 psu during peak low tide, while this parameter was consistently above 26 at the estuary mouth of Wiso River and decreased only slightly during low tide. During this period, total fresh groundwater discharge into the bay was calculated as high as $120 \times 10^3\ m^3\ d^{-1}$ from both estuaries, while during high tide, groundwater flux in each estuary amounted only to $15–20 \times 10^3\ m^3\ d^{-1}$ from total river discharge.

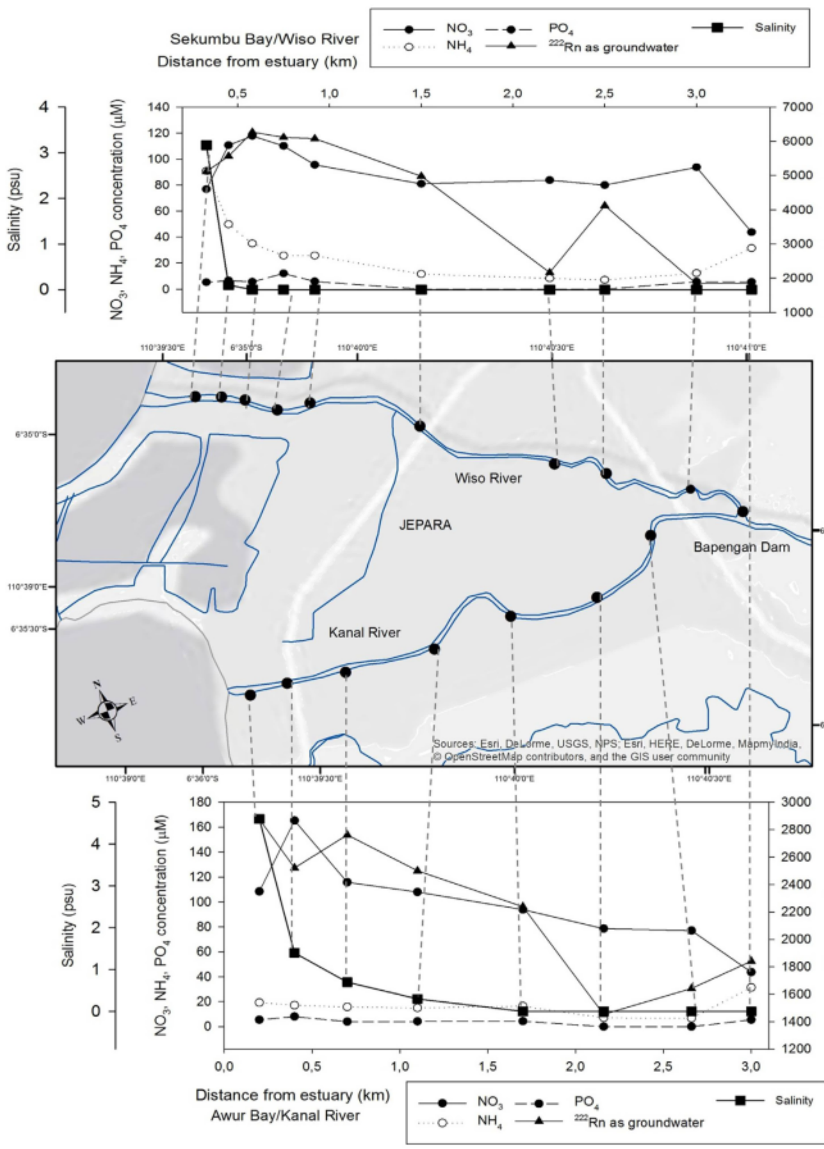

Fig. 3. Center figure shows map of both rivers which were studied in Jepara, while the upper and bottom graphs show of the concentrations nutrients and ^{222}Rn (grab samples) at their respective locations in the river.

