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RESEARCH ARTICLE

Heavy metal contamination in the marine environment of Pekalongan, Indonesia: Spatial distribution and hydrodynamic modeling

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Abstract

Pollution in Pekalongan waters is one of the environmental problems caused by the industrial city. Batik industry, the most common activity at Pekalongan, uses a lot of water and generates a lot of effluent because it involves an aqueous process when dyeing fabric. This wastewater, contains heavy metal contaminants like hexavalent chromium (Cr+6), copper (Cu), and lead (Pb), generally finds its way to rivers that run across Pekalongan. The contamination level, spatial distribution, and transport simulation of copper (Cu) and chromium (Cr) in water were investigated in the Loji and Banger rivers in Pekalongan, Indonesia. A two-dimensional hydrodynamic model was constructed to simulate heavy metal concentrations in marine environment of Pekalongan City, Indonesia. The model's kinetic processes comprised air exchange, transportation, and deposition. Thus, Cu concentrations were below Indonesia's threshold, but Cr, especially in the river and outlet river, exceeded it. Both rivers have 0.002 mg/L Cu. Since water freely flows at Banger River Estuary, Cr concentration reach the highest in both rivers, 0.1056 mg/L. The evaluation findings and the numerical Cu and Cr calculation model created in this work have important research ramifications for preventing and controlling heavy metal pollution.

KEYWORDS
Banger river, chromium (Cr), copper (Cu), heavy metal, Loji river

1 | INTRODUCTION

Pollution is a significant element in the degradation of the marine and coastal environment, as it can harm or kill biological components (biotic) and jeopardize the health of humans who ingest contaminated biota (Vatria, 2010). Whether directly or indirectly, the discharge of waste pollutants into coastal areas such as estuaries would have a collateral effect due to the ocean's dynamic nature and the available natural food chains (Wulandari et al., 2014). The most severe consequences of pollution carried by a river will be focused in the region directly adjacent to the river's discharge of fresh water (river plume) (Caddy, 2000). Heavy metal contamination is become a major concern in coastal areas, this water pollution is caused by industries, houses (residential), and agriculture, and is a contributing element to environmental degradation as evidenced by the waste quality standard (Mratihatani & Susilowati, 2013). If the fish obtained are consumed on a regular basis, it will have a negative impact on health because the river is also polluted with heavy metals. Wastewater has a significant impact on human health, causing waterborne diseases such as intestine inflammation, cholera, cystosomiasis, and hepatitis infection. In addition to being a disease transmitter, wastewater contains disease-causing agents (Firmansyah et al., 2020).

Indonesia is currently confronted with a land-based pollution problem that is inextricably linked to a cherished aspect of its culture (Ahmad et al., 2010). One of Indonesia's cultural legacies is a traditional method of fabric production known as Batik. Pekalongan is a city in Indonesia's Central Java Province that is well-known for its batik production. In general, the batik industry in Pekalongan is a cottage industry or small and medium enterprises (SMEs). Batik industrial wastewater is primarily derived from the dyeing process; in addition to high-color substances, industrial wastes batik also contains synthetic ingredients that are soluble or difficult to untangle in thickly coloured liquid waste containing heavy metals (Mratihatani & Susilowati, 2013). The type of dye used in the batik process is synthetic and as a result cannot be degraded by the natural environment. Synthetic dyes are a popular choice since they are less expensive than natural dyes. The ongoing demand for batik has led to the employment of synthetic dyes as part of the coloring process (Handayani et al., 2018). Due to rising environmental awareness and global concerns for pollution management, these dyes, as well as the waxes used in the batik process, have come under investigation. Wastewater is defined as waste products resulting from shear batik, batik-making activities, and batik laundering. Each river and drainage system in Pekalongan has poor water quality, as a result of batik production waste being dumped directly into water bodies such as irrigation or rivers (Santi et al., 2017). From previous research, Cr is toxic heavy metals that have a negative impact on a variety of microorganisms, plants, and animals. Toxic heavy metal exposure in humans can results in serious health injures and impairments, and even death in certain extremities (Rahman & Singh, 2019). Research by (Syakti et al., 2015) in Segara Anakan rivers and (Amin et al., 2009) in Dumai, Cr and Cu concentration are likely to harm organisms. Copper enters the environment via waste dumps, domestic waste water, combustion of fossil fuels and wastes, wood production, phosphate fertiliser production, and natural sources (ASTDR, 2002; Tchounwou et al., 2012), while the Cu and Cr are derived from the batik dyeing process.

