River influx drives heavy metal pollution in Manila Bay, Philippines:

An insight from multivariate analyses

Reymar R. Diwa[1], Custer C. Deocaris[1,2,3] and Lawrence P. Belo[3,4,*]

[1] Philippine Nuclear Research Institute, Department of Science & Technology, Diliman, Quezon City 1101, Philippines

[2] Technological Institute of the Philippines, Cubao, Aurora Blvd., Quezon City, Philippines

[3] BANToxics, Barangay Central, Quezon City, 1100 Philippines

[4] Chemical Engineering Department, De La Salle University, Manila 1004, Philippines

*To Whom Correspondence should be addressed: lawrence.belo@dlsu.edu.ph

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Abstract

Recent work on heavy metal pollution in Manila Bay suggests elevated concentration in the surface sediments. It is critical to identify the sources of these heavy metals to effectively rehabilitate the bay. Our study investigated the sources of the heavy metal pollution that ended up in Manila Bay and the risks associated with these toxic metals based on a recent survey conducted. Surface sediment samples with higher heavy metal concentrations were found in the upper to middle parts of the bay while lower concentrations were in the southeast areas. Multivariate analyses such as hierarchical cluster analysis (HCA), principal component analysis (PCA), and Pearson correlation analysis were used to identify the sources of the heavy metals. The heavy metal pollution in Manila Bay is attributed to several rivers draining northeast of Manila Bay, particularly the Marilao-Meycauayan-Obando River System (MMORS) which is cited as one of the 30 dirtiest river systems in the world. The ecological risks associated with heavy metals in the sediments found higher incidences of toxicity in north and middle parts of Manila Bay. Cu and Cr posed the highest risks of toxicities than any other heavy metals. Based on our analysis, the counterclockwise water gyre of the bay can explain the distribution and ecological

risks associated with the heavy metals as supported by the findings of the PCA. Given the high priority by the Philippine government to rehabilitate the bay, our study strongly shows that efforts to restore the ecological status of Manila Bay will only succeed if the pollution from major rivers draining to it will be properly addressed.

INTRODUCTION

Manila Bay is one of the most important bodies of water in the Philippines due to its cultural, historical, and economic values. Since the pre-Hispanic times, the bay served both as local and international ports opening the Philippines to the world and allowing trade (Jacinto et al., 2006). When the United States unveiled the 1905 Plan of Manila, the Manila Bay served as a centerpiece for the development of post-war Manila with neoclassical public buildings arranged to face the broad boulevards of the bay (Gamboa et al., 2019; Hines, 1972). Apart from its scenic beauty, the Manila Bay area is also the oldest traditional fishing ground in the Philippines. Gifted with abundant natural resources and has been the primary source of livelihood for residents in the bay's coastal areas, it is recognized under the Manila Bay Declaration in 2001 as a source of food, employment, and income for the people, the country's local and international gateway to promote tourism and recreation (Silvestre and Federizon, 1987; Wolanski, 2006).

Manila Bay receives commercial, industrial, and agricultural effluents as well as domestic discharges from approximately 17,000 km^2 of watershed consisting of 26 catchment areas – with main tributaries such as the heavily polluted rivers of Pasig, Bulacan, and Pampanga Rivers (Belo, 2008; Jacinto et al., 2006). As Manila experienced rapid urban population growth rate and industrialization in the past decades, the bay is confronted with several issues on the overexploitation of its resources, coupled with the continued ecological decline of marine habitats. Based on the 2015 Census, Manila's official population has topped more than 1.78 million within its 42.88 km^2 area, making the city the most population-dense (41,515 persons per km^2) in the world. Manila is much more populated than Baghdad, Iran (32,874 persons per km^2), and Mumbai, India (32,303 persons per km^2). Being the closest to the city Manila, the bay is now one of the pollution hotspots in the Seas of East Asia under the Regional

Programme on Building Partnerships in Environmental Management for the Seas of East Asia (Sta. Maria et al., 2009).

Heavy metal pollution in aquatic systems is associated with the rapid economic development of cities. It has increased considerably due to the inputs of industrial waste, sewage runoff, and agriculture discharges. Pollution by heavy metals is constantly rising, producing a severe toxic effect on all forms of living organisms (Belo et al., 2018; Cadondon et al., 2020; Hoang et al., 2020). When the trend of heavy metal contamination in rivers and lakes around the world from 1970 to 2017 was analyzed by Li et al., (2020), the group reported an increasing trend for Fe, Mn, Ni, Cu, Cr, and Cd and a decreasing trend for Zn and Pb, with the mean dissolved concentration higher in Asia than in Europe. Different regions had different heavy metal contamination sources, with mining and manufacturing sectors being critical sources of heavy metal pollution in the same time frame (1970 – 2017). There is mounting evidence of heavy metal enrichment in Manila bay's sediments, with concentrations of toxic metals reaching alarming levels (Prudente et al., 1994). Su *et al.* reported some heavy metals – particularly total Cd, total Pb, and total Cr – are present in considerable amounts in the waters, fish, and macro-invertebrates for both wet and dry seasons (Su et al., 2009). The pollutants have been documented to affect gonadal development and induced histological lesions in the digestive glands and gut in *Perna viridis* (green mussels), a pollution bioindicator (Mamon et al., 2016), although the recent report by Nacua *et al.* failed to confirm these severe deformities (Nacua et al., 2019). While the "pollution archives" with ^{210}Pb dating validates that Manila Bay receives significant discharges of domestic and industrial wastes (Sta. Maria et al., 2009), no results have been reported on the status of heavy metal pollution and its contribution to ecological risk.