4. Discussion

4.1. Nutrients in groundwater

Nutrients were measured in total 44 groundwater samples in Sekumbu and Awur Bay. NO_3 was the majority of nutrient species in groundwater samples with an average of 50% of total DIN measured, followed by NH_4 with 40% and PO_4 with 10%. This is in accordance with several studies that suggest NO_3 as dominant form of N in the aquifer of urban regions compared to reduced N, such as NH_4 and dissolved organic nitrogen (DON) due to its mobility in soil (Burkart and Stoner, 2002; Cole et al., 2006; Nolan and Stoner, 2000). NH_4 and dissolved organic matter are most likely to be attenuated during aquifer transport due to nitrification, mineralization, and sorption (Buss et al., 2004; Slomp and Van Cappellen, 2004), and DON is also generally considered to be originated from natural source rather than anthropogenic sources (Perakis and Hedin, 2002). PO4 is usually the most abundant form of P in the aquatic system compared to dissolved organic phosphorus (DOP) (Correll, 1998). Thus, although we did not measure DON or DOP fractions, we expect their contribution to the total N and P concentrations to be small.

DIN and PO_4 concentration in both Awur and Sekumbu Bay groundwater were substantially higher than in lower populated tropical areas e.g. Hawaii and Mauritius (Knee et al., 2008; Povinec et al., 2012), and even higher than in populated areas such as Bangkok or Manila (Burnett et al., 2007; Taniguchi et al., 2008). Our average PO_4 and DIN groundwater concentration in Jepara were higher and lower compared to Jakarta (Delinom et al., 2009), respectively, the latter has a population of 10 million inhabitants, with an eight times higher population density than Jepara.

High nutrient concentrations in Jepara can be related to a combination of natural geological conditions, anthropogenic activity, and land use. Human activities surrounding the study sites can be categorized into residential area, agriculture, and livestock, as these three activities account for 52%, 30%, and 4% of land use, respectively. They represent the dominant anthropological sources compared to others such as aquaculture (1%, Central Java Water Resources Management Agency (2016)).

According to Katam (2015), fertilization application in Jepara's farming land is on average 170 kg ha^{-1} year^{-1} for N based fertilizer and 13 kg ha^{-1} year^{-1} for P based fertilizer, which is similar to fertilization rates from China of 160 kg N ha^{-1} year^{-1} (Kaiser et al., 2013), and well above the global average of 90 kg N ha^{-1} year^{-1} (Jennerjahn, 2012). Considering that uptake efficiency of fertilizer amounts to 40% on average in tropical regions for both N and P based fertilizer (Han et al., 2010), total nutrient excess from fertilizer in Sekumbu Bay and Awur Bay area into the groundwater would accumulate to 510 kg N ha^{-1} year^{-1} and 48 kg P ha^{-1} year^{-1} or equivalent with 670 ton N year^{-1} and 63 ton P year^{-1} (Table 3), even though the environmental impact in receiving surface water would be delayed due to residence time of nitrogen in the aquifer (Ascott et al., 2017). High P loading from fertilizer can overwhelm soil sorption capacity for P, which increases PO_4 mobility in groundwater (Slomp and Van Cappellen, 2004). Together with fertilization, volcanic origin of soil and sediment in the Jepara area could affect high PO_4 mobility in our study site as P is less likely to bond with volcanic soils and sediments (Correll, 1998).

Furthermore, co-occurrence of high NO_3 and NH_4 concentrations in groundwater is not unusual in locations with wastewater contribution from individual septic systems (Kroeger and Charette, 2008). Based on Jepara Regional Planning Agency (2010), only an average 25% of houses in Sekumbu and Awur Bay area are connected to a sanitation system, which means the rest of the households potentially supply 1120 ton N year^{-1} and 190 ton P year^{-1} of untreated wastewater into the groundwater. Significant pollutant input from non-point sources into the aquifer could also come from livestock farming in some area (Ma et al., 2011). In Jepara, manure waste contributes 950 ton N year^{-1} and 300 ton P year^{-1} of potential diffuse pollutant into the soil and aquifer system.