Cr has been implicated in animal genetic mutations and possesses additional carcinogenic properties (Shanker & Venkateswarlu, 2011). Cr are likely to end up in textile effluent as a catalyst in aqueous textile processes, specifically dyeing. Additional research has demonstrated that dyes have the potential to significantly reduce the overall biomass of freshwater aquatic systems (Samchetshabam et al., 2017). Contamination of rivers and other aquatic systems can have a detrimental effect on the socioeconomic stability of communities that rely on these environments for services. In humans, long-term exposure to dyes and their influence on health should be understood. For instance, skin irritation is a frequent health complication associated with prolonged exposure to textile dyes, as are respiratory problems (Hassaan & Nemr, 2017). While it is obvious that textile waste treatment is necessary, not all treatment processes are effective. Metals such as chromium (Cr), copper (Cu), zinc (Zn), and arsenic (As) found in effluent must be considered (Hassaan & Nemr, 2017). Textile manufacture may have heavy metal concentrations from synthetic dyes. Heavy metals, such as Cd, Cu, and Pb, can be found in reactive dyes. Heavy metals like Cu, Cr, Cd, Fe, Pb, Ni, and Zn are found in several dyes used in the textile industry (Oginawati et al., 2022). Excessive copper intake can have negative health consequences associated with deficiency, including dizziness, nausea, and stomach cramps, as well as a chronic effect on organ damage, including kidney and liver disorders (Stern, 2010). Additionally, the majority of textile businesses in Pekalongan are small and medium-sized enterprises (SMEs), which lack the necessary wastewater treatment facilities (Madhav et al., 2018; Samchetshabam et al., 2017). Therefore, this research aimed to study the distribution characteristics, sources and dynamic mechanism of heavy metals in the water and sediments of marine environment of Pekalongan, Central Java, Indonesia.

2 | MATERIAL AND METHOD

2.1 | Water samples

In June 2020, water samples were collected from thirteen locations for heavy metal sampling. The study area and sample sites are depicted in Exhibit 1. At least two water samples were taken using a Nansen bottle. The water sample was taken in the water column below the surface water (5–15 cm), then placed in a non-translucent sample bottle with a capacity of 2 L and stored in a cooler (cool box), where ice cubes are added to keep the temperature in the cool box close to 0°C. Once all the water samples have been placed in the cool box, the cool box was tightly closed to avoid biological, chemical, or physical changes occurring between data collection and analysis (Fitriyah et al., 2013).

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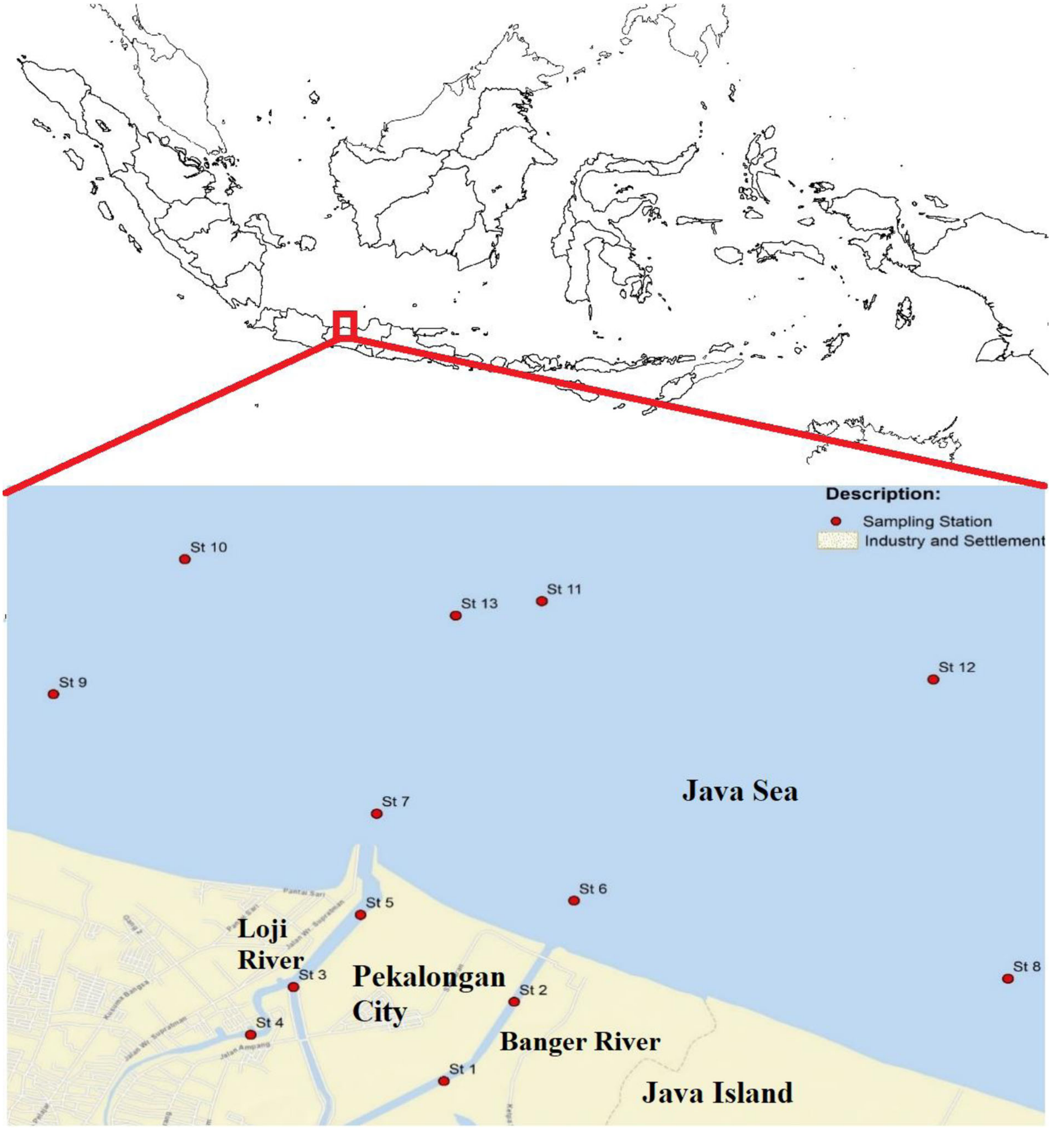