In the last two decades, pollution in Manila Bay has gained significant attention from both the academe and the regulatory bodies in the Philippines, thus leading to the establishment of the Integrated Environmental Monitoring Program for Manila Bay (IEMP-MB). As a result, heavy public and private investments have been allotted to address the pollution and resource degradation of Manila Bay. The national government has recently organized clean ups, required all discharge waters to undergo treatment through sewage plants, provided resettlement to informal settlers in the esteros draining to Pasig River (which is one of the major water bodies draining to Manila Bay), and authorized the

controversial "beach nourishment: on a 300 m beach of the bay by reclamation using dolomite sands costing roughly USD 580,000 (Cahiles, 2020; Castelo, 2019; Rafales, 2020). Still, very little has been done to address the pollution triggered by major rivers outside of Metro Manila that also drain to the bay. Despite several studies describing the concentration of heavy metals in the surface sediments of Manila Bay, to the best of the authors' knowledge, no research has yet identified the sources of these toxic metals. To address this critical knowledge gap, we aimed not only to identify the sources of heavy metals but also to assess their apportionment and their effects on key ecological risk indices. Specifically, the objectives of this study are (1) to identify the various sources of metal pollution in Manila Bay, including their contributions and apportionment using multivariate analyses, (2) to assess the ecological risks associated with heavy metals in the sediments, and (3) to determine the key factor that controls the distribution of the heavy metals in the surface of Manila Bay. This study is expected to help provide a stronger basis for environmental policies associated with the rehabilitation and protection of the ecological environment in this significant body of water in Southeast Asia.

METHODOLOGY

Study Area

Manila Bay is a semi-enclosed estuary situated in the western part of Luzon between 14.23° and 14.87° N and 120.53° and 121.03°E. Connected to the West Philippine Sea and the larger South China Sea through a 16.7-km-wide entrance, the bay is bounded by Cavite and Metro Manila on the east, Bulacan and Pampanga on the north, and Bataan on the west and northwest (**Figure 1**). It has a surface area of 1,800 km^2, a mean depth of 25 m, and an estimated volume of 31 km^3. It is located adjacent to the capital of the Philippines, Manila. This natural harbor is the country's major hub and international gateway that holds a tremendous social, economic, and political importance as it facilitates commerce and trade between the Philippines and the neighboring countries. With a coastline length equivalent to 190 km which is like that of the entire Singapore, Manila Bay serves as a drain to several prominent rivers like the Pasig River, Pampanga River, Angat River, and Marilao-Meycauayan-Obando River System (MMORS).

The wind blows at specific periods of the year control the gyres of Manila Bay. There are northeasterly winds from October to January, southeasterly winds from February to May, and southwesterly winds from June to September (De Las Alas and Sodusta, 1985; Villanoy and Martin, 1997). Villanoy and Martin (1997) proposed that the water current of Manila Bay is being controlled by a combination of wind and tide. However, due to the absence of a circulation model that combines the effects of both the wind and tide during the time of sampling, this study used the circulation model of a study by De Las Alas & Sodusta (1985).

Sediment Heavy Metal Data

To identify the sources of pollutants in the surface of Manila Bay, we used the datasets on heavy metal concentrations in sediments of Manila Bay. From 10-11 February 2005, during the dry season, IEMP-MB collected sediment samples on the surface of Manila Bay in nine (9) locations (**Figure 1**). The elemental compositions of the samples were analyzed using energy-dispersive X-ray Fluorescence (XRF) at the Philippine Nuclear Research Institute (Diliman, Quezon City, Philippines) using secondary targets Ag and Fe in radioisotope excited XRF using [241]Am source (Olivares et al., 2019). **Table 1** provides a summary of the concentrations of the 9 heavy metals in the surface sediments of Manila Bay.

Statistical Analysis

The statistical software R ver. 4.0.4 was used to perform multivariate analysis. Hierarchical cluster analysis (HCA), principal component analysis (PCA), and Pearson correlation analysis (CA) were used to identify the sources of heavy metal pollutants in Manila Bay. HCA is an algorithm that groups similar objects into clusters based on proximity measures and hierarchically arranges a sequence of partitions for a data set (Köhn and Hubert, 2015). PCA is used to reduce the dimensionality of the data to a new set of variables with minimal loss of information (Jolliffe, 2002). CA is a measure of strength of linear correlation between two sets of variables. The combination of these multivariate analyses has been found useful to identify the sources of heavy metals in the environment by grouping them according to similar sources (Buttafuoco et al., 2010; Guagliardi et al., 2012; Weissmannova et al., 2019). The *p*-values were used to evaluate the validity of the CA and PCA. The calculation of distance between the

elements for the clustering by HCA was achieved by using the Canberra method. The algorithm used to link the clusters was the Complete method. Correlation coefficients $r > 0.700$ were deemed strong correlations for this study.