Overall nutrient contribution to groundwater from agriculture, livestock, and sewage due to land use around Wiso River and Kanal River are shown in Table 3. The difference between calculated input from land surface and measured export of groundwater flux to the river equals 2200 ton N year^{-1} and 380 ton P year^{-1} nutrient loss in the soil and aquifer which are potentially attributable to surface runoff and biogeochemical processes in the nutrient path such as denitrification, mineralization, or sorption in the aquifer (Bowen et al., 2007).

Despite groundwater velocity and residence time can reduce nutrient concentrations before they discharge into river system (Michael, 2005), a considerable portion of groundwater nutrients still enters the stream water, partially due to lack of significant retention alongside the riverbank, which is full of congested residential areas without a riparian vegetation zone. To limit the uncertainty of the quantity of nutrient loading from aquifer into the river, groundwater sampling points were chosen from wells that located no further than 100 m from the coastline and 50 m from the river bank.

4.2. Groundwater discharge and transport in stream water

In order to estimate groundwater discharge into the river and estuary, ^{222}Rn concentrations were measured in groundwater, river water, and coastal seawater. The range of ^{222}Rn concentrations of coastal groundwater endmember in this area (3000–16,000 Bq m^{-3}) is comparable to other reported ^{222}Rn activities from areas with a similar geological setting in Indonesia (Bakti et al., 2014) and elsewhere (D'Alessandro and Vita, 2003; Hwang et al., 2005; Moreno et al., 2014; Wu et al., 2013). ^{222}Rn activities in coastal groundwater of Awur Bay were two times higher than ^{222}Rn activities in Sekumbu Bay (Table 1). Swarzenski et al. (2007) implied that ^{222}Rn variability in the same area can be counted as function of local geology and residence times. Based on geological map by Suwarti (1992), the whole coastal plain of Jepara is situated in the same geological foundation, thus it is more likely that the difference in ^{222}Rn in coastal groundwater is derived from the difference in local groundwater residence times.

Vertical salinity and DO profiles in the downstream section of the river show low salinity in the bottom part, which could indicate that groundwater discharges from the river bottom (Fig. 5). Impermeable retaining walls which both channels have at the riverbanks could be the explanation for that observation. Stream dredging, implemented by the local government, inhibits the development of impermeable stream bottom sediments and thus improves groundwater flow (as reported elsewhere by Santos et al. (2008)). This can also explain higher groundwater discharge rates in Wiso River compared to Kanal River

Table 2
Estimation of groundwater discharge in Sekumbu and Awur Bay based on average ^{222}Rn from shallow coastal groundwater samples.

Parameters		Wiso River/Sekumbu Bay	Kanal River/Awur Bay
Upstream area (10^3 m^2)		89	78
Average depth (m)		0.9	1.2
Residence time (d)		0.13	0.19
Background ^{226}Ra (Bq m^{-3})		8	13.5
Atmospheric loss (Bq m^{-2} d^{-1})		550	635
River discharge (10^3 m^3 d^{-1})		750	470
Average volumetric groundwater discharge rate in downstream and estuary (10^3 m^3 d^{-1})		347	114
Average of groundwater fraction in river discharge/Q_{Gw}:Q_R (%)		42%	18%
Groundwater nutrient fluxes (10^3 mol d^{-1})	NO_3	58	14
	NH_4	27	7
	PO_4	3	2

(Table 2), where Wiso River is wider than Kanal River, thus it has more bottom surface to discharge the groundwater.

In both of the rivers, ^{222}Rn numbers fluctuated in the upstream part (Fig. 3). Three circumstances could cause this ^{222}Rn fluctuation: geology, hydrodynamic condition, and river construction (e.g. retaining walls or stream dredging). As mentioned earlier, both rivers are located in the same geological foundation with no known fracture zones which rules out a variation in geology. Secondly, based on our field observation, the upstream part of both rivers were well mixed due to shallow water depth, thus, ^{222}Rn fluctuation due to saline water effect from the estuary is unlikely. However, we also observed that the retaining wall of the river was cracked in some places, and the stream dredging was not equally implemented in all parts of the river in the same time. We expect this to be the main reason for the varying ^{222}Rn signals in the upstream area.

