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ESTIMATION OF AMMONIUM SOURCES IN INDONESIAN COASTAL ALLUVIAL GROUNDWATER USING Cl^- AND GIS

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ABSTRACT: Indonesian coastal alluvial plain has a large groundwater storage capacity. However, human perturbation to the groundwater environment has resulted in groundwater contamination. Nitrogen species are parameter those usually detected in Indonesian coastal groundwater. The main objective of this study was to evaluate the possible sources of ammonium (NH_4^+-N), as one form of nitrogen species, using both chemical properties and geographic information. The study site was Indramayu, a developing rural area located in the northern part of West Java. In August 2017, 20 groundwater and river water samples were collected from sites representative of various types of land-use. The chloride (Cl^-) concentration in the samples ranged from 15 mg/L to 12,000 mg/L. Most of the locations represented fresh-brackish to brackish-salt water conditions, indicating the influence of seawater on the groundwater samples. Furthermore, analysis of several nitrogen species identified high NH_4^+-N concentrations in the samples. Based on the relationship between Cl^- and NH_4^+-N, samples were clustered into three groups. The first and second groups exhibited linear correlations, while in the third group, no relationship between these two parameters was identified. The land-use associated with each group was then evaluated. The main NH_4^+-N source in the first group was identified as organic sediment, where NH_4^+-N may be released through ion exchange with sodium, facilitated by the high salinity conditions. The source of NH_4^+-N in the second group was suggested to be a combination of sediment, fertilizer, and human and animal waste. Finally, it was hypothesized that the source of NH_4^+ in the third group is mainly human waste.

Keywords: Indramayu, Groundwater, Chloride, Ammonium, Land-use

1. INTRODUCTION

Groundwater is the largest unfrozen freshwater resource on earth, with a total volume of approximately 11 million km^3 [1]. This highly valuable resource could be used as drinking water for at least 50% of the world's population [2]. Despite this, unsustainable groundwater abstraction has led to a global groundwater depletion problem [1], [3]–[4]. Groundwater depletion can have knock-on effects, which are usually complex and dependent on the condition of the aquifer [1]. The direct effect, a decrease in the water table, can lead to serious issues, such as land subsidence and groundwater contamination. Such problems are already occurring in Indonesia. According to UNESCO [3], Indonesia is one of the top ten groundwater abstracting countries, with an abstraction rate of 14 km^3/year; this is predicted to increase by 1–2% annually [3].

Recently, these groundwater depletion effects have been increasingly observed in Indonesian coastal alluvial plains. An aquifer in alluvial plain has a large groundwater storage capacity, and thus forms the main source of clean water for many developing cities. Thus, urbanization is one of the major triggers of groundwater contamination in these environments [5]-[6]. Jakarta and Semarang are examples of Indonesian coastal alluvial plain cities facing groundwater perturbation and nitrogen species contamination because of urbanization. For example, in Jakarta, extensive groundwater depletion has caused land-subsidence [7]-[8], shallow and deep groundwater mixing, seawater intrusion, and nitrogen contamination, particularly nitrate, [5], [9]–[10]. Similarly, groundwater abstraction in Semarang has also resulted in land-subsidence [11], seawater intrusion, and nitrate contamination [12].

In this study, an investigation of groundwater in a coastal alluvial plain located in Indramayu, Indonesia, which can be found in the northern part of West Java (Fig. 1), was conducted. Indramayu is currently a rural area but has the potential to become urbanized in the future. The aquifer has a large unconfined groundwater potential, around 65,213.8 m^3/day [13]. However, the research concerning quality is still limited. Unlike the big cities of Jakarta and Semarang, human environmental impact is not significant in Indramayu. This relatively pristine condition is ideal for studying the nature of the groundwater environment during the early stages of urbanization.

In this study, the nitrogen concentration in the groundwater, particularly ammonium (NH_4^+-N), was studied. This type of nitrogen species has been little studied in Indonesia. While NH_4^+-N is a good indicator to recognize groundwater contamination from human waste, fertilizer uses, and contaminated land [14]. NH_4^+-N source apportionment is attempted by evaluating the relationship between NH_4^+-N and chloride (Cl^-) concentrations, in addition to an assessment of land-use.

2. METHODS

2.1 Study area

The study site was located in the eastern part of Indramayu, on the North Coast of Java (Fig. 1). The total area of Indramayu is approximately 2,100 km^2, with a coastline length of about 147 km [15]. The topography of the area is low land to shore, with an average slope of 0-2%. The area has a tropical climate, with an average annual rainfall of 2,146 mm/year.

2.1.1 Geology

The Indramayu coast is a river delta, formed by sedimentation processes fed by the Cimanuk River [16]. Generally, Indramayu soil was formed from an alluvial plain environment, but also includes deposits from beach ridge, coastal, deltaic, and floodplain environments, along with the erosion of tuffaceous sandstone and conglomerate (Fig. 1).