Risk Assessments

The ecological risks of the heavy metals in the sediments were assessed using Sediment Quality Guidelines (SQG) and Marine Sediment Pollution Index (MSPI). The quotients of the concentrations of the individual heavy metals to the concentrations in the SQG were derived by using the Threshold Effect Level (TEL) developed for sediments in Florida coastal waters (MacDonald et al., 2000; Macdonald et al., 1996). The mean of the quotients was used to represent the sediment quality of the sampling locations.

The MSPI of the sediments was derived using the procedure developed by Shin and Lam (2001) in deriving the MSPI of the marine sediments surrounding Hong Kong (Shin and Lam, 2001). Out of the 22 elements analyzed by Olivares *et al.* (2020) in the samples, sixteen elements (*i.e.*, Al, Br, Ca, Cl, Cr, Cu, Fe, K, Mg, Na, Pb, Rb, Si, S, Sr, and Ti) were selected to reflect the sediment pollution based on the absolute values of the correlation between the principal components and variables (> 0.700) as suggested by Comrey and Lee (Comrey and Lee, 1992). MSPI was calculated using the equation:

$$MSPI = (\textstyle\sum q_i w_i)^2 / 100,$$

where q_i is the sediment quality rating of the ith element and w_i is the weight attributed to the ith element. For each of the sixteen elements, the sediment quality rating was based on comparison to the percentile ranging from 10-100 in the dataset (*e.g.*, a rating of 10 was given if the concentration of the element falls between the 0-10 percentile). The weight attributed to each of the elements was calculated using normalized eigenvalues of the principal components where the high correlation values of the elements were distinguished. The MSPI ratings reflect the extent of sediment pollution in the surface of Manila Bay. MSPI is then rated from 0 to 100 with the following qualitative ratings: 0-20 for 'excellent'; 20-40 for 'good'; 40-60 for 'moderate'; 60-80 for 'poor'; and 80-100 for 'bad'.

Spatial mapping

The mean incidences of toxicity and MSPI were spatially interpolated using the Kriging method of Surfer® 11.1.719 (Golden Software, LLC). The color-coding scheme of the MSPI was defined based on the qualitative description of MSPI ratings.

RESULTS AND DISCUSSION

Heavy metal pollution relative to other bodies of water

The heavy metal concentrations in surface sediments of Manila Bay (Olivares et al., 2019) were compared to other coastal bodies in East and Southeast Asia and in the Philippines as shown in Table 1. The mean Cr and Cu in Manila Bay are 1.5 and 1.4 times the mean in East and Southeast Asian marine sediments. Most of the mean of the heavy metals are also less than the mean heavy metal concentrations in marine sediments of the Philippines except for Pb and Zn. These heavy metals are equivalent to 1.6 and 1.2 times of the Philippine average, respectively.

Source identification of heavy metals

Multivariate analyses are useful to discriminate the contributors of heavy metal pollution in the natural environment by grouping them according to comparable sources (Buttafuoco et al., 2010; Guagliardi et al., 2012). HCA, PCA, and CA were performed in this study to identify the sources of pollution in the surface sediments of Manila Bay.

The dendrogram that resulted from HCA analysis shows the three primary clusters of sampling locations as shown in **Figure 2a**. The southeastern part of Manila Bay (locations 6 and 7) forms the first cluster; the central and southwest parts (locations 4, 5, 8, and 9), the second cluster; and the northern part of the bay (locations 1, 2, and 3), the third cluster. To identify the sources of the heavy metals, we again used the HCA to identify the clustering of metals by their sources (**Figure 2b**). The metals are clustered into two primary groups. The first cluster consists of Na, Cl, Si, and Fe which are associated with seawater

composition and terrestrial sources (Hans Wedepohl, 1995; Millero et al., 2008). The rest of the metals such as Ca, Mg, Sr, Br, Rb, Mo, Y, S, and K as well as the heavy metals such Al, Mn, Pb, Ni, Cu, Cr, Zn, and Ti are gathered together to form the second cluster which can be associated with industrial and terrestrial sources (Alloway, 2013; Hans Wedepohl, 1995).

The PCA approach further identifies the sources of heavy metal pollutants. Eight dimensions or principal components were identified by the analysis. Seven of the principal components have eigenvalues greater than 1.0 % and these seven explain 99.1 % of the total variance in the dataset. The relations among the elements based on the first three principal components that represent seawater composition, terrestrial sources, and industrial sources, respectively, is shown in **Figure 2c**. The elements roughly cluster into three groups indicating similarity in distribution patterns and sources. The first group consists of Na, Cl, S, and Br which are associated with the seawater composition. The second group incorporates Al, Ca, Fe, K, Mg, Si, Sr, and Ti which are related to terrestrial sources. The elements, i.e. Cr, Ni, Zn, Cu, Mn, Pb, Mo, Y, and Zr including Rb, are in close proximity with each other forming the third cluster. The latter cluster appears to be associated with industrial sources, such as tanneries, Pb-acid battery recycling, gold smelting, jewelry refining, agro-based industries, pyrotechnics, and electroplating (Johnson et al., 2006; Koch, 2004; McMurtry et al., 1995; Sun et al., 2017; Vivas et al., 2019). **Supplementary Table 5** summarizes the eigenvalues, proportion of variance, factor loadings, and elemental contribution of the principal components identified by the PCA. **Supplementary Table 6** summarizes the contribution of each principal component to the sampling locations.

