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Baseline

Influence of urban runoff, inappropriate waste disposal practices and World War II on the heavy metal status of sediments in the southern half of Saipan Lagoon, Saipan, CNMI

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ABSTRACT

Heavy metals were examined in sediments from the southern half of Saipan Lagoon. These waters provided tactical access for US troops during WWII and were heavily shelled at the time. Mercury profiles in sediments were, to some extent, reflective of this event. Samples from the southern end of the lagoon, where an old post-war dumpsite once existed, were found to be substantially enriched with Pb, Cu and Zn. Further north, the lagoon was primarily impacted by urban runoff. Metal enrichment in sediments from this region was generally highest at storm drain outlets and attenuated seawards. Moderate enrichment was rarely exceeded for any element other than Hg beyond the 50 m mark. Sediment quality guidelines used to flag potentially adverse ecological health effects revealed no PEL exceedances. TEL exceedances for Pb and Cu were identified in sediments near the former dumpsite. The public health implications of the data are briefly addressed.

Saipan is the largest and most densely populated island in the Commonwealth of the Northern Mariana Islands (CNMI) and is located approximately 118 nautical miles NNE of Guam. The current population stands at around 48,000 (Eugenio, 2012). While the economy benefits substantially from external funding from the U.S. Government (via grants, contracts and assistance programs, etc.), internal revenues are generated primarily from tourism. The eastern side of Saipan is composed primarily of rugged, rocky cliffs, whereas the western side is bounded by a large lagoon that extends almost the entire length of the island. Locally referred to as Saipan Lagoon, this body of water harbors a rich diversity of marine life and is one of the island's major tourist attractions. The lagoon is also of great cultural significance to local residents, many of whom favor its waters for subsistence fishing as well as aesthetic enjoyment. The sustainable management of this environmentally sensitive region is, therefore, of critical socioeconomic importance to the people of Saipan and central to their traditional beliefs and cultural ties to coral reefs and the ocean.

Prior to WWII, anthropogenic impacts on the lagoon were largely reflective of land clearing efforts for housing and agriculture. These activities accelerated natural soil erosion processes and increased sedimentation rates and nutrient enrichment in nearshore waters. Saipan's post-war emphasis on tourism and trade has seen parallel growths in housing, commercial developments and light industry, which in turn has greatly increased the range of potential contaminants entering the lagoon. This is especially true in the central region of the lagoon, which borders the most industrialized coastline on island. Earlier monitoring efforts in this section focused on some of the more recalcitrant chemical groups likely to be released into the environment (Denton et al., 2001) and identified heavy metals as contaminants of primary concern in sediments and biota (Denton et al., 2006, 2009, 2010, 2011a,b).

Waters further south in the lagoon are primarily impacted by stormwater runoff from commercial and residential premises, highways and unpaved roads (Bearden et al., 2010). The runoff, which for the most part flows from properties within 0.5 km of the coast, is channeled into the lagoon via a system of swales, culverts and storm drains. While increased sedimentation rates and nutrient enrichment in adjacent nearshore waters remain areas of concern (Bearden et al., 2010) and have reportedly caused some ecological restructuring in places (Houk and Van Woesik, 2008; Houk and Camacho, 2010), relatively little is known regarding the distribution and abundance of other likely contaminants mobilized into these waters, particularly those that accumulate in sediments and biota, and degrade the edible quality of fisheries

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resources harvested for food. Heavy metals certainly fall into this category and are typically elevated in urban and highway runoff (USEPA, 1983; Makepeace et al., 1995). Recent work has also shown that stormwater discharges emptying into the southern half of Saipan Lagoon are no exception in this regard (Environet Inc., 2007). Several other potential sources of heavy metal contamination also exist along this stretch of coastline and in the lagoon itself, including various wartime artifacts (tanks, munitions, unexploded ordnance and ordnance fragments) associated with the invasion of Saipan by US troops during WWII (AMPRO, 2005). Additionally, Agingan Point at the southern end of the lagoon (Fig. 1) was used as a civilian dumpsite shortly after WWII up until the 1970s (Kluge, 1969). Visible reminders of this unregulated waste disposal practice still exists in the lagoon and adjacent forereef areas to this day, despite earlier clean-up efforts by local authorities. Determining the heavy metals status of sediments in these waters as a prelude to biota analysis was therefore seen as a logical extension of our earlier surveillance studies in the lagoon and is the focus of the study reported here.