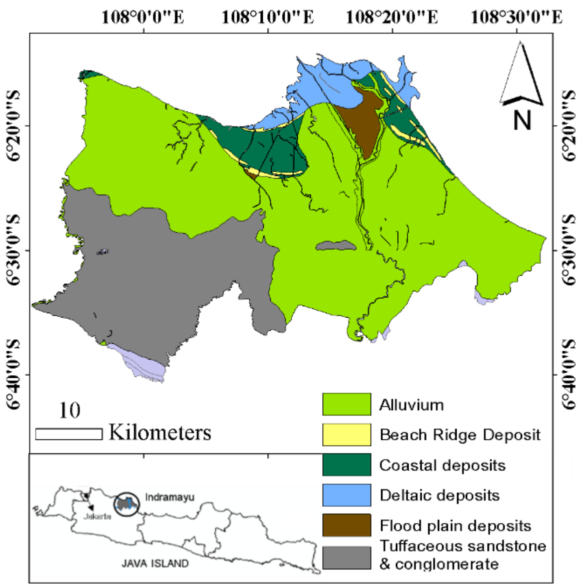

Fig. 1 Geology of Indramayu.

The alluvial plain is the dominant geologic formation in the area, with a groundwater water level of approximately 0.4 m to 3.2 m [17] and unconfined groundwater reserves of approximately 370 L/s [18]. The shallow groundwater table along with high permeability of soil make alluvial plain groundwater usually vulnerable to contamination.

2.1.2 Land-uses

Land-use in the Indramayu area is depicted in Fig. 2. The dominant land-use types are paddy fields and brackish fishponds. Settlements are concentrated on the delta formation, next to Cimanuk River. Oil and gas operations are also present, as this is the main industry located in the northeastern region of Java. This industry is owned by the Indonesian government.

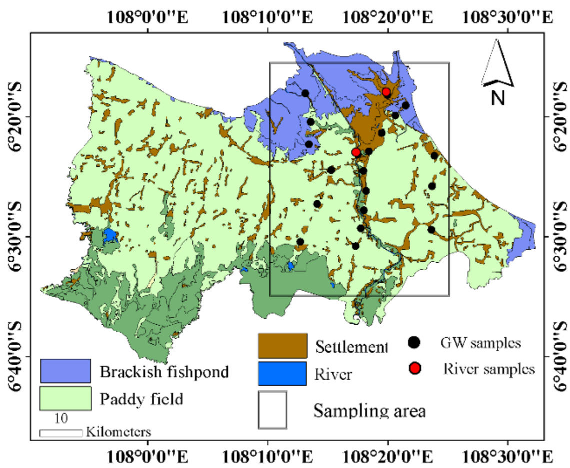

Fig. 2 Land-use types of Indramayu and sampling locations.

2.2 Sample collection

A total of 20 samples were collected from unconfined groundwater aquifers (18 samples) and the Cimanuk River (2 samples), in August 2017, during the dry season (Fig. 2). Sampling locations were selected to represent the dominant land-use types in Indramayu area, namely Paddy field, brackish fishpond, and settlement. The field campaign took place during the dry season with rainfall amounts between 20 – 50 mm.

2.3 Parameter and chemical analysis

This study focuses on the analysis of nitrogen species, including nitrate (NO_3^--N), nitrite (NO_2^--N), and ammonium (NH_4^+-N), in addition to chloride (Cl^-). Initially, samples were filtered through a 0.2 µm polytetrafluoroethylene to remove materials (i.e. bacteria) those can affect the concentration of nitrogen species during storage time. Then, the samples were stored in a freeze condition until analysis.

The concentrations of nitrogen species were determined with a continuous-flow automated nutrient analyzer and Cl^- was quantified with ion chromatography. The investigation of ammonium sources was conducted to analyzing the relationship between NH_4^+-N and Cl^-. This relationship was then confirmed by land-use evaluation.

3. RESULTS AND DISCUSSION

3.1 Chloride and nitrogen species

3.1.1 Chloride concentration

The groundwater Cl^- concentrations varied from 15 mg/L to 12,000 mg/L. Using the Stuyfzand classification [19], the groundwater samples can be classified as fresh, fresh-brackish, brackish, brackish-salt, and salt water based on Cl^- concentration (Fig. 3). Freshwater was mainly identified in the settlement areas, while other classifications were identified in paddy fields and brackish fishpond areas. Samples with high Cl^- (> 150 mg/L) were dominant over low Cl^- (<150 mg/L).