EXHIBIT 1 Study area and location of sample sites. [Color figure can be viewed at wileyonlinelibrary.com]

keep the temperature in the cool box close to 0°C.

2.2 | Bathymetry

The seabed grading method is intended to obtain bathymetric information. By using an echosounder integrated with GPS, the device used is the Garmin map 585 s. Depth measurements of sea level by means of echosounder conducted from a motor boat, with a maximum boat speed of five knots (2.5 m/s) and the ship's condition stabilized with strips sounding intervals of 100 m. The coordinates of the measurement points are obtained using a GPS (Global Positioning System) which is integrated with the echosounder. Bathymetric mapping to be carried out is divided into two phases: the preparation phase in the form of secondary data collection, which is a topographic map of the land/coastal and regional bathymetric maps in the form of the Marine Environment Map. Subsequently conducted primary data collection, which includes: measurement sounding and measuring tides.

EXHIBIT 2 Waste water sample analysis results

Station	Longitude	Latitude	Copper (Cu)* (mg/L)	Chromium (Cr) (mg/L)	Characteristics
St 1	109.691268	−6.877119	<0.002	0.0509	Banger river, fishpond, Mangrove
St 2	109.698129	−6.873144	<0.002	0.0553	Mouth of Banger river, fishpond, Residence
St 3	109.702438	−6.867390	<0.002	0.0419	Batik industries, Boat dock, Residence
St 4	109.688881	−6.866300	<0.002	0.0319	Residence, Food industries, boat dock
St 5	109.686258	−6.869778	<0.002	0.0298	Mouth of Loji river, Port, Fish shelter
St 6	109.693002	−6.861048	<0.002	0.1056	Banger river estuary, Coast of slamaran beach
St 7	109.706112	−6.860040	<0.002	<0.009	Loji river estuary
St 8	109.694006	−6.853703	<0.002	<0.009	East coast, Agriculture
St 9	109.73275	−6.865694	<0.002	<0.009	West coast, fishpond
St 10	109.674159	−6.845015	<0.002	<0.009	High sea
St 11	109.682224	−6.835191	<0.002	0.0103	High sea
St 12	109.704121	−6.838242	<0.002	<0.009	High sea
St 13	109.728166	−6.843929	<0.002	<0.009	High sea

*Detection limit for Cu Analysis can only detect above 0.002 mg/L.