Surface sediments (~2 cm depth) were collected from 22 storm drain discharge points in the southern half of the lagoon between the villages of Garapan and San Antonio (Fig. 1). Samples were also taken for analysis along transect lines extending seaward from 16 of these drains. The latter samples were taken at locations 10, 25, 50, 100 and 250 m offshore along all transects, and at 500 and 1000 m distances on some that extended into the wider portions of the lagoon (Fig. 1). All site locations were fixed by GPS and offshore collections beyond the 100 m mark were made from a small, motorized boat using mask and snorkel.

Sediments throughout the study area were largely composed of biogenic carbonates (e.g., coral, shells, calcareous algae, foraminifera) mixed with alluvium from the weathering of inland limestone

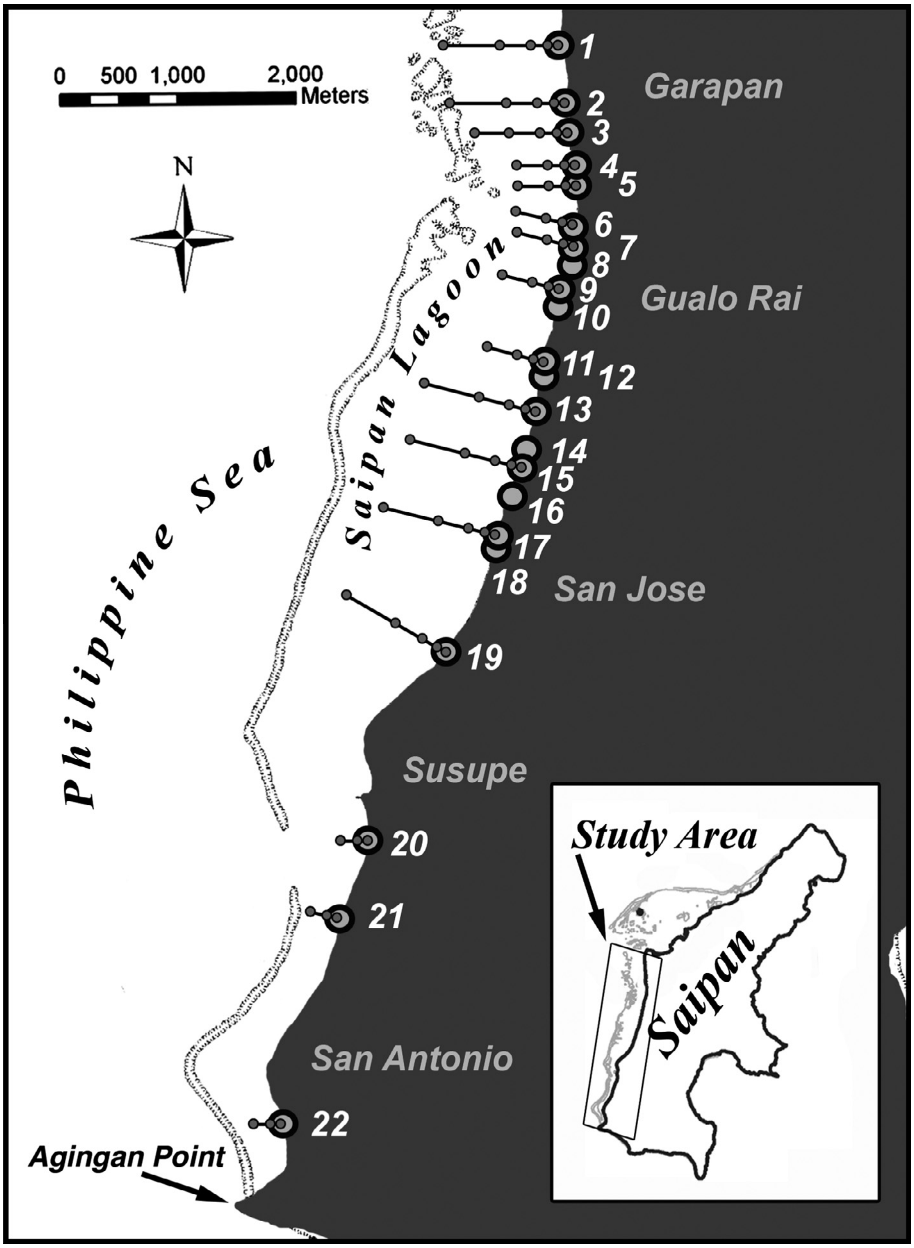

Fig. 1. Western seaboard of southern Saipan showing major villages and locations of stormwater discharge points along the coast (numbers assigned for the sole purpose of this investigation). Sediment samples collected from each discharge point and at intervals along seaward transects originating from 16 of them as shown (scale does not permit resolution of transect sampling points less than 100 m offshore). Insert: map of Saipan highlighting the study area relative to the rest of the Saipan Lagoon.

