Doctoral Thesis

Impact of brackish-water aquaculture activity on groundwater vulnerability in coastal alluvial plain: an evaluation to reach sustainability

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Outline of Dissertation

GENERAL SUMMARY

This dissertation thesis's main goal is to provide a comprehensive evaluation of brackish-water aquaculture`s impact on the vulnerability of groundwater in the coastal alluvial plain. Groundwater in coastal Southeast Asia is reported experiencing a quality decrease because of human activities, e.g., urbanization and groundwater extraction. However, research results show that the influence of brackish-water aquaculture, a well-known Southeast Asia activity, is required for a deeper investigation. An analysis of an integrated physio-chemical and stable isotopes in Indramayu, which is one of the most extensive brackish-water aquacultures in Indonesia, shows deteriorating conditions that are no less severe than other economic activities. The comparison between the dominant land-uses in the study area shows that brackish-water aquaculture (LC) groundwater significantly has higher saline water content than agriculture and residential groundwater (UC). It is found that the Cl^{-} contents in LC groundwater is ranged from 1660 to 16,100 mg/L, averagely more than 10-fold higher than UC groundwater. The highest Cl^{-} concentration is observed in confined LC groundwater within the depth of 20–30 m. The hydrochemistry data and isotopes ratios of $\delta^{2}H$ and $\delta^{18}O$ in water confirm enrichment of seawater that is intentionally introduced to the fishpond.

The results also highlight a significant correlation (*P*-value < 0.1) of high salinity versus trace metals and ammonium-nitrogen contents. Whilst high salinity is potentially categorized as an "anthropogenic" contaminant; the trace metals are conceivably a "natural contaminant" that experience a dissolution process supported by high salinity and reductive aquifer. In respect to reductive condition, both LC and UC groundwater have NH_4^{+}–N as the predominant dissolved organic nitrogen as one indication of the low-redox environment arise. Equal with Cl^{-}, NH_4^{+}–N concentrations in LC groundwater (from 2.4 to 13.0 mg/L) is higher (7-fold) than UC groundwater. The $\delta^{15}N_{NH4}$ values suggest mineralization of organic material in the LC sediments as the primary source of NH_4^{+}–N. Furthermore, only in the LC site, NH_4^{+}–N is significantly correlated with Na^{+}, which implies that mobilization of ammonium sediment to groundwater is promoted by cation exchange.

Finally, nearly 2 x 10^6 m^3 confined groundwater in the LC region is contaminated by Cl^- and trace metals contaminations and vulnerable to elevated NH_4^+–N. This groundwater is not recommended to directly use as clean water because the concentrations of Cl^-, SO_4^{2-}, Na^+, Fe^{2+}, and Mn^{2+} are significantly higher than the guidelines regulated by the World Health organization and Indonesian Government.

SUMMARY CHAPTER 1 – 7

CHAPTER 1 INTRODUCTION

Chapter 1 describes general introduction, background, and objectives. There are three subjects as the focus those are:

(i) Evaluation of saline water contamination determined from hydrogeochemical characteristics (major cations and anions, and bromide concentrations) and environmental stable isotopes (deuterium δ^2H and $\delta^{18}O$ in water, and $\delta^{34}S$ and $\delta^{18}O$ in sulfate).

(ii) Evaluation of dissolved metals contamination and environmental factors that contribute to their presence elucidated from arsenic, iron, manganese, salinity, redox condition, dissolved organic, and geology features.

(iii) Evaluation of nitrogen contamination by analyzing dissolved in organic nitrogen, stable isotope of nitrogen-15, and geology features.

CHAPTER 2 STUDY SITE AND ANALITYCAL METHODS

Chapter 2 provides knowledge related to selected study site, samples collection, methods, and study literature. The area under study is Indramayu, which is located on the north coast of Java Island, Indonesia (107°52′–108°36′ E and 6°15′–6°40′ W). Indramayu is vital as a rice- and fish-production area with increasing gross domestic regional product (GRDP) particularly in the farming and fishery sector. It is a lowland area with groundwater level is generally 3.5 m.a.s.l and flows toward the sea.

The hydrogeology profile shows a thick clay in the north, which comprises a slight to dense carbonate clay with shell fragments.