Our Pearson Correlation analysis supports the findings of the PCA. The high positive correlation ($r >$ 0.700) found between the metals indicates similarity in sources. A high correlation was found between Na and the individual Cl, S, and Br (r 0.77 – 0.94) as shown by **Supplementary Table 7**. The terrestrial components showed very high correlation to one another (r 0.70 – 0.94). Fe showed high correlation with the terrestrial components such as Al (r 0.94), Si (r 0.93), K (r 0.85), and Ti (r 0.93) indicating that most of Fe in the surface sediments of Manila Bay is terrestrial in origin. Ti showed high correlation

with K (r 0.77). Zr also showed high correlation to terrestrial components such as Al (r 0.77), Fe (r 0.84), and Ti (r 0.77).

Ecological risks associated with heavy metal pollution

Finally, the ecological risks associated with the heavy metals in Manila Bay were assessed. The Threshold Effect Level (TEL) in sediments of marine and coastal waters is the concentration below which harmful effects are unlikely to be observed (MacDonald *et al.*, 2000). The contributions of the individual heavy metals to the TEL were in the order of Cu (50.8 %) > Cr (20.3 %) > Ni (12.2 %) > Zn (9.9 %) > Pb (6.8 %) as shown in **Figure 3a**. The $Q_{m\text{-}TEL}$ of all the samples are greater than 1.0, indicating that the sediments are polluted by heavy metals. The mean threshold effect level quotient ($Q_{m\text{-}TEL}$) of the sampling locations is presented in **Figure 3b**.

The guidelines also project the incidences of toxicity caused by heavy metal pollution by direct comparison with TEL and PEL values. The Probable Effect Level (PEL) in sediments of marine and coastal waters are the concentrations above which harmful biological effects are likely to be observed. **Supplementary Table 2** shows the incidences of toxicity of the individual heavy metals in each of the sampling locations. Sampling locations 1, 2, 3, 4, 5, and 11 have mean incidences of toxicity of 11.1%; locations 6 and 8 have 10.0% mean incidences of toxicity; and location 7 has 7.7% mean incidence of toxicity. For individual metals, Cu and Cr contributed most to the toxicity of the surface sediments.

Sixteen elements such as Al, Br, Ca, Cl, Cr, Cu, Fe, K, Mg, Na, Pb, Rb, Si, S, Sr, and Ti were used for the derivation of MSPI. The absolute value of the factor loadings of these elements is greater than 0.700 based on the results of the PCA. **Supplementary Tables 3** and **4** summarize the sediment quality rating and weight of each of the elements, respectively. MSPI is rated from 0 to 100 with the following qualitative ratings: 0-20 'excellent'; 20-40 'good'; 40-60 'moderate'; 60-80 'poor'; and 80-100 'bad'. According to the MSPI ratings **(Figure 3c)**, location 4 has 'excellent' sediment quality and all other locations have 'good' sediment qualities.

From our analysis, it is noteworthy that despite lower MSPI values, sampling locations at the north, middle, and west of Manila Bay have higher incidences of toxicity. Sampling sites with lower incidences of toxicity southeast of the bay such as at locations 6 and 7 have higher MSPI ratings as shown in **Figure 4**. With this trend, similarities in the sources of the heavy metals with the first and second principal components associated with terrestrial and industrial sources, respectively can be deduced.

Source apportionment of heavy metal pollution shows contribution from highly pollutive tributaries

The distribution of the heavy metals in sediments can be attributed to several rivers draining northeast of Manila Bay, particularly, the Marilao-Meycauayan-Obando River System (MMORS). Meycauayan River, for instance, was listed as one of the 30 most polluted rivers in the world (Blacksmith Institute, 2007). The stretch of MMORS along the municipalities of Marilao, Meycauayan, and Obando is densely-populated and is a major hub of industries like tanneries, Pb-acid battery recycling, agro-based industries, gold smelting, jewelry refining, and backyard pyrotechnics that reportedly discharge their untreated wastewaters to the rivers (Vivas et al., 2019).