formations (Carruth, 2003; Cloud, 1959) and quarried limestone back-fill used for road constructions along the coast. They transitioned from well sorted medium to coarse grained sand in the intertidal zone to variable mixes of medium to fine grained sand, silt and clay immediately offshore. Clean, well sorted, coarser deposits were generally encountered further out. A band of fine sedimentary material extended 50–100 m offshore between storm drains 1–16 and supported dense stands of a tall seagrass (*Enhalus acoroides*). Groundwater intrusion was very evident along this particular stretch of coastline. Further south, nearshore sediments were notably coarser and a variety of benthic algae and a smaller seagrass (*Halodule uninervis*) were the dominant macrophytes.

Sediments collected for analysis were gently scooped up in hand-held, acid-washed, plastic containers. An initial sample was collected from each storm drain discharge point on July 31, 2008. A second sample was taken from discharge points at transect origins on August 5–7, 2009. All offshore sediments were also collected over the latter time frame. In the latter instance, three separate sediment samples were taken within an approximate ~3 m diameter circle at each location.

In the laboratory, the samples were air dried at 30 °C, gently disaggregated and sieved through a 1-mm Nylon screen. Heavy metal analyses were performed by atomic absorption spectroscopy following conventional wet oxidation in hot nitric acid for 3 h at 100 °C. All analytical procedures and QA/QC protocols were the same as those described in an earlier communication (Denton and Morrison, 2009). Mean metal recoveries from the standard soil reference material, PriorityPollutnT™/CLP Inorganic Soils (Catalog No. PPS-46; Lot No. 242) were within 90% of certified values for all elements and were considered acceptable.

The trace metal data are summarized in Table 1 (Appendix A) for each site. Iron was consistently the most abundant element examined while Cd was rarely detected and Ag not at all. Levels of most detectable elements were generally higher at storm drain discharge points and attenuated seaward. Appreciable variability in metal concentrations was noted between discharge points for some elements and not others. Max–min geometric mean ratios for Cr and Ni, for example, were four and eight respectively, compared with 69 and 91 for Mn and Fe. Within-site variations were generally much lower with close to 80% of all 2008/2009 storm drain data set comparisons differing by less than a factor of five. Between-site variability diminished rapidly seaward with max–min geometric mean ratios for all metals except Hg, differing by a factor of three, or less, at the 500 m mark. Offshore replicates for most elements seldom differed by more than a factor of two at any site. Again Hg was the notable exception.

Several contributing factors could account for the relatively high metal variations between discharge points including discrete

Table 2
Trace metal enrichment in surface sediments from saipan lagoon.

Offshore distance (m)	Enrichment category	Sites showing some degree of metal enrichment above baseline							
		Cd	Cu	Fe	Hg	Mn	Ni	Pb	Zn
0	Extremely high	–	–	–	–	–	–	22	3
	Very high	–	–	–	–	–	–	–	2,5,11,14,18,22
	Significant	–	3,11,22	3,11	2,4,5,22	11	–	1–6,10,11,13–15,18	1,4,6–8,10,12,13,15–17,19–21
	Moderate	3,11	2,4–6,13–15,20	2,4,22	1,3,6,8–16,19,21	6–8	11	9,12,16,19	9
10	Extremely high	–	–	–	–	–	–	22	–
	Very high	–	22	–	–	–	–	–	22
	Significant	–	3	22	22	–	–	2–4	2–5,19
	Moderate	–	–	3,4	3,5,9,17,20	–	–	1,5,6,9,11,13,15,17,19	1,6,9,11,13,15,17
25	Extremely high	–	–	–	–	–	–	22	3,22
	Very high	–	22	–	–	–	–	–	
	Significant	–	3	22	9,22	–	–	2–4	1,2
	Moderate	–	–	3	2,3,5,7,13,15,17,19,20	–	–	1,5,6,11,13	4–6,11,13
50	Extremely high	–	–	–	–	–	–	–	–
	Very high	–	–	–	–	–	–	–	–
	Significant	–	–	–	7,9	–	–	3,22	1,3,22
	Moderate	–	3,22	2,3,6	1,2,5,6,11,15,19,22	–	–	1,2	2,6
100	Extremely high	–	–	–	–	–	–	–	–
	Very high	–	–	–	–	–	–	–	–
	Significant	–	–	–	7	–	–	22	22
	Moderate	–	–	3	1–3,5,6,9,15,22	–	–	–	2,3
250	Extremely high	–	–	–	–	–	–	–	–
	Very high	–	–	–	7	–	–	–	22
	Significant	–	22	22	11,15	–	–	22	2,20
	Moderate	–	7,9,13,	1–3,7	5,9,13,17,19,20,21	2–4,6,7,9,15	–	–	1,21
500	Extremely high	–	–	–	–	–	–	–	–
	Very high	–	–	–	7	–	–	–	–
	Significant	–	–	–	9,11,19	–	–	–	–
	Moderate	–	9	–	5,6,15	–	–	–	–
1000	Extremely high	–	–	–	–	–	–	–	–
	Very high	–	–	–	–	–	–	–	–
	Significant	–	–	–	19	–	–	–	–
	Moderate	–	–	–	15	–	–	–	15