2.3 | Current measurement and analysis

Measurement of water flow data using the Eulerian/Euler method. Retrieval and recording of current measurement data at this location using the ADCP Argonaut SonTek XR with wavelength sensor beam 0.75 MHz and autonomous multicell system. The accuracy of this measurement module is 0.01 cm/s, with a maximum column depth of 15 m measurement, a maximum of 10 cells can be measured, and a minimum layer thickness of 0.8 m with a maximum speed that can be recorded measuring is 6 m/s. This equipment is included with measurements in the dynamic column. Data recording of the speed and direction of the field current is needed to verify the accuracy of the model. Determination of 1 observation station by recording the amount of current for 3 days with a time interval of 10 min. Determination of the location of current measurements using the area sampling method, the method used to determine the sample if the data source is very broad and the sampling is based on a predetermined area (Kowalik & Murty, 1993). Determination of the flow measurement point carried out in a safe area and not a lot of shipping activities. The measuring depth is divided into five active layers, to obtain a variable current velocity profile along the measured water column. Thick layer of each division is 2 m on every layer, including the depth of 15 m, 12 m, 9 m, 6 and 3 m bpl (See Exhibit 2).

Measurement of water flow data using the Eulerian/Euler method. Retrieval and recording of current measurement data at this location using the ADCP Argonaut SonTek XR with wavelength sensor beam 0.75 MHz and autonomous multicell system

Field flow data analysis is presented in graphical form so that it is easy to describe. Current data processing is done with the 2 D Numerical Model.

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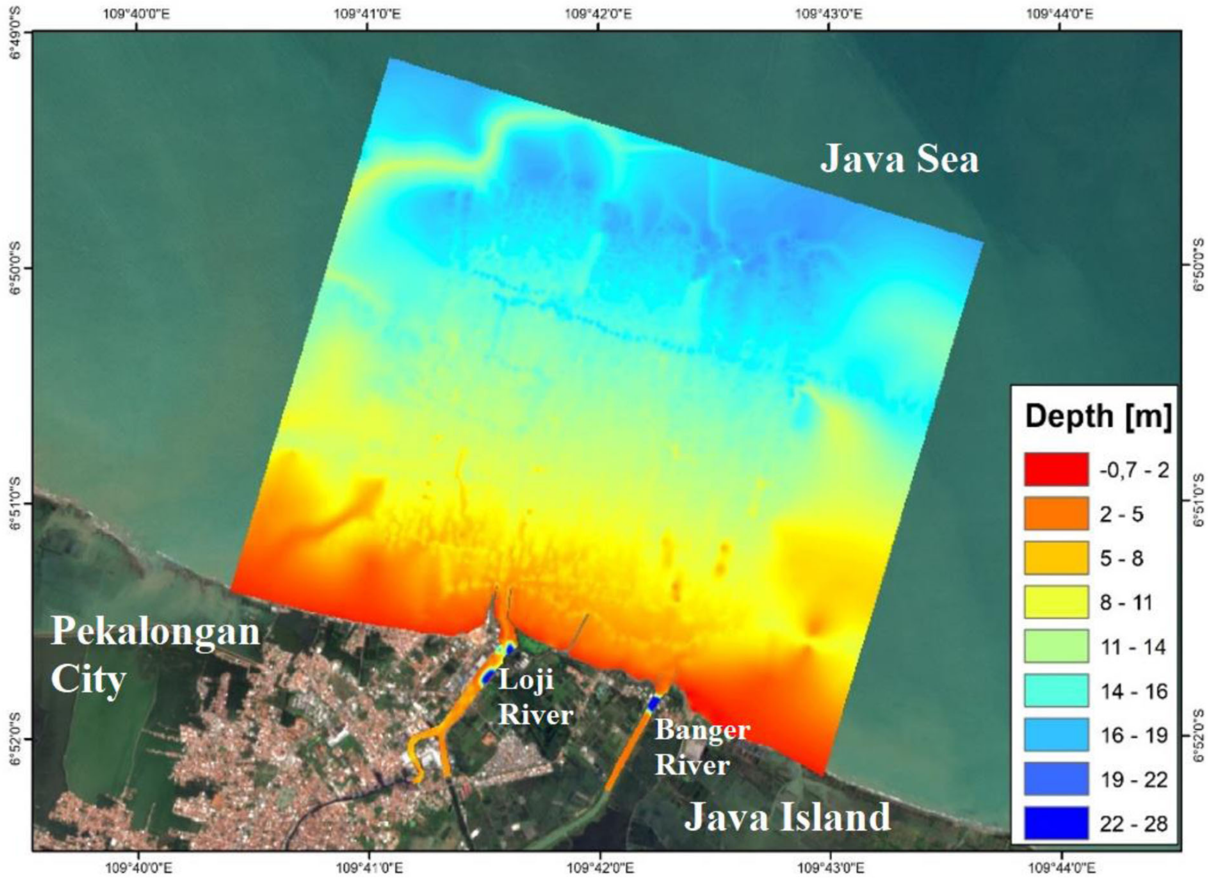