Interestingly, the low correlation between the heavy metals indicates that there may be unique sources of these pollutants. These heavy metals are attributed to several and diverse industries operating along the stretch of MMORS. For example, the influx of Cr is most likely due to the paint and leather tanning industries (Johnson et al., 2006). Pb is associated with Pb-acid battery recycling and municipal wastes which are concentrated in Brgy. Banga and Brgy Calvario at Meycauyan, Bulacan (Diwa et al., 2021; Sun et al., 2017). Ni was probably sourced from electroplating industries but can also be associated with agricultural activities (McMurtry et al., 1995). Olivares *et al.* (2019) proposed that Ni could be lithogenic or terrestrial in origin since Ni concentrations did not exceed the criteria values. However, in our multivariate analyses, the clustering of Ni with other heavy metals proved otherwise. Like Ni, Zn can also be sourced from electroplating industries and can be associated with battery recycling, agricultural activities, and municipal wastes (Araújo et al., 2017). Cu is often present in car lubricants (Al-Khashman, 2007). Of the minor pollutants observed, we attribute Rb in the cluster of the major

heavy metals to pyrotechnic industries that are abundant in Meycauayan and in the neighboring towns in Bulacan, *e.g.* Bocaue (Koch, 2004).

Unexpectedly, the effect of heavy metal pollution by other prominent rivers draining to Manila Bay has been invalidated. For instance, the heavy metal toxicities in locations 3 and 9 could be easily attributed to influx from Pampanga River and Pasig River, respectively. But this has been discredited by the PCA that shows the low apportionment of the principal component 2 (or industrial sources) to these locations. This has been supported by the results of Samar *et al.* (2013, unpublished) that identified low heavy metal concentrations in the water near the mouth of Pampanga River. Meanwhile, heavy metals were detected in higher concentration in Pasig River during the dry season from October to May. But these toxic metals may not have reached Manila Bay at the time of sampling due to possible backflow along Pasig River, that is, saline water from Manila Bay flows into the Pasig River towards the end of the dry season (Paronda et al., 2019).

Water gyre phenomenon drives the spatial distribution of ecological toxicity risks

We discovered that the spatial variation of heavy metal toxicity and MSPI vary significantly depending on the three wind blow patterns occurring at different times of the year that control the water gyres of Manila Bay (**Figure 4**). Consistently, the water gyre at the time of sampling was controlled by the southeasterly winds that occur during February to May of the year (De Las Alas and Sodusta, 1985).

The heavy metal influx from MMORS reaches the northeast of Manila Bay in location 1 and spreads to the west and east sides through the counterclockwise water gyre occurring on upper-middle parts of the bay traversing locations 1, 2, 3, 4, 5, 8, and 9. This mechanism is supported by the PCA which shows that the principal component 2 contributes most of the heavy metals like Cr and Cu which are deposited mainly in locations 1, 2, and 5. This mechanism, however, conflicts with the observed lower toxicities but higher MSPI in the southeast of the bay despite being involved in the gyre. A plausible explanation to this is that this part of the bay is being supplied with nutrients, as well as sediments, from the West Philippine Sea (Pokavanich and Nadaoka, 2006). The prevailing longshore current carries and deposits terrestrial, low toxicity sediments to these locations. This hypothesis is strongly supported by our PCA

result which confirms that the principal component 1 contributes elements of terrestrial origin such as Si, Al, and Fe that are deposited in locations 6 and 7. Moreover, the high correlation between the terrestrial elements indicates a common source, supporting the idea that these elements must have been sourced from the West Philippine Sea.

An evident relationship between the sampling locations supporting the relationship between the water gyre and the associated ecological risks and MSPI is shown by the clustering of sampling locations. The cluster of locations 1, 2, and 3 indicate the influx of heavy metal pollutants since these locations are close to where MMORS drains. The cluster of locations 4, 5, 8, and 9 receives heavy metals that were originally discharged to the first cluster through the water gyre. This transport effect appears to be limited in locations 6 and 7 because these locations are more influenced by the incoming current from the West Philippine Sea and are therefore unaffected by the heavy metal influx in locations 1, 2, and 3.

Implications for Manila Bay clean-up

In January 2019, the Department of Environment and Natural Resources (DENR) formally started the rehabilitation program of Manila Bay. This aims to restore the water quality of Manila Bay in accordance with the *Writ of Continuing Mandamus* issued by the Supreme Court of the Philippines in 2016 that ordered several government agencies to restore the bay to make it fit for swimming and for other recreational activities. This program which includes cleanup, water quality improvement, resettlement of informal settlers, and education has an estimated total funding of PhP 47 billion (est. 970 M USD) (CNN Philippines, 2019).

DENR ordered the strict implementation of Republic Act 9275 or the Philippine Clean Water Act of 2004 and warned against suspensions or total closure of the industries that pollute the bay. In July 2019, the first of the four planned sewage treatment plants opened. The cleanup removed 3810 tons of debris in Manila bay coastline and drainage systems draining to Manila Bay by August 2019. In November 2019, around 70,000 informal settler families from Metro Manila and neighboring provinces were relocated. And by September 2020, the government filled the 300 m coastline in Manila with crushed dolomite for beach nourishment (Cahiles, 2020; Gamboa, 2020).

The findings of our study suggest that the government through the Department of Environment and Natural Resources (DENR), Metropolitan Manila Development Authority (MMDA), and local governments may also need to adopt stricter policies to address pollution caused by rivers outside of Metro Manila that drain to the Manila Bay. The following are some key recommendations that the local and national government may consider addressing the heavy metal pollution in Manila Bay:

- We showed in this study that MMORS is the source of heavy metal pollution in Manila Bay. The effect of Pasig River, however, remains to be unclear due to possible backflow at the time of sampling (Paronda et al., 2019). It is recommendable for the government to address the heavy metal pollution by industrial discharge in Bulacan, Philippines such as tanneries, Pb-acid battery recycling, gold smelting, jewelry refining, agro-based industries, pyrotechnics, and electroplating to effectively rehabilitate the bay.