Samples collected from storm drain discharge points and nearshore waters up to 100 m offshore were normalized against intertidal and nearshore calcium carbonat-rich sediments from Pago Bay, Guam (Denton and Morrison, 2009). Geometric mean values ($n = 26$) for Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb and Zn in this reference material were: <0.20, 2.81, 0.81, 300, 0.003, 14.6, 0.76, 0.41, and 0.96 μg/g respectively. All other sediments were normalized against samples collected 1000 m offshore during the present study ($n = 21$). Corresponding geometric mean values for the above metals in this material were: <0.20, 2.26,<0.20, 46, 0.0017, 2.00,<0.40,<0.40 and <0.20 μg/g respectively. All non detects were assigned values equal to 50% of their respective analytical detection limits. Dashes indicate no enrichment at category indicated.

locational difference in the frequency, duration and intensity of storm events and antecedent dry periods prior to sampling, as well as proximity differences to metal sources within feeder watersheds. However, perhaps the greatest source of variation throughout this entire study was associated with particle size distribution discrepancies between the samples analyzed. To compensate for such sediment heterogeneity and highlight anthropogenic metal inputs, the data were normalized against Cr using the approach described by Salomons and Förstner (1984). Chromium was consistently detected in all samples and the relatively narrow range of individual values encountered (1.27–12.3 µg/g dry wt.) suggested zero to minimal anthropogenic input of this element throughout the study area. Although Fe and Mn are more popularly used as proxies for the finer sediment fraction (<63 µm), the likely presence of steel and ferrous metal fragments in the lagoon sediments from WWII days precluded their use for such purposes here.

Anthropogenic metal inputs throughout the study area were identified by comparing the Cr normalized data of the samples with that obtained from a suitable reference material to obtain enrichment factors (EFs). The equation used to derive EF may be written as follows:

$$\mathrm{EF} = \frac{[C_{\mathrm{metal}}/C_{\mathrm{Cr}}]_{\mathrm{sample}}}{[C_{\mathrm{metal}}/C_{\mathrm{Cr}}]_{\mathrm{reference\ material}}}$$

where C_{metal} and C_{Cr} are the geometric mean concentrations of the metal of interest and Cr in the sample and reference material respectively.

Reference material datasets were obtained for beach/nearshore sediments from the northern half of Pago Bay in Guam (Denton and Morrison, 2009) and from samples collected 1000 m offshore during the present investigation. The latter samples were used to calculate EF values for offshore sediments beyond the seagrass zone (250–1000 m offshore), while the Pago Bay deposits, being geochemically and texturally similar to nearshore sediments in Saipan Lagoon, were used to derive EF values for all other samples (0–100 m offshore). Both reference material datasets were not influenced by urban runoff and only minimally impacted by anthropogenic activities, if at all. The obtained EF values were ranked using a scaling system similar to that developed by Sutherland (2000) where $\mathrm{EF} < 2$ = no enrichment; $2 < \mathrm{EF} < 5$ = moderate enrichment; $5 < \mathrm{EF} < 20$ = significant enrichment; $20 < \mathrm{EF} < 40$ = very high enrichment, and $\mathrm{EF} > 40$ = extremely high enrichment. The results of this analysis are summarized in Table 2 and briefly discussed below.

Sediments from all storm drain discharge points (22 sites) were shown to be enriched with Zn and the great majority were contaminated with Hg (18 sites) and Pb (17 sites). One in every two storm drain outlets showed some degree of Cu enrichment while five, or less, were contaminated with Cd, Fe, Mn and Ni. Sediments from the site 11 storm drain outlet were the only samples enriched with all eight metals. The fact that this particular storm drain was located directly across the highway from a car dealership and vehicle maintenance/service area may have been the reason for this.

