Preliminary Study Contamination of Organochlorine Pesticide (Heptachlor) and Heavy Metal (Arsenic) in Shallow Groundwater Aquifer of Semarang Coastal Areas

Baskoro Rochaddi1, Chrisna Adhi Suryono2, Warsito Atmodjo3 and Alfi Satriadi3
1Oceanography Department, Diponegoro University, Semarang, Indonesia
2Marine Science Department, Diponegoro University, Semarang, Indonesia
Email : rochaddi@ymail.com

Abstract. The present study was conducted to assess the level of pesticide and heavy metal contamination in shallow aquifer of Semarang coastal areas. Results indicated that Heptachlor and Arsenic were detected in the water samples in the range 0.023-0.055 µg L\(^{-1}\) and 0.03-1.63 µg L\(^{-1}\), respectively. Compared to the standard limits of the organochlorine contents in the water sample by World Health Organization (WHO) limits and Indonesian Drinking and Domestic Water Quality Standard for Ground Water (IWQS), groundwater of Semarang Coastal Areas was contaminated with pesticide and heavy metal. This study has proven the presence of organochlorine and heavy metal contamination of some shallow aquifer supplies in the coastal areas of Semarang.

Keyword: Pesticide, Heavy Metal, Shallow Aquifer

1. Introduction
Semarang coastal land and water has been contaminated by heavy metals and pesticides. The anthropogenic heavy metals have found in coastal water and land in western part of Semarang [1]. Furthermore, found several heavy metals in marine sediments such as Cr, Pb, and Cu [2]. Some of the organochlorine pesticides on marine water (Heptachlor, Aldrin, Endosulfan, Endrin, and pp-DDT) has been investigated, and it showed that all of the marine water samples contaminated with organochlorine pesticides[3]. The previous research proved that the coastal are of Semarang has been contaminated by heavy metals and organochlorine both on the marine water and sediment. The heavy metals in water and sediments there is predicted come from outside as well as inside of Semarang coastal water. Semarang coastal area has been surrounded by many rivers such as Semarang river, West Banjir Kanal River, East Banjir Kanal River, Babon and Banger river. While the heavy metals which come from inside coastal water are assumed come from metal corrosion of ships and buildings.

Pesticides are usually used in agriculture since their benefits to mankind, and the environment outweigh the risks associated with their application[4]. However, rice cultivation, especially in low-lying paddy fields, is inherently risky to the environment since paddy water normally flows out into the surrounding water[5]. There have been several cases found where rice pesticides moved out of their intended area and contaminated the surrounding environments like rivers and lakes[6-8]. Consequently, large-scale application of these toxic materials in agriculture areas can contribute to the presence of those compounds in surface and groundwater, lakes, estuary and ultimately in the coastal areas. Most of these compounds are recalcitrant to biodegradation and their entry into the aquifer might be, poses many challenges to the existing of drinking well water from pollution. The possibility of pesticides polluting the coastal or marine environment is considered unlikely since paddy effluents normally travel long distances before they reach coastal areas. By then, most of the residues are degraded by physicochemical and biological factors. However, this is only true for rice paddy fields located upstream or more inland. Nowadays, nevertheless, humans are forced to utilize even the coastal lowlands for rice cultivation [9] due to the limited area of agricultural lands. Consequently, this increases the possibility of pesticides ending up in coastal environments.
Arsenic (As) is one of the most important toxicants that is widely distributed in nature and occurs in the form of inorganic and organic compounds. Exposure to inorganic compounds may occur in a variety of ways through certain industrial effluents, chemical alloys, pesticides, wood preservative agents, combustion of fossil fuels, catalysts, glass, fire retardant, occupational hazards in mining that are anthropogenic and natural sources [10]. Arsenic is known to cause arsenicosis due to its manifestation in drinking water, the most common species being arsenate As (V) and arsenite As (III) [11]. Arsenic contamination of groundwater has led to a massive epidemic of arsenic poisoning in Bangladesh [12-13] and neighboring countries [14-15]. It is estimated that approximately 57 million people in Bangladesh [16] and 11 million people in Vietnam [14] are drinking groundwater with arsenic concentrations elevated above the World Health Organization's standard of 10 parts per billion [17]. This tragedy was occurring as the high arsenic levels present in drinking water. Bangladesh maintains an allowable limit of 50 ppb, and the limit in Canada is 25 ppb [18]. The arsenic found in Bengali and Canada drinking water is present naturally in the sediment but is released only by humans, primarily from the digging of shallow wells. Similarly unacceptably high arsenic levels are found in shallow-well domestic drinking water in the American Midwest [19].

Recently there has been increasing anxieties concerning arsenic-related problems. Occurrence of arsenic contamination has been reported worldwide, for example, Mongolia [20], USA [19][21-22], Bangladesh [12-13], Taiwan [23], Spain [24], et cetera. In Indonesia, no significant As contamination has been reported yet. However, local contaminations in soils, sea, and groundwaters with high As and Hg levels have frequently been revealed particularly around Buyat Bay mining areas [25-26].