A total area of Indramayu is greater than 240,000 ha. The main land-use categories are agriculture, brackish-water aquaculture, and residential. In this study brackish-water aquaculture is consistently written as LC while other land-uses is written as UC.

Samples are collected for sediments and groundwater. The sediments samples are collected at different depths from two boreholes in UC and LC areas to understand the geological conditions of the aquifer system. Groundwater samples are collected two times, in August 2017 (dry) and November 2019 (very dry). There are 18 groundwater samples (5 from LC; 13 from UC) and 28 groundwater samples (10 from brackish-water aquaculture area; 18 from agriculture and settlement areas) collected in the first and second sampling periods.

Water quality are analyzed both in the field and in the laboratory. Water temperature (WT), pH, oxidation–reduction potential (ORP), dissolved oxygen (DO), dissolved Fe (Fe^{2+}), and bicarbonate (HCO_3^-) are measured as field parameters. The laboratory analysis is conducted for three categories, those are chemicals parameter, stable isotopes, and biological parameters. The chemical parameters are: (i) major anions and cations, (ii) metals (arsenic, dissolved iron and manganese), and (iii) dissolved inorganic nitrogen (nitrate, nitrite, and ammonium). The stable isotopes analysis is for (i) Deuterium and ^{18}Oxygen in water, (ii) ^{34}Sulfur and ^{18}Oxygen in sulfate, and (iii) ^{5}Nitrogen in ammonium.

CHAPTER 3 BRACKISH-WATER AQUACULTURE IMPACT ON SALINITY IN GROUNDWATER

Chapter 3 aims to respond to the first specific objective. This chapter describes the possibility of elevated salinity in the groundwater system due to brackish-water aquaculture activity. It provides a detail examination of the salinity level in unconfined and confined groundwater by using hydrochemical approach. Also, groundwater origin and saline water source is elucidated from stable isotopes of δ^2H and $\delta^{18}O$ in water, and $\delta^{34}S$ and $\delta^{18}O$ in sulfate.

The hydrochemistry data coupled with stable H and O isotope ratios in water, and stable S and O isotope ratios in sulfate reveals that brackish-water aquaculture activity in selected study area potentially contributes to the elevated salinity. The groundwater in the brackish-water aquaculture region has significantly different chemical characteristic compared to agriculture and settlement areas. The chemical properties of groundwater in the brackish-water aquaculture area are as follows:

1. The hydrochemical characteristics of groundwater indicates high saline-water contents with characteristics as follows: (i) high and dominant contents of Cl^- and Na^+; (ii) strong correlation between Na^+ and Cl^-; (iii) enrichment of Ca^{2+} concentration and depreciation of Na^+ content; and (iv) a relatively high Mg^{2+}/Ca^{2+} ratio but significantly low HCO_3^-/Cl^- ratio.
2. The δ^2H and $\delta^{18}O$ compositions in water are significantly higher than other types of land-use and situated in the vicinity of seawater mixing line. It implies high saline-water content and evaporation process. Moreover, the impact of saline-water is likely more severe when the precipitation is reduced.
3. The comparison of $\delta^{34}S$ and $\delta^{18}O$ composition in groundwater and local seawater describes the possibility of saline-water present in some samples. At the same time, groundwater from agriculture and settlements potentially contaminates by detergent and fertilizer.

CHAPTER 4 BRACKISH-WATER AQUACULTURE IMPACT ON DISSOLVED METALS IN GROUNDWATER

This chapter presents explanation of the second specific objective. Also, it determines the vulnerable area to dissolved metals contaminations and describes environmental factors that potentially influence the increase in these metals' concentration. This chapter has been published in *Springer Nature Applied Sciences* (https://doi.org/10.1007/s42452-021-04385-y).

The results show that the groundwater of Indramayu is potentially vulnerable to natural contamination of Fe^{2+} and Mn^{2+}. On the other hand, the concentrations of As are very low or under the threshold of drinking-water regulation. The most vulnerable area is located primarily at the north tip of Indramayu, a brackish -water aquaculture area composed of deltaic deposits. With respect to clean water regulations, the groundwater in Indramayu requires treatment before utilization, specifically focusing on Mn^{2+}, then Fe^{2+}. The significant different of metals concentrations between two times of sampling periods are detected in several point which the metals concentrations are increased significantly.