Zinc also produced the strongest pollution signal overall with EF values falling into the *significant enrichment* category, or higher, in sediments from all but one storm drain outlet. The pollution signal for Pb was also relatively strong with EF values falling into the *significant enrichment* category or higher at 13 of these sites. In contrast EF values for the remaining metals were predominantly confined to the *moderate enrichment* category or lower.

Only two case of *extremely high* metal enrichment were identified in this region of the beach: at site 3 for Zn and site 22 for Pb. The former drain was located 30 m south of a public boat ramp, the associated activities of which have undoubtedly contributed towards the elevated loadings of Zn and other metals found here. The latter drain discharged onto the beach approximately 370 m north of the Agingan Point former dumpsite area. Although heavy metal enrichment was expected in this part of the lagoon, the total area impacted was surprising and extended from the storm drain to the reef crest, a distance of approximately 350 m. The Pb footprint alone covered an approximate area of 100,000 sq m (~0.1 sq km). Other heavy metals of toxicological concern in this region were Cu and Zn (Tables 1 and 2).

Overall ~80% of all offshore sites exhibited some degree of enrichment for at least one element examined. It is interesting to note that Hg surpassed Zn as the most commonly encountered contaminant metal beyond the 10 m mark and exhibited some degree of enrichment in sediments from just over half (55%) of all offshore sampling sites. By comparison, slightly more than one third (36%) of all offshore sites were enriched with Zn. Equivalent values for all other detectable metals in descending rank order were: Pb (28%) > Fe (14%) > Cu (11%) > Mn (7%).

In general, EFs for most metals diminished with increased distance offshore. This is to be expected for metal contributions mobilized into the lagoon from land-based sources. There were instances, however, where the opposite trend was apparent leading us to suspect the existence of discrete metal 'hot-spots' within the lagoon itself. Distribution profiles for Hg were especially noteworthy in this regard and frequently showed elevated levels in sediments collected 250–1000 m offshore. The most striking example of this anomaly occurred along the storm drain 7 transect and is illustrated in Fig. 2. It can be seen that mean mercury concentrations in surface sediments rose sharply seawards and peaked 250 m offshore. Other such examples within this distance range were evident on transects originating from storm drains 11, 15, and 19 (Tables 1 and 2).

Interestingly, these offshore mercury spikes were frequently accompanied by increased Cu, Fe, Mn and Zn enrichment, which suggest they were remnant artifacts of the US invasion of Saipan in 1944. Mercury fulminate, for example, was the primary explosive used in primers and detonators of artillery shells and percussion caps of bullets during WWII (US Navy, 1947). Mercury switches were also commonplace in certain types of projectiles and rockets used at the time (US Navy, 1946). Given the enormity of fire power delivered by both US and Japanese forces before and during the Saipan invasion (Crowl, 1960; Trueman, 2000; Moore,

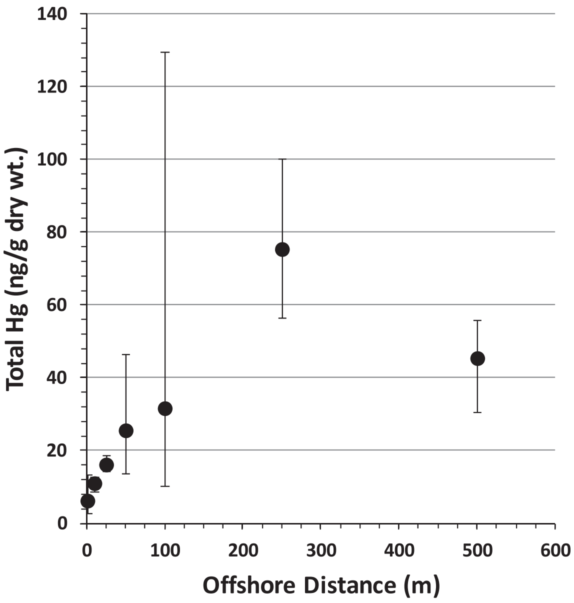

Fig. 2. Mercury levels in sediments collected along a seaward transect originating from storm drain 7. Plots are geometric means; whiskers are concentration ranges.

Table 3
Sediment quality guideline (SQG) exceedances in current study.