Clean water is probably the biggest problem in many houses of coastal urban settlement. Based on field investigation, many homes in this region are not connected to the city water supply, and wells are often unpredictable. Most houses do not have overhead tanks for catchment or storage but, instead, have large ground storage tanks from which the water is pumped on demand by a small electric pump. The most prevalent of these is the shallow aquifer (dug well) which has become the major source of water in the coastal inhabitants. However, groundwaters and surface waters are never pure that probably contain a variety of organic compounds [27-28]. Organic chemicals such as polychlorinated biphenyls, PAHs and organochlorine pesticides have been identified in the surface water [29-31] and groundwater [32-34]. Contaminants from residential areas, industries and agricultural practices through migration and infiltration enter to ground waters [36-37]. Therefore, a study of the distribution and concentration of organochlorine in the shallow aquifer become the most relevant. This research aims to examine the organochlorine pesticide (Heptachlor) and Heavy metal (Arsenic) content of the shallow aquifer in Semarang coastal areas and assess how far conform to WHO/ Indonesia recommendation for fresh water/ drinking water.

2. Material And Methods

Water sample collection: Sampling points were located on coastal areas flowing from the west part to the east part of Semarang. Figure 1 shows the sampling sites; the water samples were collected in polyethylene plastic bottles from 30 different dug wells across those cities. The water samples were collected in 1000 mL plastic bottles. Then, bottles were properly labeled and tightly sealed. All the water samples were brought to the laboratory for performing organochlorine pesticide (Heptachlor) and Heavy Metal (Arsenic) analyses. The parameters analyzed were psycho-hydrochemical properties and organochlorine pesticide (Heptachlor) and Heavy Metal (Arsenic) contents of shallow groundwater. The pH, salinity, and temperature were measured at the time of samples collection using pH meter, hand refractor salinometer and a standard centigrade thermometer (Horiba Co. Ltd., Japan. The analytical Arsenic procedure applied was the method of the Standard Method for the Examination of Water and Waste Water [37] using 15% methylene chloride in n-hexane and capillary columns. One-liter samples were extracted with a solvent mixture and then concentrated in a Kuderna-Danish apparatus. The extracts were cleaned up with Florisil column. The final pesticide extract was obtained in 5 mL of hexane. The samples were then analyzed by gas chromatograph Model Hitachi 163 FPD.
(Flame Photometer Detector), and nitrogen High Pure (HP) was used as the carrier gas. A 2 in a glass column (3 m, n ID) packed with 3% Silicon OV1 on 80-100 mesh Supelcon was used. Gas flow at 30 mL min⁻¹, column temperature at 160-230°C, detector temperature at 290°C and the injector temperature at 290°C were maintained. Arsenic metal analysis. The water samples were acidified with an aliquot of HNO₃. About 100 mL each of the well-mixed acidified water was digested with five mL HCl. The digested samples were then analyzed for As and Fe using atomic absorption spectrometer Perkin Elmer Model 5000 [38]. Statistical analysis was conducted among the standard parameters using Linear correlation method. This was performed to know whether any relationship among the parameters existed or not.

![Figure 1. The map of sampling site location](image)

3. Results And Discussion

Semarang coastal areas are divided into three areas (West, central and East). This city was chosen for study because of their representative for the main industrial and human center of northern coastal urban. Hence, those cities have a great probability of the existence of declining water quality resources. Pollution of groundwater has been reported for a number of urban throughout the world on a wide range of pollutants, such as organochlorine, heavy metals, nitrate, fecal, bacteria, virus, domestic waste, pesticide [39-47]. This is mainly due to a higher population density and more intensified agricultural and industrial activities [48-50]. In the present study, the results of some physicochemical parameters and selected priority Organochlorine Pesticide (Heptachlor) and Heavy Metal (Arsenic) in Shallow Groundwater Aquifer of Semarang Coastal Areas are preserved in Table1 and 2, respectively.

Table 1. Physico-chemical properties of water samples of shallow groundwater in the Semarang coastal areas