As seen in Fig. 4, the peak ^{222}Rn signal rates were observed during low tide in both estuaries, as also observed elsewhere (Makings et al., 2014; Peterson et al., 2010; Swarzenski et al., 2007). This would suggest that either groundwater was released as a result of water level fluctuation, or that high ^{222}Rn river water was mixed with low ^{222}Rn seawater during high tide (Swarzenski et al., 2007). During our measurement, surface water level dropped 3–4 times during low tide event, and ^{222}Rn activities were increased 3–4 times in the same period. The same level of co-variance between water level and ^{222}Rn concentration suggests that tidal variation effect seems to be smaller than expected at our sites.

Daily variation of groundwater flux in this calculation can be considered as minimum, as ^{222}Rn correction for the formula does not display big gap between minimum and maximum flux in the 24-hour measurement. However, groundwater discharge rates may vary seasonally due to changes in precipitation and evaporation, especially considering groundwater discharge in this location originates from a shallow aquifer. In the dry season, we expect a higher percentage of groundwater

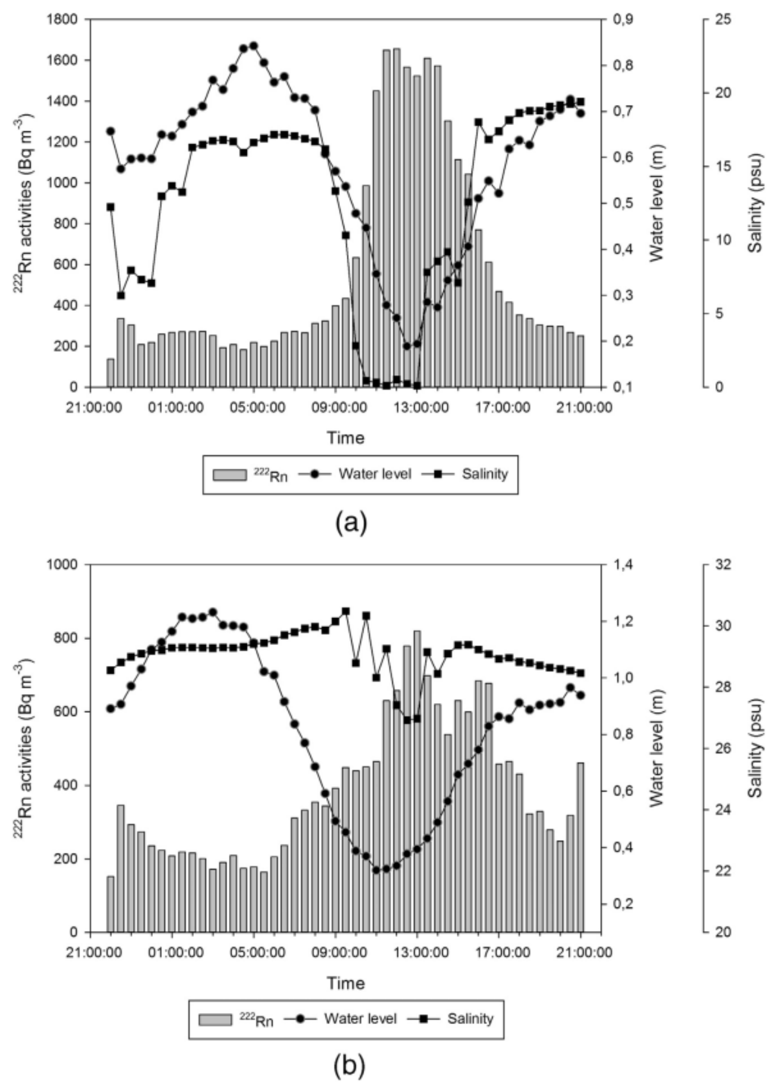

Fig. 4. Plot of ^{222}Rn activities, water level, and salinity during estuarine time series measurement in (a) Kanal River/Awur Bay (sampling date 13 November 2016) and (b) Wiso River/Sekumbu Bay (sampling date 22 November 2016).