High salinity is suspected of supporting the leaching of Fe^{2+} and Mn^{2+} from minerals and soils through ion-exchange processes. The reduced environment indicated by DO and NO_3^-–N reduction is another crucial factor in the dissolution of Fe^{2+} and Mn^{2+} from soils and minerals to groundwater. This reduction process is possibly promoted by microorganisms and thus occurs as a function of organic matter. Finally, it is necessary to consider Fe and Mn as damaging natural contaminations to alluvial coastal aquifers. Consequently, appropriate groundwater management strategy is required to maintain sustainability in the region.

CHAPTER 5 BRACKISH-WATER AQUACULTURE IMPACT ON NITROGEN IN GROUNDWATER

Chapter 5 is the explanation of the third specific objective. This chapter evaluates the predominant dissolved inorganic nitrogen (DIN) in the groundwater system. Furthermore, potential sources of predominant DIN are elucidated from a combination of stable isotope of ^{15}Nitrogen, coliform bacterial, major cations, geology features, and land uses. This chapter has been published in *Water* (https://dx.doi.org/10.3390/w13010025).

NH_4^+–N in the LC region, potentially originated mainly from the mineralization of soil organic nitrogen to ammonium. In agreement with this origin, the ratios of $\delta^{15}N$ in the sediments indicate the mineralization of nitrogen in sediment. However, contamination by anthropogenic activity is possible considering the high values of total coliform bacteria. The strongly positive and significant relationship of NH_4^+–N and Na^+ suggests that under high salinity, the exchangeable NH_4^+ is mobilized from sediments to the groundwater through cation exchange. Additionally, the high salinity of groundwater possibly arises from the brackish-water pond.

Further, attenuation of ammonium-nitrogen from manure, sewage, and pit latrines occurs in the groundwater in the UC region. Both total coliform and *E. coli* values confirm this condition. The ratios of $\delta^{15}N$ in several layers of sediments suggest the possibility of nitrogen mineralization to ammonium; nevertheless, the nitrogen contents suggest that this process is more likely in the sediments of lower coastal region. The significantly lower salinity followed by weak and not significant relationships of NH_4^+–N and all major cations indicate less possibility of NH_4^+–N mobilize to groundwater through cation exchange.

CHAPTER 6 DEGRADATION OF GROUNDWATER RESOURCES

Analysis of hydrochemical parameters and environmental stable isotopes, along with an evaluation of land-uses and geology features reveals that brackish-water aquaculture activity potentially contributes to groundwater quality degradation. The groundwater is vulnerable to the following: (1) salinization (Cl^-, SO_4^{2-}, and Na^+), (2) dissolution of trace metals (Fe^{2+} and Mn^{2+}), and (3) elevated concentration of ammonium-nitrogen (NH_4^+–N). Furthermore, the groundwater vulnerability levels and specific source for selected contaminants are diverse.

In general, salinity (reflected with Cl^-), dissolved metals (reflected with Mn^{2+}), and NH_4^+–N concentrations are relatively low in the northern aquifer and gradually increase to the southern aquifer. The confined groundwater losses due to brackish-water aquaculture activity are potentially ±2.8 x 10^6 m^3 and 1.7 x 10^6 m^3, respectively due to salinity and trace metals contamination. Equally, there are possibly ±1.8 x 10^6 m^3 confined groundwater of brackish-water aquaculture site vulnerable to elevated NH_4^+–N contents.

CHAPTER 7 CONCLUSION

The groundwater quality analysis results show that the brackish-water aquculture activity contributes to high contents of saline water, dissolved iron and manganese. In the meantime, although no indication of dissolved inorganic nitrogen, the groundwater in LC site is potentially vulnerable to elevated concentration of ammonium-nitrogen. Consequently, the groundwater is not suitable for drinking-water purpose. In addition, this resource is possibly problematic in the future without appropriate management. To protect the sustainability, the government should consider groundwater resources carefully in management of brackish-water aquaculture activity.

Published Paper

Rusydi AF, Saito M, Ioka S, Maria R, Onodera SI (2019) Estimation of ammonium sources in Indonesian coastal alluvial groundwater using Cl^- and GIS. Int J of Geomate 17(62):53–58. https://doi.org/10.21660/2019.62.4749

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