| Sampling Site No | Colour    | Odor     | Taste | pH | Salinity | Temp |
|------------------|-----------|----------|-------|----|----------|------|
| 1                | Brownish  | Offensive| Tasteless | 7  | 2        | 29   |
| 2                | Bluish    | Offensive| Tasteless | 7  | 0        | 29   |
| 3                | Colourless| Offensive| Tasteless | 7  | 2        | 29   |
| 4                | Colourless| Offensive| Tasteless | 7  | 2        | 29   |
| 5                | Greysh    | Offensive| Tasteless | 7  | 0        | 29   |
| 6                | Colourless| Offensive| Tasteless | 7  | 2        | 29   |
| 7                | Colourless| Offensive| Tasteless | 7  | 0        | 29   |
| 8                | Colourless| Offensive| Tasteless | 7  | 5        | 29   |
This study indicates that low variation existed among some physicochemical parameter (temperature, salinity, pH). Colour, taste, and odors were detected in water samples. Some of the water samples, particularly some water samples, did not comply with the standard limits for drinking. The water samples have color and offensive odor. The water temperature values obtained in this study varied slightly ranged from 28.5-29.5°C with a mean of 28.93°C. This is the most common water temperature in the tropical zone. This temperature range of water samples is supposed to be affected by the intensity of the sunlight as the temperature rose from 28.5-29.5°C on relatively hot day afternoon. Some workers reported that the temperature of water from tropical climate was little affected by seasonal variations of temperature [51-52]. Salinity range was varied from 0 to 9.0‰ with a mean of 2.1±2.51‰. The highest desirable level for pH (7.0) is within the range of 6.5-8.0 values for drinking purposes [53].

| Number | Contaminant Method | Remark |
|--------|-------------------|--------|
| 1      | 0.038             | 0.54   |
| 2      | bd                | 0.13   |
| 3      | 0.058             | bd     |
| 4      | 0.041             | 1.63   |
| 5      | bd                | 0.5    |
| 6      | 0.025             | 6.21   |
| 7      | bd                | 3.42   |
| 8      | bd                | 0.03   |
| 9      | 0.023             | bd     |
| 10     | 0.027             | bd     |
| Range  | 0.0-0.058         | 0-6.21 |
| Means + SD | 0.0228 ± 0.02     | 1.2463 ± 2.05 |

WHO/lna 0.03/0.03 0.5

Table 2. Range, frequency of occurrence and mean ± standard deviation of organochlorine pesticide (Heptachlor) and Heavy Metal (Arsenic) residue (µg L⁻¹) from shallow groundwater Semarang coastal areas

Bd: bellow detected

Water samples from coastal areas of Semarang showed the presence of selected organochlorine pesticide (Heptachlor) (Table 2). Ten samples (60%) contained residues of heptachlor, ranging from 0 to 0.055 µg L⁻¹, and 70 % of samples were Arsenic contaminated whereas ranging from 0 to 6.21µg L⁻¹. Comparison of the organochlorine contents in the water sample with World Health Organization (WHO) limits and Indonesian Drinking and Domestic Water Quality Standard for Groundwater showed that some of the household wells were contaminated with organochlorine.

The mean levels of heptachlor were exceeded the maximum permissible levels for drinking water. Variation in residual levels in these samples could be the due location of the sampling. The present study sufficiently shows the presence of organochlorine residues in most of the samples. Comparison of data on organochlorine residues of Semarang areas water samples showed that the highest levels and frequency of occurrence on heptachlor. Although we have not analyzed surface water samples for organochlorine residues (Heptachlor) in the present investigation, comparing the results reported here with other regional studies on the river and sea water contamination [54], different contamination levels with heptachlor. Heptachlor was not detected. It is not known whether this is due to irresponsible insecticide handling practice or groundwater transport processes.
The appearance of arsenic in groundwater and drinking water is a worldwide concern since it has been found that arsenic is one of the most toxic and carcinogenic chemical elements. In the present study As metal concentration was ranging from 0 - 6.21µgL⁻¹ respectively in three different areas of Semarang coastal land. The maximum As concentration was observed in Central part of Semarang (6.21 µgL⁻¹) and (3.42 µgL⁻¹) in East part of Semarang. This condition could be explained that first, arsenic can enter the water supply from natural deposits in the earth or industrial and agricultural pollution. It is a natural element used for a variety of purposes within industry and agriculture. It is also a by-product of copper smelting, mining, and coal burning. Industries in Semarang were suspected release much more arsenic into the environment every year. Once released, arsenic remains in the environment for a long time. However, in the present results of this study when it was compared to the previous results in Buyat Bay for Drinking Water[53], it was lower. Because it may be stated that the environmental parameters like salinity, temperature, dissolved oxygen, and pH also have some effect on the accumulation of trace metals.

4. Conclusion
Characteristics of selected physicochemical parameters and organochlorine pesticide (Heptachlor) and Heavy Metal (Arsenic) residues in shallow groundwater on Semarang coastal areas has been determined in this study. Heptachlor was above the maximum permissible value recommended by WHO and Indonesian Water Quality Standard. Low concentrations of arsenic in water which are continuing to be used need to be monitored over time by authorities concerned to ensure that concentrations of arsenic in water of these shallow aquifer remain at an acceptable level over time. Some of these aquifer needs further purification to ensure their sustainability for consumption of coastal urban inhabitants. It is necessary to study further in these areas during the wet and dry period due to fully evaluate the organochlorine and heavy metal impact of pollution on shallow aquifer in seasonal variation.

5. Acknowledgment
This article is part of the research which supported by the grant from non APBN 2017 Faculty of Fisheries and Marine Sciences Diponegoro University

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