Table 3
Groundwater nutrient inputs and outputs in Jepara.

Nutrient pollution source	Terrestrial nutrient load to groundwater (ton year^{-1})[a,b,c,d]		Percentage of terrestrial contribution to groundwater (%)		Groundwater export to river (ton year^{-1})[e]		Nutrient loss in soil and aquifer (ton year^{-1})	
	N	P	N	P	N	P	N	P
Fertilizer	670	63	24	10	540	180	2200	380
Sewage	1120	190	41	35				
Manure	950	300	35	55				

[a] Jepara Bureau of Statistics (2016).
[b] Katam (2017).
[c] Henze et al. (2008).
[d] Ruddy et al. (2006).
[e] From this study.

fractions in river water discharge, due to a less pronounced seasonality in groundwater discharge than river discharge.

The groundwater discharge from Wiso and Kanal River is comparable with other estuary studies in other regions, albeit in the higher end of discharge quantity. It also must be noted that the rivers studied in the comparison studies are significantly longer than the two in this study (Table 4). Thus, the environmental implication of groundwater discharge into Wiso and Kanal River per total area is large, especially as the groundwater brings high concentrations of terrestrial nutrients as described in the previous section. Our total groundwater nutrient flux estimation from both sites is 106 × 10^3 mol d^{-1} or equivalent with 0.63 mol m^{-2} d^{-1} for DIN and 5 × 10^3 mol d^{-1} or approximately 0.03 mol m^{-2} d^{-1} for PO_4, which are considerably higher than other estuarine nutrient studies as the such Loxahatchee River estuary (Swarzenski et al., 2006), the Korogoro Creek estuary (Sadat-Noori et al., 2016), or industrialized Masan Bay in South Korea (Lee et al., 2009). This indicates that a combination of population, anthropogenic activity, and environmental infrastructure contributes to nutrient flux into Jepara's estuaries and coastal area.

4.3. Nutrient dynamics in stream water and estuary

Combined, stream NO_3 and NH_4 concentrations in Jepara sites were higher than less urbanized tropical rivers e.g. Amazon (Cai et al., 2004), comparable with Chao Phraya river in Bangkok (Burnett et al., 2007) or other Indonesian rivers which have similar population density (Jennerjahn et al., 2004), but still lower than higher populated Chinese rivers (Liu et al., 2009). NO_3 species made up the dominant percentage of nutrient in both rivers by average 78–80%, similar to the global average of 77% (Turner et al., 2003). This indicates medium anthropogenic modification scale into coastal system in Jepara, as areas with higher level of human modification usually has higher NH_4 percentage (~50%) in the river discharging into ocean (Liu et al., 2009). Average PO_4 concentration in both rivers are considered at average level compared to global data Smith et al. (2003).

Fig. 6 shows a calculated and measured budget for NO_3, NH_4, and PO_4 flux in Wiso River and Kanal River during the wet season 2016. Wiso River gained additional nutrients, while Kanal River obtained only NH_4 but lost NO_3 and PO_4·These transformations can be attributed to surface runoff or various biogeochemical process transformations and nutrient cycling, such as organic remineralization, possible due to oxic conditions or availability of organic constituents as COD (Table 1).