Metal	Threshold effects level (TEL) (μg/g)[a]	Site exceedance (offshore distance)[b]	Probable effects level (PEL) (μg/g)[a]	Site exceedance (offshore distance)
Ag	0.73	None	1.77	None
Cd	0.68	11 (0 m)	4.21	None
Cr	52.3	None	160	None
Cu	18.7	11 (0 m), 20 (0 m), 22 (10 m)	108	None
Fe[c]	21,200	None	43,766	None
Hg	0.13	7 (100 m)	0.7	None
Mn[c]	460	None	1100	None
Ni	15.9	None	42.8	None
Pb	30.2	22 (0, 10 and 25 m)	112	None
Zn	124	20 (0 m)	271	None

[a] SQGs from MacDonald et al. (1996).
[b] Exceedance shown by one or more replicates.
[c] SQGs for Fe and Mn from Persaud et al. (1992) for 'Lowest Effect Level' and 'Severe Effect Level'.

2002), it seems reasonable to assume that the pockets of metal contamination created by exploding ordnance and lost ammunition are still detectable in places along the coast and in the lagoon to this day.

The numerical sediment quality guidelines (SQGs) developed by MacDonald et al. (1996) for Florida coastal waters are applicable to calcium carbonate-rich sediments and therefore provide a useful means of predicting the potential ecological impact of sediment associated contaminants in the study reported here. These guidelines, referred to as the threshold effects level (TEL) and probable effects level (PEL) SQGs, define three ranges of contaminant concentrations where adverse biological effects rarely occur (<TEL), occasionally occur (TEL to PEL), or frequently occur (>PEL). The guidelines developed by these authors are listed in Table 3 and were available for all metals examined except Fe and Mn. SQGs for the latter elements were therefore adopted from those developed earlier by Persaud et al. (1992), (cited in Jaagumagi, 1992).

In comparing these SQGs with our site geometric mean data listed in Table 1, no PEL (or equivalent Persaud et al., 1992 guideline) exceedances were evident for any metal considered (Table 3). However, nearshore sediments impacted by the earlier dumping activities at Agingan Point (site 22) exceeded TELs for Cu and Pb. Additionally, TELs for Cd and Cu were closely approached in beach and nearshore sediments adjacent to the public boat ramp (site 3). Occasionally, a TEL for a particular metal was equaled, or exceeded in a single site replicate but not by the site geometric mean. Such was the case for Hg in sediments taken 100 m offshore at site 7 (max: 0.13 μg/g); Cd and Cu in beach sand from site 11 (max: 0.71 μg/g and 19.5 μg/g respectively); Pb in beach sand from site 22 (max: 31.0 μg/g), and Zn and Cu in beach sand from site 20 (max: 163 μg/g and 38.0 μg/g respectively). The site 20 outlet was of particular interest to us because it was the only point of discharge for runoff from Susupe village, the second largest coastal village in the study area after Garapan (Fig. 1). Evidence of Zn and Cu enrichment in beach deposits down gradient of this outlet was, therefore, expected in view of traffic densities along the adjacent stretch of coastline. The unusually large temporal discrepancies between the 2008 and 2009 data sets (Table 1) at this location was also presumed to reflect the limitations of the sampling strategy relative to storm events rather than to major change in metal loadings within the storm drain catchment area.

In summary, this investigation provides convincing evidence that heavy metals are continually being mobilized into the southern half of Saipan Lagoon from land-based sources in stormwater discharges. Undoubtedly, traffic from the main coastal highway is a major contributing source for these contaminants, as are the commercial premises dotted along its length. The findings of the study also point towards lagoon-based sources of heavy metals believed to be of WWII origin. Mercury, in particular, was widely distributed within these waters at moderate levels of enrichment and occasionally above. To what extent this has influenced Hg levels in fisheries resources within this section of the lagoon remains to be evaluated. Very few sites examined in this study were contaminated with any metal above threshold concentrations considered ecologically harmful. Those that were, were confined to relatively small sections of the intertidal zone or nearshore waters with one notable exception. The former dumpsite area at the southern end of the lagoon (site 22) was extensively contaminated with several elements that could conceivably induce adverse biological effects in sensitive species. Ironically, the natural beauty of this heavily contaminated section of Saipan Lagoon belies its sad history and continues to attract tourists and local residents who fish the waters and harvest bivalves, seaweed and other benthic organisms for food. Public health risks associated with the unrestricted consumption of these resources are currently being addressed.

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Appendix A. Supplementary material

Supplementary data associated with this article (Table 1) can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2014.01.014.

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