- Since Cr and Cu are the two heavy metals that have the highest incidences of toxicity in Manila Bay, the government should adopt stricter policies to industries contributing these pollutants in Meycauayan, Bulacan, the tannery capital of the Philippines.

- Government prioritization of more challenging issues for Manila Bay rehabilitation like heavy metal pollution. Heavy metal pollution in aquatic systems is one of the most challenging pollution issues due to the toxicity, abundance, persistence, and subsequent bioaccumulation of heavy metals (Barlas et al., 2005). Manila Bay can never be completely safe for swimming and for other recreational activities if the heavy metals remain in the water and sediments in concentrations considered unsafe. Projects to improve the appearance of the bay like reclamation by dolomite sands can come after.

- Replanting of mangroves, particularly species with phytoremediation abilities, in the coastal areas of Bulacan to filter the polluted water and create a natural "engineering barrier" to prevent the heavy metals from reaching the bay and be circulated by the water gyre to other parts of Manila Bay.

- Despite being an important source for marine produce for consumers in Metro Manila and neighboring provinces, the effect of heavy metal toxicity to marine organisms in Manila Bay

remains to be poorly understood. The government is also recommended to fund research projects to study the incidence of toxicity in sediment-dwelling organisms found at the surface of Manila Bay to provide an empirical basis of the heavy metal toxicities discussed in this study (MacDonald et al., 2000; Macdonald et al., 1996).

In this study, the relationship of other water gyres occurring at different times of the year to the risk indices was not considered. There was also no empirical data on the water gyre conditions of the bay at the time of sampling. We recommend studying the spatial distribution of heavy metals and their associated toxicities for other seasons with wind patterns producing distinct water gyres such as northeasterly winds from October to January and southwesterly winds from June to September. The incidence of toxicities associated with heavy metals in this study is also based on comparison with the consensus-based SQG values. Studies about the toxicity effects of heavy metals to sediment-dwelling organisms in Manila Bay are therefore recommendable.

CONCLUSIONS

The elevated concentration of heavy metals found in the surface sediments of Manila Bay is attributed to heavy metal influx from MMORS. MMORS is listed as one of the 30 most polluted river systems in the world on the account of haphazard waste discharge from industries like tanneries, Pb-acid battery recycling, gold smelting, pyrotechnics, etc. Despite having generally low mean incidences of toxicity, these heavy metals may have potential ecological risks as suggested by the SQG. The counterclockwise water gyre prevailing in the bay at the time of sampling spreads the heavy metals coming from MMORS to other parts of the bay. Meanwhile, the sediment pollution in the southeastern part of Manila Bay appears to be controlled by the longshore current from the West Philippine Sea that carries low toxicity metals of terrestrial origin. It has been widely communicated that one of the main objectives of the ongoing rehabilitation of Manila Bay is to decrease the amount of heavy metals. The results of this study suggest that understanding of the heavy metal sources is a very critical knowledge to effectively rehabilitate Manila Bay. Rehabilitation efforts should therefore not be limited to addressing pollution

in the bay but also in the major rivers draining into it, particularly, the Marilao-Meycauayan-Obando River System.