Also, in particular Wiso River gained a higher amount of NO_3 and NH_4 loading than Kanal River (Fig. 6). Land use along Wiso and Kanal River are typically similar except in the downstream part of Wiso River, where there are two aquaculture ponds (Fig. 1). Since aquaculture waste can contain high levels of nitrogen and COD (Cao et al., 2007), it could be the main source of NO_3 and NH_4 in the downstream part of Wiso River, and in areas close to the ponds in coastal seawater of Awur Bay (Fig. 2B). Previous studies in other coastal regions of Jepara found that the estuary tends to serve as nutrient sink for waste discharged from coastal fishpond cultures (Pranowo et al., 2004).

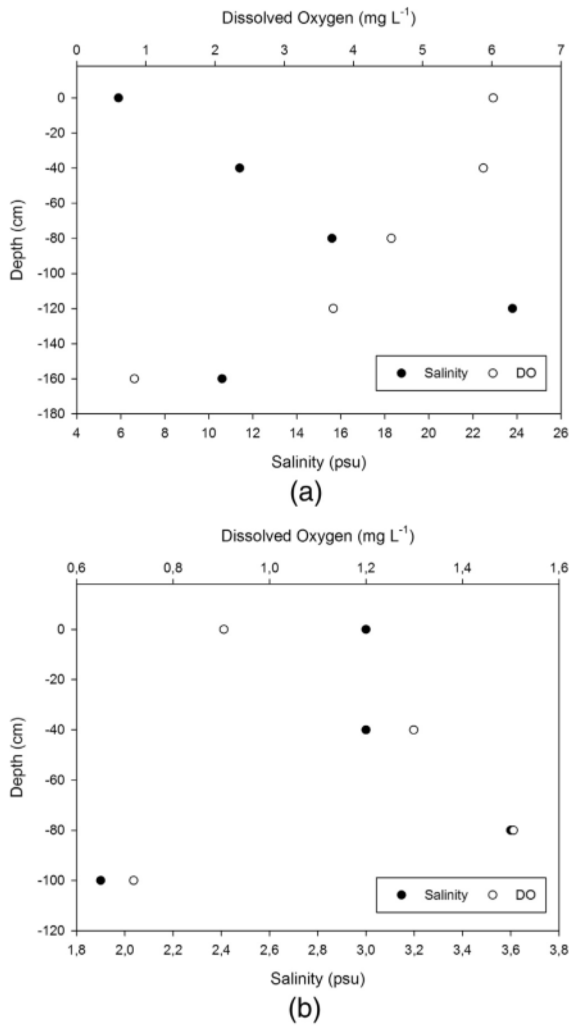

Fig. 5. Vertical profile of DO and salinity in the (a) Kanal River/Awur Bay and (b) Wiso River/Sekumbu Bay estuaries.

Table 4
Comparison of groundwater discharge into different estuaries.

Study	Location	Total length of river studied (km)	Environmental condition	Groundwater discharge ($\times 10^3$ m^3 d^{-1})
Peterson et al. (2010)	South Prong River, US	9	Subtropical estuary	3.3–480
Burnett et al. (2010)	C-25 Canal, US	8.5	Subtropical estuary	312–331
Tamborski et al. (2017)	Forge River, US	14.5	Subtropical estuary	12.2
Sadat-Noori et al. (2016)	Korogoro Creek, Australia	5	Subtropical estuary	86.4
This study	Wiso and Kanal river, Indonesia	5.5	Tropical estuary	461

4.4. Environmental implications

Wiso and Kanal River estuaries discharge approximately 44×10^3 mol d^{-1} of DIN and 7×10^3 mol d^{-1} of DIP to Jepara coastal area (Fig. 6). Their effect can be seen on the eutrophication index that shows moderate and severe eutrophication in Sekumbu Bay and Awur Bay, respectively (Table 1). Compared to previous studies implemented in Sekumbu Bay (Ayuningsih et al., 2014; Maslukah et al., 2014), our study shows higher primary productivity in the estuary and coastal area as represented by one order of magnitude increase of chlorophyll-*a* and other nutrient concentration within three years, attributable to population growth and expanding human activities. Coastal samples in Sekumbu Bay show nutrient concentrations similar to Manila Bay (Jacinto et al., 1998). However, NH_4 and PO_4 concentrations in Awur Bay were one magnitude higher, which may indicate persistent local inputs, such as or polluted surface runoff or marine-based sources, e.g. primary production (Kaiser et al., 2013).