EXHIBIT 3 Variation of water depth in marine environment of Pekalongan. [Color figure can be viewed at wileyonlinelibrary.com]

done with the 2 D Numerical Mode

2.4 | Recording of water elevation

Tide measurements were taken to ascertain tidal characteristics, allowing for the determination of sea level elevation, tide type, and tidal components via the Virtuose single channel logger RBR. Following that, the tide data were analyzed to determine the tide type, mean sea level (MSL), and water level (maximal, minimum, mean). The MSL is determined using harmonic component analysis of tidal data. This analysis includes calculating the tidal harmonic constant value using the admiralty method and determining the tide type using Formzahl numbers. The Least Squares method is used to calculate the value. The analysis will produce tidal components that will become variables in the mathematical modeling of the current (Ongkosongo & Suyarso, 1989).

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2.5 | Mathematic two-dimensional model

Two-Dimensional hydrodynamic mathematical model is used to show the physical phenomena movement of ocean currents. DFlow modelling in 3D Delft enables the simulation of physical and chemical variations in coastal/estuarine environments. In this case, it includes wave, tidal, and water elevation changes caused by wind or waves, current flow caused by salinity or temperature gradients, sand and mud (sediment) transport, water quality, and changes in bathymetry/morphology of the beach, among others. Delft3D is also useful for operational purposes, such as forecasting storms, waves, and algae blooms (Deltares, 2011).

3 | RESULT AND DISCUSSION

3.1 | Measurement of water level and bathymetry

Based on the field measurement, the bathymetry in Pekalongan Waters varies between 3–22.7 m (Exhibit 3). The results of the measurements and recording of water elevation indicated that the tidal type was mixed tides that tilt twice daily, with the tide receding twice in a single day. The initial tide's shape was not identical to the shape of the second wave, and it was diurnally inclined (Table S2). Using the least square method, the harmonic analysis calculation yielded the amplitude (A) and phase difference (g°) values. Separating the components of the water level was applied to the measurement results. The results indicated that the astronomical and residual components played a role in forming the water level, respectively; each component was assigned a percentage of dominance: 92.6% for the astronomical component and 7.4% for the residual component.

3.2 | Hydrodynamic modeling

The data from the model are calculated and compared with the data from the measurement results in the waters, the data being tested are currents and tides at the measurement location. Water level elevation data were obtained from tide recording with RBR Virtuose. Current data is measurement data measured using the Acoustic Doppler Current Profiler (ADCP) XR. Hydrodynamic simulation is a simulation in one scenario during the eastern season (June), the validation results will determine the accuracy during measurement (3 × 24 h) using the simulation results in the future.

The data from the model are calculated and compared with the data from the measurement results in the waters, the data being tested are currents and tides at the measurement location. Water level elevation data were obtained from tide recording with RBR.

According to hydrodynamic simulations, the difference in water level between modelling and measurement is 0.0349 m, with a percentage of error of 3.24% for the overall value of data (Figure S1). The error value for the ocean current velocity component revealed differences of up to 0.069 m/s in the east-west component flows. The north-south current velocity component (v-velocity) is 0.019 m/s different. Both of these components were visually compared using a scatter graph and have an error of 18.9% (Figure S2). The validation in this model used the RMS (Root Mean Square) method where RMS is a non-dimensional value to indicate a match for two kinds of data (Evans, 1993; Hutasuhut et al., 2022; Wulandari et al., 2021). These validation values are acceptable and can be used in conjunction with pollutant-tracer simulation. Evans (1993) also explain the tolerance limit for water level is 0,1 m while the speeds to within 0.2 m/s show that the model can be accepted with a small error tolerance. The model data was calculated and compared to the data from the measurement results in the waters; the data being compared are the currents and tides at the measurement location. Determine the location of current measurements using the area sampling method, which was used to determine the sample size when the data source is extremely broad and the sampling area is predetermined (Kowalik & Murty, 1993). Exhibit 4 depicts the current pattern in each condition. The back and forth movement of the current reflects the tidal and tidal conditions in Pekalongan waters. At low tide, the Loji River is seen pushing the outflow from the river body, with a flow velocity of 0.1–0.35 m/s. Banger River, a nearby river, exhibits the same characteristics with a speed of 0.2–0.3 m/s. The fastest currents are found off the coast of Pekalongan, with velocity intervals of 0.5–0.65 m/s.