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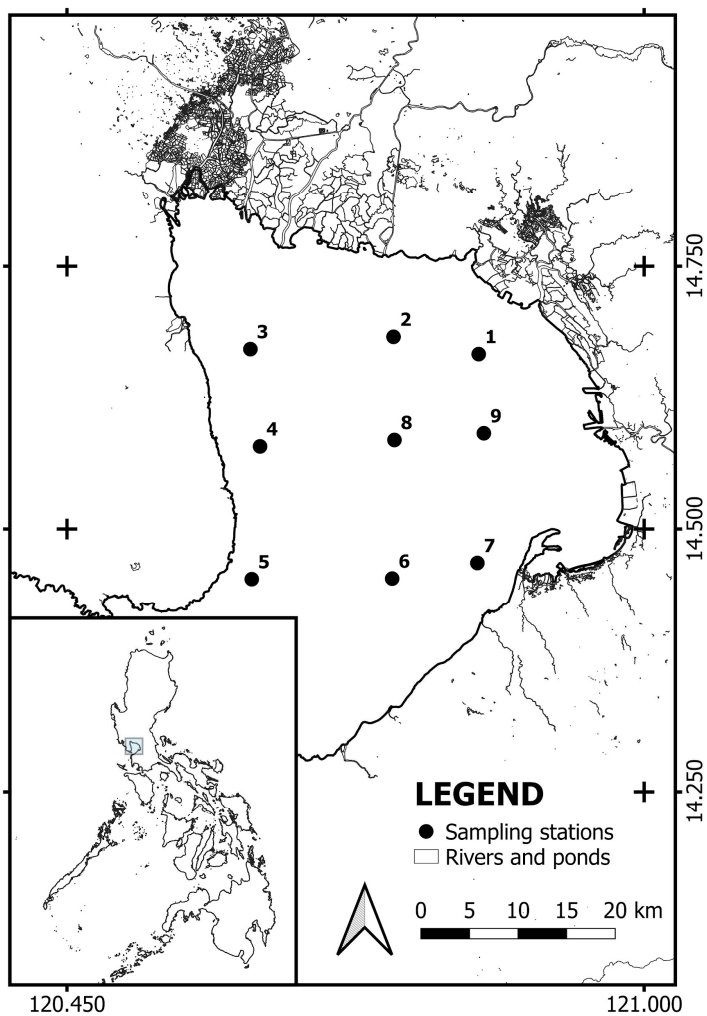
Figure 1. Location map of the nine samples collected from the surface of Manila Bay
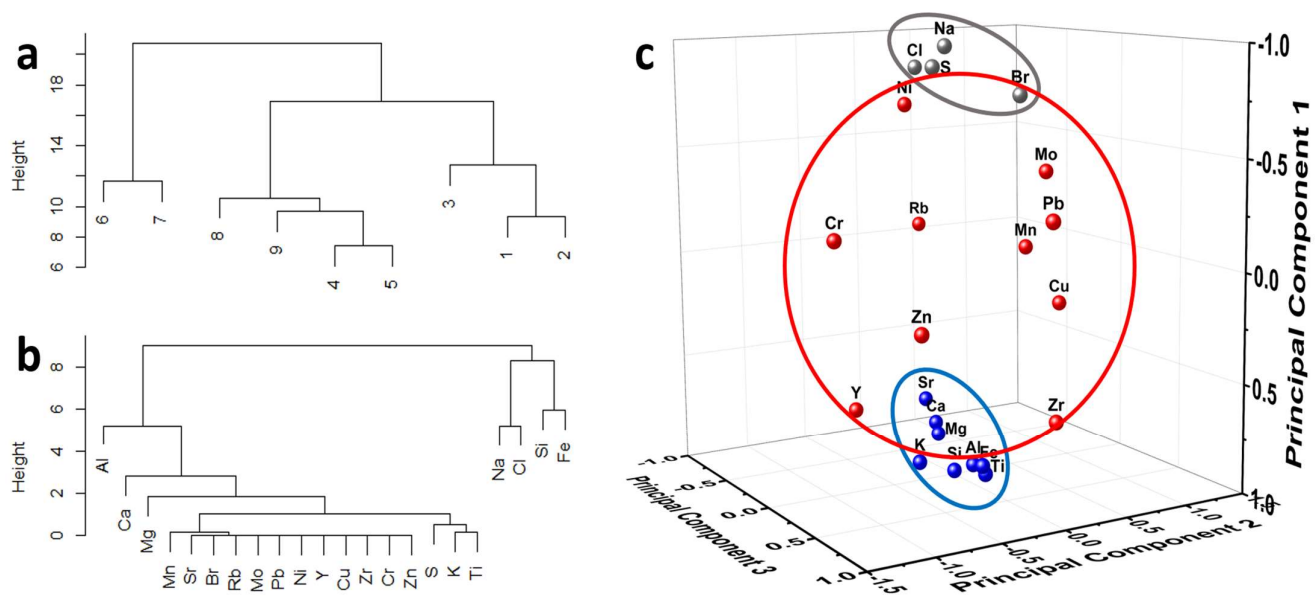
Figure 2. Hierarchical dendrogram of (a) sampling locations and (b) elements in surface sediments of Manila Bay show clustering according to similarity in sources. The 3D plot of (c) factor loadings by the PCA further shows the clusters of metals according to source: gray is seawater composition, blue is terrestrial, and red is industrial.
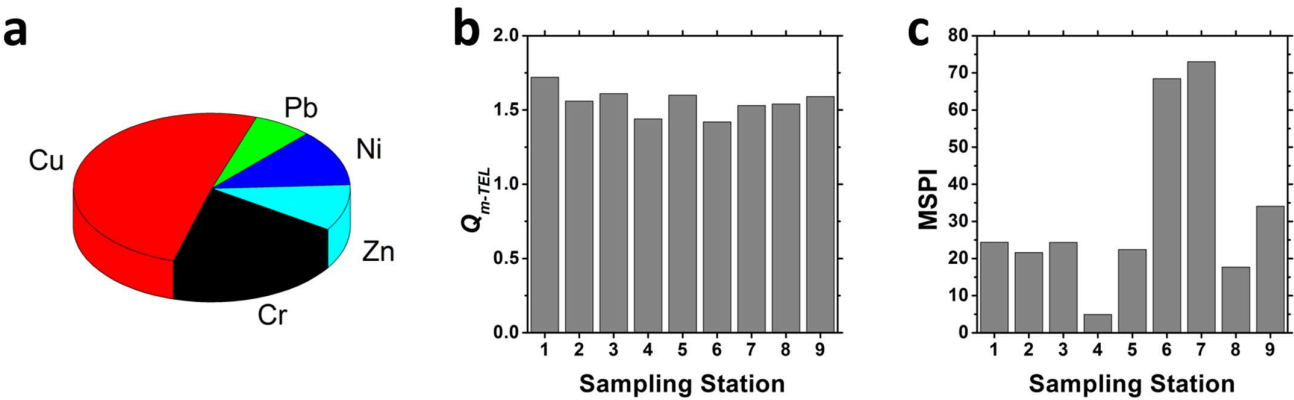
Figure 3. Plots of (a) percent contribution of the heavy metals to TEL, (b) mean TEL of sampling locations, and (c) MSPI of sampling locations in Manila Bay. All samples are polluted with respect to heavy metals with possible risk to ecology. The MSPI shows that Location 4 has excellent sediment quality (MSPI 0-20) whereas other locations have good sediment qualities (MSPI 21-40).
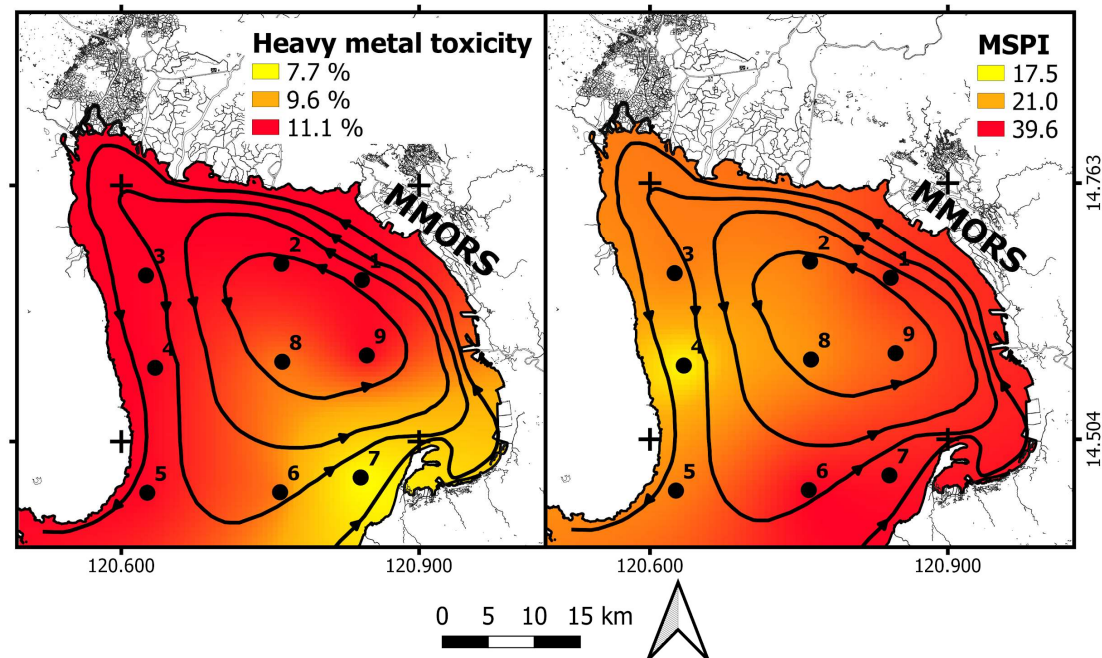
Figure 4. *Maps of (left) mean incidence of toxicity and (right) MSPI of surface sediments of Manila Bay. The heavy metals found in the sediments are attributed to the industrial influx from MMORS, located northeast of Manila Bay, which is cited as one of the most polluted river systems in the world. The