From a stoichiometric perspective, DIN:DIP ratios in receiving coastal seawater shows a deviation from the Redfield ratio of 16:1 (Redfield, 1958). Sekumbu Bay coastal seawater is P limited (DIN:DIP of 51), while Awur Bay leans towards N-limitation (DIN:DIP of 3). Deviation from the Redfield ratio could change ecosystem structure such as shifting plankton community and increasing biomass, phytoplankton, and macro algae production, which further affecting light penetration and dissolved oxygen to coastal biota (Billen and Garnier, 2007; Slomp and Van Cappellen, 2004).

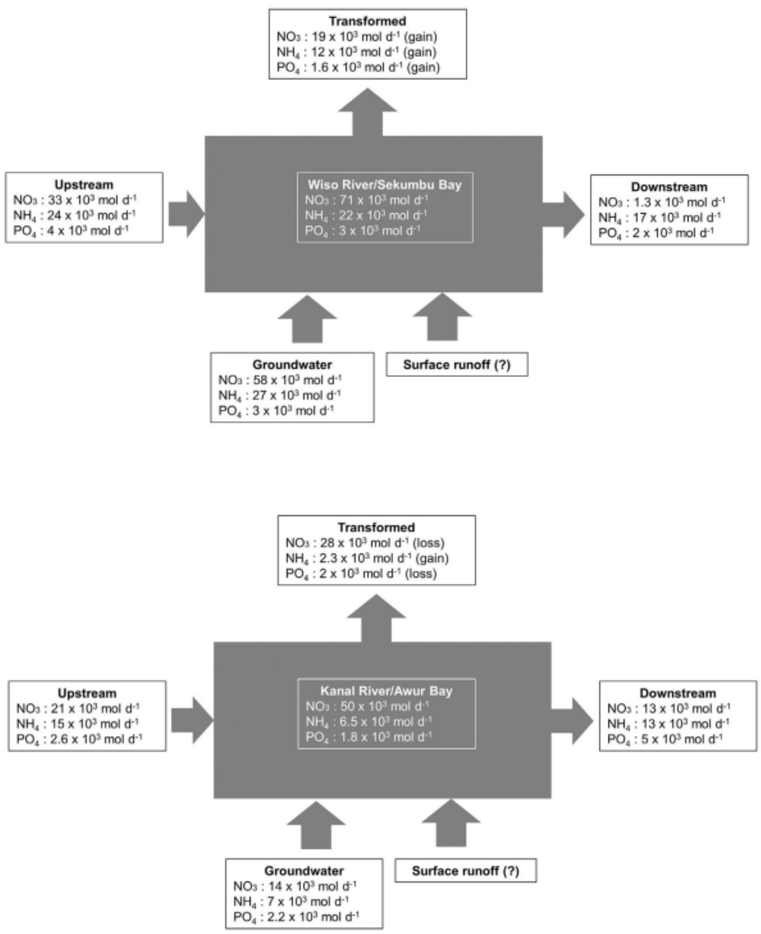

Fig. 6. Measured and calculated nutrient flux budget in Wiso River (above) and Kanal River (bottom).

Considering sewage leaking contributes as the highest point source contribution compared to other anthropological activities (Table 3), a wastewater treatment plant could be beneficial to reduce nutrient pollution in the future (Kroeger and Charette, 2008; Mitsch et al., 1999). A proper sanitation system is important since Jepara has 1.5% annual population growth (Jepara Bureau of Statistics, 2016) which means Wiso and Kanal River will receive additional 190 ton N $year^{-1}$ and 32 ton P $year^{-1}$ by 2030 assuming there is no improvement in sanitation system, plus the additional nutrient transport as a result of long-decade intensive agriculture that is delayed due to groundwater residence time (Ascott et al., 2017; Michael et al., 2005). A phosphate detergent ban could also be introduced to specifically limit P input to the groundwater system (Litke, 1999).