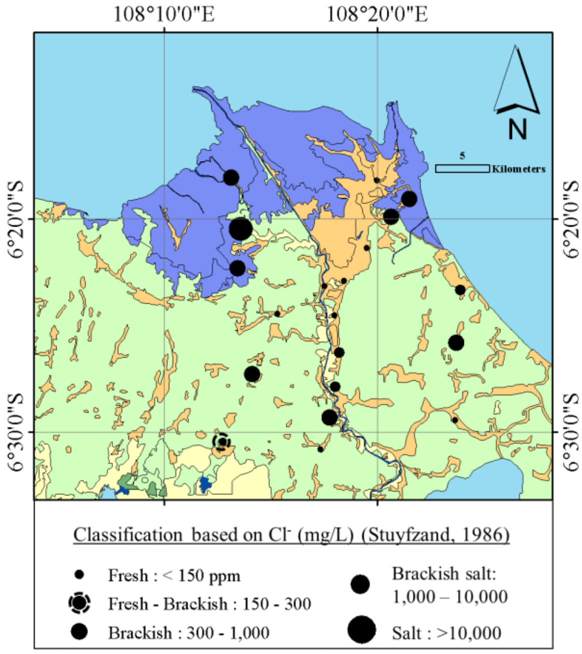

Fig. 3 Cl^- classification distribution.

Cl^- is ubiquitous in water and a conservative substance because relatively stable to chemical reactions. Therefore, Cl^- is usually used as a tracer to help evaluate water quality and chemical reactions occurring in the aquatic environment. There are several natural sources of Cl^- in the freshwater environment, including the intrusion of saltwater from the ocean, natural weathering of bedrock or geologic deposits containing halite, unconsolidated sediments and soils, saline groundwater (brines), and volcanic activity [20].

In coastal regions, a major source of Cl^- is likely the result of saltwater intrusion from the ocean. Therefore, it was assumed the groundwater associated with a coastal area in this study, is heavily influenced by the seawater intrusions, owing to the high Cl^- concentrations observed.

3.1.2 Nitrogen species concentrations

Recent research suggests that nitrogen contamination poses significant risks to the environment [21–25]. According to many previous studies [5–6], [25–27], the dominant speciation of nitrogen contamination in groundwater worldwide is NO_3^--N. In contrast, groundwater analysis of samples from Indramayu, exhibited higher NH_4^+-N concentrations in comparison to NO_3^--N (Fig. 4). The concentration of NH_4^+-N fell between < 0.04 – 9.04 mg/L and was detected at the majority of sites, while NO_3^--N could only dominant at two sites (Fig. 4). NH_4^+-N occurs in the groundwater under reduction condition [28-29] and indicates the presence of human and animal wastes, and agricultural influences, such as fertilizer [30].

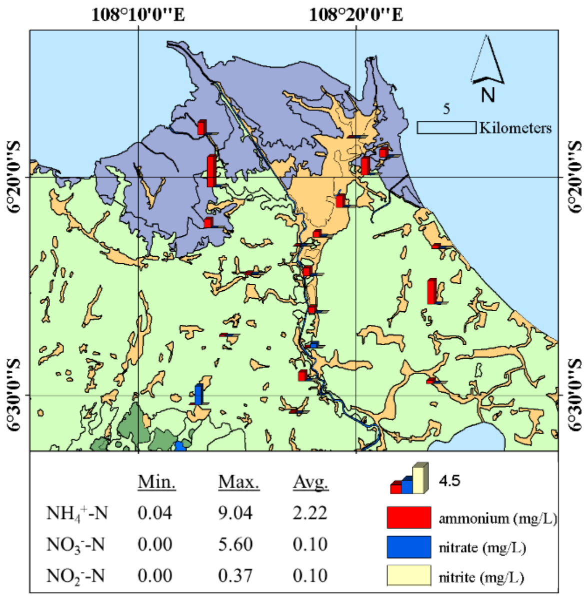

Fig. 4 Nitrogen species percentage and distribution.

As shown in Fig. 4, relatively high concentrations of NH_4^+-N were not only found in the settlement and paddy field areas, but also in the brackish fishpond. Therefore, another source of NH_4^+-N other than human and animal wastes and fertilizer is suspected.

3.1.3 Ammonium sources based on chloride distribution and land-uses evaluation

The previous study showed that ammonium has a close correlation with TDS [31], while TDS in natural water is related to salinity [32]. In this paper, the non-reactive nature of Cl^- along with land-use type variability was used to evaluate the source apportionment of NH_4^+-N in groundwater other than human and animal waste. Fig. 5 depicts the correlation between observed NH_4^+-N and Cl^- concentrations.