heavy metal influx from MMORS is delivered to other parts of the bay such as 2, 3, 4, 5, 8, and 9 through the water gyre that is controlled by the wind at the time of sampling (De Las Alas and Sodusta, 1985). This explains the high incidence of toxicities associated with the heavy metals in the north, middle, and west parts of the bay. The southeast part such as locations 6 and 7, however, is affected by the longshore current that delivers low toxicity metals of terrestrial origin from the West Philippine Sea.*

TABLES:

Table 1. Summary of heavy metal composition of surface sediment samples from nine locations in Manila Bay (Olivares *et al.*, 2019).

Element	Marine sediments in E and SE Asia*		Marine sediments in the Philippines**		This study	
	Mean	Range	Mean	Range	Mean	Range
Al	-	-	-	-	31811	16000-68200
Ti	-	-	-	-	3285	1780-5410
Cr	54.0	0.13-1.50	216.67	162.44-269.89	82.7	49.9-139
Mn	-	-	-	-	1130	841-1810
Fe	-	-	45800	14800-91100	44244	37200-56600
Ni	25.7	1.00-37.4	58.07	19.10-290.11	15.1	9.92-18.7
Cu	53.2	3.00-148	191.60	2.05-1074	73.9	56.7-90.3
Zn	155	4.00-595	77.13	1.91-144	96.0	74.6-124
Pb	35.4	1.00-111	10.08	0.54-62.27	16.0	8.69-26.6

* Fang and Yang (2011)
** Lo and Sakamoto (2005); Marges et al. (2011); Elvira et al. (2016); Badong and Bersabal (2018); Dacera et al. (2018); Nillos et al. (2020)