Diffuse or non-point source pollutions such as from fertilizer or manure, which in total contributes to 59% of N and 65% of P via groundwater discharge in the study site, could be controlled by modification of the farming methods or nutrient retainer along the riverbank and coastline e.g. wetland or riparian systems (Mitsch et al., 1999), particularly in the area with detected high radon activity. This could be advantageous considering local government plans to open 5160 ha of new agricultural field by 2030 in the areas surrounding Wiso River and Kanal River (Jepara Regional Planning Agency, 2011), yielding additional 80 kg N ha^{-1} $year^{-1}$ and 48 kg P ha^{-1} $year^{-1}$ into land surface adjacent to Wiso River and Kanal River. Studies regarding wetland that specifically constructed to prevent ground and surface water quality deterioration shows N and P removal range of 40–50% depending on the type of wetland and inflow loading (Vymazal, 2007; Yeh and Wu, 2009).

As groundwater fluxes comprise a significant fraction of the surface discharge flux into the ocean, monitoring programs for groundwater system could include construction of monitoring wells inshore and offshore and development of continuous environmental assessment program, which is still absent from the local environmental regulation. For the assessment program to be successful, a detailed description of local hydrostratigraphy, groundwater chemistry and hydraulic head, and validated groundwater model have to be developed (McCoy and Corbett, 2009).

5. Conclusions

Groundwater and nutrient discharge into streams and estuaries was measured in Jepara, Indonesia. This study confirms a significant groundwater-derived nutrient discharge to Jepara coastal area via stream water. Our results show that the main sources of these nutrients are sewage, manure, and fertilizer. Considering the size of the upstream discharge area, Jepara's estuaries receive higher nutrient loading per total surface area compared to other major cities due to an improper sanitation system and lack of barriers for diffuse source pollution such as fertilizer and livestock waste.

Increasing trends of nutrient concentration and primary productivity level in a short time span of years in Jepara's receiving coastal seawater also suggests its sensitivity towards change in population and human activities in its adjacent watershed. This could endanger the environmental state of Jepara's coastal aquatic area since the city is projected to have 1.5% population growth by 2030. Considering that nutrient input from coastal groundwater discharge contributes up to 106×10^3 mol d^{-1} of DIN and 5×10^3 mol d^{-1} of DIP, environmental management related to water quality in aquifer should be considered in order to prevent further water quality deterioration in the coastal area.

In general, global nutrient budgets often include megacities in developing regions in the assessment of anthropogenic effect to coastal areas. However, based on the observations from this study, direct nutrient fluxes to the sea from the abundant medium population coastal cities with highly modified regional land use and little developed central sewage systems may contribute significantly to the total nutrient discharge into the ocean. Thus, it should not be overlooked in the large scale nutrient budget calculations to obtain a more accurate assessment of the effect of human activities in the coastal region.

Acknowledgements

The authors wish to thank the student assistants during field expedition, Florian Senger, Ardha Yosef, Filius Justitia, and Rengga Wahyu; Diponegoro University lab technician Andreas Nur Hidayat; as well as Dr. Subagiyo and Dr. Rudhi Pribadi for the logistic field assistance. We also thank the Dean of Faculty of Fisheries and Marine Science Diponegoro University, Prof. Agus Sabdono for his support. Three anonymous reviewers are acknowledged for their constructive comments on this manuscript. This research was financially supported by the German Academic Exchange Service (DAAD; Sustainable Water Management Grant No 57156376 to Dini Adyasari) and the German Federal Ministry for Education and Science (BMBF Grant No #01LN1307A to Nils Moosdorf).

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