3.3 | Advection-diffusion of copper (Cu) and chromium (Cr) concentrations

The comparison of modelling and field sampling for Cu and Cr parameters demonstrates the difference in samples taken at the time of measurement; samples were not taken simultaneously, for example, during low tide and ebb tide approaching the post. According to the simulation results, Cr has a minimum-maximum value variation of approximately 0.005 mg/L and 0.1056 mg/L, compared to the spatial distribution results at each sample point, which show a minimum of 0.0009 mg/L and a maximum of 0.091 mg/L. For each parameter, the average error (mean relative error) was 0.2844 mg/L for Cr and 0.35 mg/L for Cu. The spatial distribution pattern of Cu and Cr contaminants in Pekalongan was depicted during the spring tide phase, when the ebb and flood tide conditions exist (Exhibit 5). Cu and Cr distributions follow the current pattern due to flow as a material transport agent contained in water bodies. Cr was concentrated in the mouth areas of the Loji and Banger rivers. These two rivers are the pollutant sources. Flow acts as a transport agent for material contained within bodies of water, ensuring that Cu and Cr distributions follow the current pattern. The current pattern of movement was southeast, east, and some vectors move northeast. The pattern of spatial dispersion was also observed in the concentration distributions of Cu and Cr. Cu and Cr concentrations as shown in Exhibit 2 indicated that Cu concentrations were less than 0.0002 mg/L. Meanwhile, the Cr concentration in the mouth of the Loji river was less than 0.009 mg/L, except for St

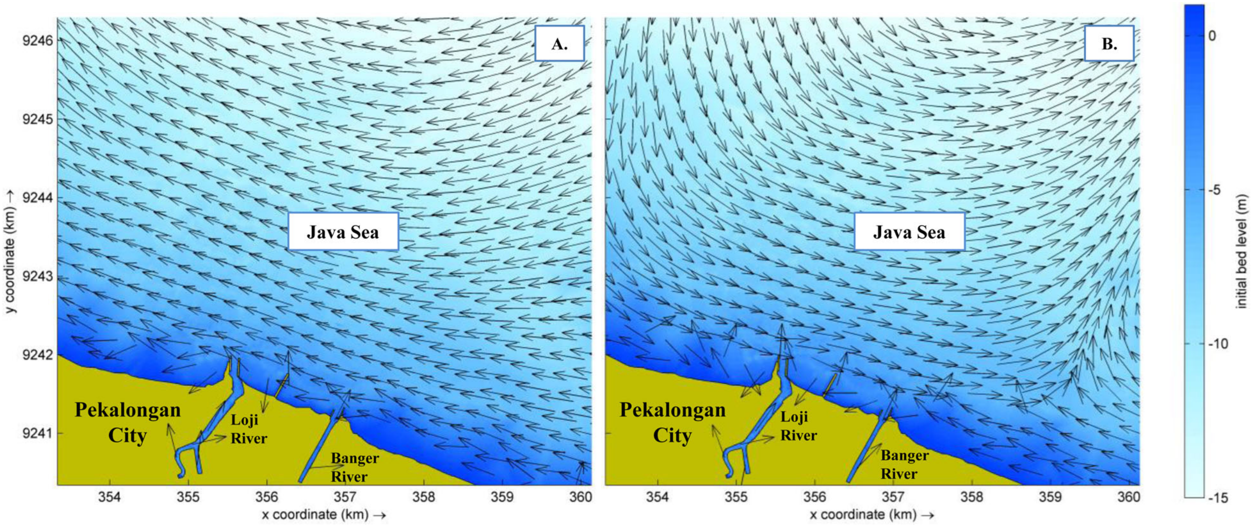

EXHIBIT 4 Simulation of direction of surface current during spring tide in marine environment of Pekalongan: ebb tide conditions (A), flood tide conditions (B). [Color figure can be viewed at wileyonlinelibrary.com]