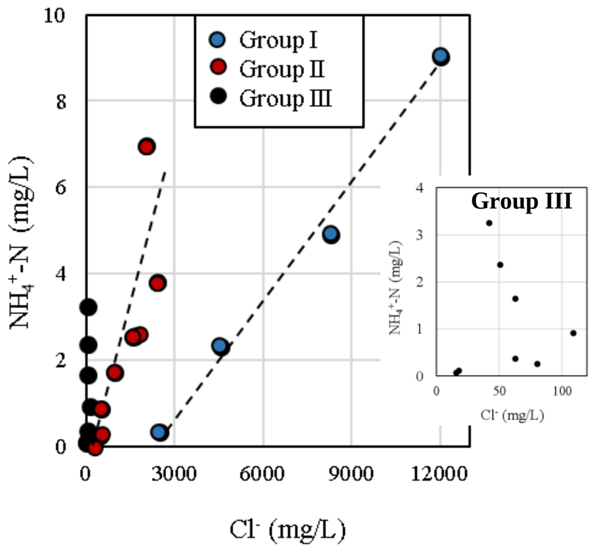

Fig. 5 Correlation between NH_4^+-N and Cl^-

Based on the correlations, underlying values can be clustered into three groups (I –Ⅲ). Groups I and II form linear correlations, while group III shows no relationship between NH_4^+-N and Cl^-. Based on the groups formed, a land-use evaluation was conducted to determine the possible sources of NH_4^+-N (Fig. 6).

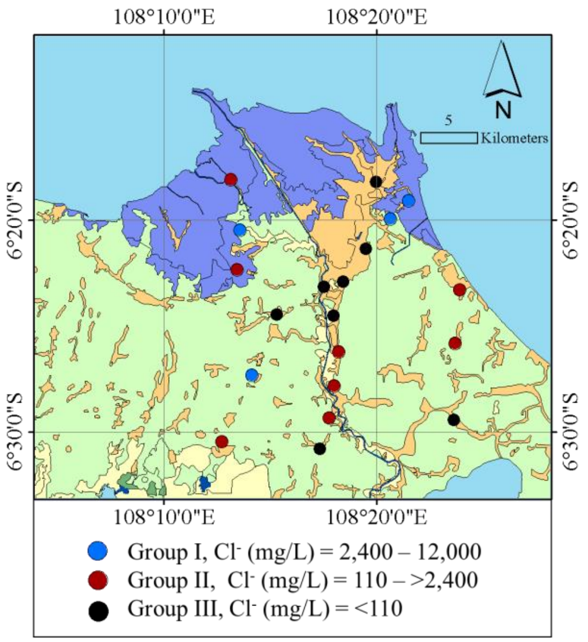

Fig. 6 Distribution of 3 groups clustered by Cl^- and NH_4^+-N concentrations

Samples in group I exhibited the largest range in Cl^- concentration and linear correlation with NH_4^+-N concentrations. Group I samples are mainly located in the brackish fishpond, with the exception of a single point is located in a paddy field area. This point has a much higher Cl^- concentration (2,470 mg/L) compared to other points located in paddy field areas. Therefore, the NH_4^+-N an associated with group I is suggested mainly derived from organic sediment with the ion exchange process.

Ion exchange is one of an important process for ammonium cycle. Whereas, salinity plays a significant role in a cation exchange process of ammonium [28, 33]. Seitzinger [33] found that freshwater sediment can store exchangeable ammonium (NH_4^+) greater than seawater sediment. Simply, the concentration of NH_4^+ will release easier from sediment to water under high salinity condition. The NH_4^+ ion exchange with sodium (Na^+) could be facilitated under high salinity conditions. Another possibility is from dissimilatory nitrate reduction to ammonium (DNRA) reactions, which usually occurs under anaerobic condition and high concentration of organic material [29, 34].

Group II also produced a linear correlation between NH_4^+-N and Cl^- concentrations. The concentration of Cl^- ranged from more than 110 and 2,400 mg/L. The land-use type associated with this group is more varied than the other two groups, and include brackish fishpond, paddy field, and settlement. Therefore, the source of NH_4^+-N for samples from group II is suggested to be from organic sediment, fertilizer, and human and animal wastes.

Finally, group III demonstrates no relationship between NH_4^+-N and Cl^- concentrations. The samples for this group were associated with the lowest Cl^- concentrations (<110 mg/L). The land-use type associated with this group is mainly settlements. It is therefore suggested, that the source of NH_4^+-N for group III is mainly human waste. The process of NH_4^+-N releases to groundwater for this group is assumed to be different from other groups. The reduction condition could occur in this area.

4. CONCLUSION

In the early stages of urbanization of a coastal alluvial plain, groundwater resources may be mineralized by NH_4^+-N. The source of NH_4^+-N is not limited to fertilizer and human and animal waste, but also from organic sediment. Organic sediment NH_4^+ can be released through ion exchange reactions with sodium (Na^+) under high salinity conditions. In further research, a more in-depth study can be carried out concerning other conditions that support ion exchange of ammonium in addition to high salinity.

5. ACKNOWLEDGMENTS

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