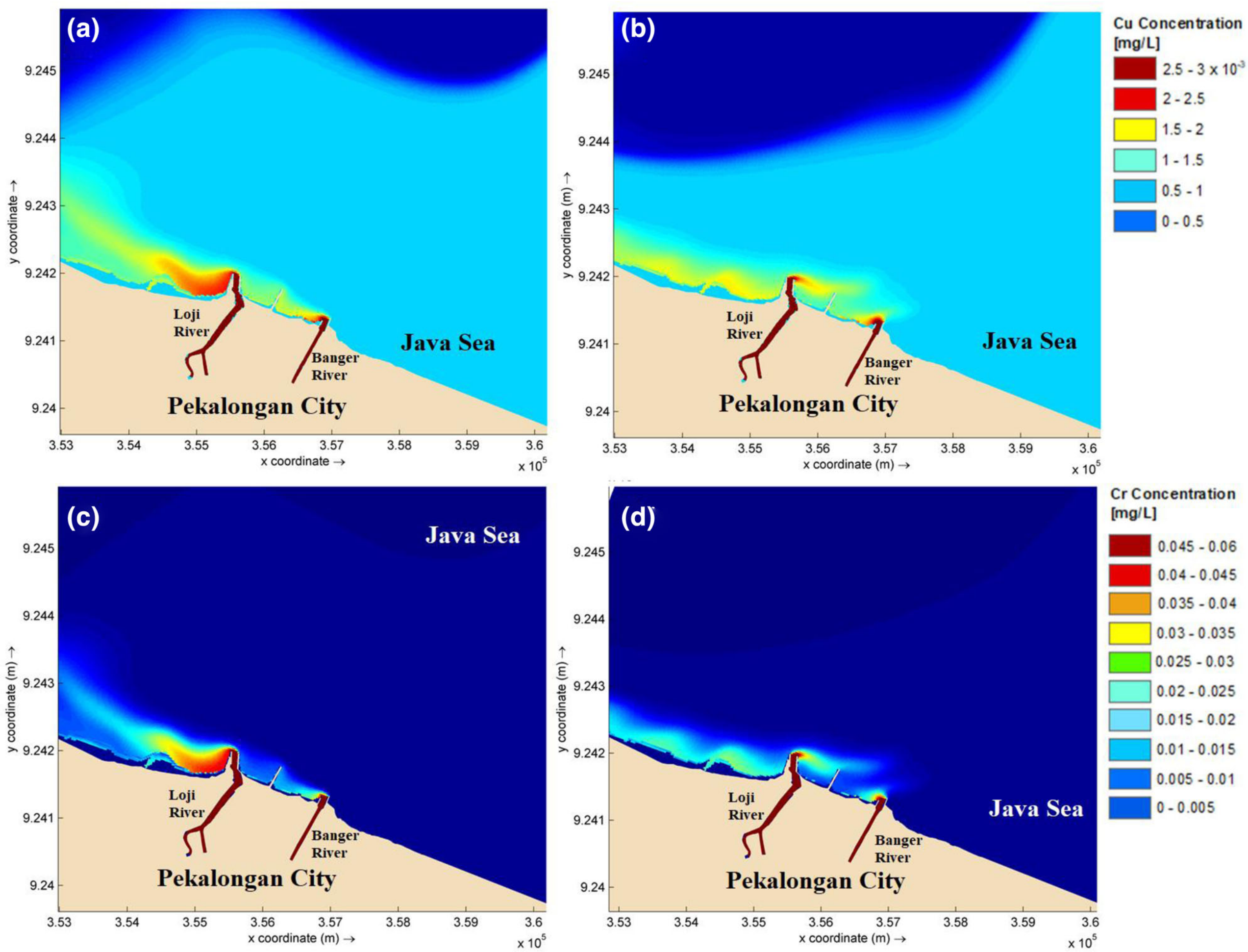

EXHIBIT 5 Spatial distribution maps of the heavy metals in the marine environment of Pekalongan: Concentration distribution of vopper during ebb tide condition (A) and flood tide condition (B); and concentration distribution of chromium during ebb tide condition (C) and flood tide condition (D). [Color figure can be viewed at wileyonlinelibrary.com]

EXHIBIT 6 Cases of heavy metals in some countries

Location	Sample	Cu	Cr	References
Loji and Banger Rivers, Pekalongan, Indonesia	Waters	<0.002 mg/L	<0.009–0.0553 mg/L	This study
Port Phillip Bay,Australia	Waters	$0.47 \pm 0.05\ \mu g/kg$	$0.04 \pm 0.02\ \mu g/kg$	(Fabris et al., 1999)
Trengganu, Malaysia	Species	$56.09 \pm 8.69\ \mu g/g$	$1.34 \pm 0.24\ \mu g/g$	(Rahouma et al., 2013)
Hooghly–Matla estuarine system, India	Sediments	51.8–189 mg/kg	27.7–66 mg/kg	(Ghosh et al., 2019)
Saigon river, Vietnam	Waters	0.92—13.3 $\mu g/L$	0.036—0.849 $\mu g/L$	(Strady et al., 2017)
Turag River, Bangladesh	Waters	0.09 ppm	0.024 ppm	(Afrin et al., 2015)
Taihu Lake, China	Waters	6.844 $\mu g/L$	0.472 $\mu g/L$	(Tao et al., 2012)
USA, Gulf of Mexico	Sediments	35 mg/kg	110 mg/kg	(Vázquez-Botello et al., 2004)
Tugu District Coast, Semarang, Indonesia	Waters	<0.005–4.42 ppm	0.15–11.77 ppm	(Suryono, 2017)
Banjir Kanal Barat, Semarang, Indonesia	Sediments	30.54–55.09 ppm	–	(Maslukah, 2013)
Banjir Kanal Timur, Semarang, Indonesia	Sediments	–	5.786 ppm	(Wulandari, 2012)
Marunda Beach, Jakarta, Indonesia	Sediments		12.79 ppm	(Nurhidayah et al., 2020)
Tanjung Mas Harbor, Semarang, Indonesia	Waters	–	0.14 ppm	(Suryono and Djunaedi, 2017)
Citarum River, Indonesia	Sediments	47.5 mg/kg	8.8 mg/kg	(Yenny et al., 2020)

11 in the upper Banger River, where it was 0.0103 mg/L. The low concentration of Cr downstream (St 7) of the Loji river was due to the shape of the mouth river, causing the concentration of the heavy metal to accumulate prior to the river's mouth (St 5). The highest concentration of Cr was located in the mouth of Banger River (St 6), due to the outflow being openly free and thus accumulating not far from the mouth. According to Indonesian regulations, the copper concentration was within the standard threshold, but the chromium concentration, particularly in the river and mouth river of the Loji River, has exceeded the standard level. This serves as a warning to the Pekalongan City area that it requires additional evaluation in order to control heavy metal contamination. We can compare the Cr data in other countries and find that the majority of the area is less polluted than Pekalongan City. Suryono and Djunaedi (2016), Maslukah (2013), and Wulandari (2012), Nurhidayah et al. (2020), which explain the value of Cu and Cr in Jakarta and Semarang, show that Cr in Semarang ranges from 0.14–11.77 ppm, while Cu in Semarang ranges from 0.005–55.09 ppm. Copper and chromium in Jakarta are shown in Exhibit 6. It is 47.5 ppm for Cu and 8.8—12.79 ppm for Cr. The copper and chromium concentrations at various locations in Exhibit 6 exceed the quality standard established by Indonesia Republic Government Regulation No 22 of 2021. Except for the chromium and copper concentrations in Saigon River, Vietnam (Strady et al., 2017), which are lower than the copper and chromium standards in the Loji and Banger Rivers, these results also show values higher than the average copper and chromium values on the Loji and Banger Rivers. Copper is most valuable in Trengganu, Malaysia (Rahouma et al., 2013). The study's lowest point is at the confluence of the Loji and Banger rivers in Pekalongan, Indonesia. The Gulf of Mexico in the United States of America had the highest chromium concentration (Vázquez-Botello et al., 2004), while the Loji and Banger rivers in Pekalongan, Indonesia, had the lowest. The quality of water contaminated by heavy metals is regulated by Government Regulation No. 22 of 2021 on Environmental Protection and Management, which establishes a quality standard threshold for each designation, including ports, marine tourism, and marine biota. According to Central Java regulation no 13 of 2018 on the Zonation Plan for Coastal Areas and Small Islands, Loji and Banger river estuaries, are designated as a fisheries zone.

4 | CONCLUSION

The distribution of Cr and Cu follows the current pattern due to the tidal conditions, the ebb and flow of the current as a material transport agent contained in water bodies. The residual component dominated the water level by 7.4%, while the astronomical component influenced by 92.6%. Hydrodynamic simulations show a 0.0349 m water level difference between modeling and measurement, with a 3.24% data error. The ocean current velocity component error showed east-west component flows of up to 0.069 m/s. North-south current velocity (v-velocity) differs by 0.019 m/s. A scatter graph showed 18.9% error for these components. Pollutant-tracer simulations can use these validation values. At low tide, the Loji River pushes outflow at 0.1–0.35 m/s. A nearby river, Banger River, has the same characteristics at 0.2—0.3 m/s. The fastest currents out of Pekalongan are 0.5–0.65 m/s. The results of the simulation show that Cr has a minimum-maximum value variation of roughly 0.005 mg/L and 0.1056 mg/L, in contrast to the spatial distribution results at each sample point, which show a minimum of 0.0009 mg/L and a maximum of 0.091 mg/L. The Banger River Estuary along Slamaran Beach had the highest chromium concentration at 0.1056 mg/L. Due to the detection limit of 0.002 mg/L, the Cu concentration in all sampling locations is less than 0.002 mg/L, exceeding the quality standard threshold of Government Regulation No. 22 of 2021 on Environmental Protection and Management, which is 0.005 mg/L for marine biota.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

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SUPPORTING INFORMATION

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