Heavy metal contamination of coastal lagoon sediments: Fongafale Islet, Funafuti Atoll, Tuvalu

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24 **Abstract**

25 To evaluate contamination of coastal sediments along Fongafale Islet, Central Pacific,
26 a field survey was conducted in densely populated, sparsely populated, open dumping and
27 undisturbed natural areas. Current measurements in shallow water of the lagoon indicated
28 that contaminants from the densely populated area would only be transported for a small
29 proportion of a tidal cycle. Acid-volatile sulfides were detected in both the intertidal
30 beach and near shore zones of the densely populated area, whereas these were no
31 detection in the other areas. This observation lends support to argument that the coastal
32 pollution mechanism that during ebb tide, domestic wastewater leaking from poorly
33 constructed sanitary facilities seeps into the coast. The total concentrations of Cr, Mn, Ni,
34 Cu, Zn, Cd and Pb were relatively high in all of the areas except the undisturbed natural
35 area. The indices of contamination factor, pollution load index and geo accumulation
36 index were indicative of heavy metal pollution in the three areas. The densely populated
37 area has the most significant contamination; domestic wastewater led to significant
38 contamination of coastal sediments with Cr, Zn, Cu, Pb and Cd. The open dumping area
39 is noteworthy with respect to Mn and Ni, which can be derived from disposed batteries.

40

41 **Keywords:** Heavy metals; Acid-volatile sulfides; Sediment; Principal component
42 analysis; Pacific island

43

44 **1. Introduction**

45 Globally coral reefs are under significant ecological decline with estimates
46 suggesting that 20% of the world's reefs have been destroyed and a further 25% at risk of
47 collapse(Hughes et al., 2003; Wilkinson, 2004). Such ecologically driven-decline is being
48 forced by a number of factors which include: global environmental changes (increasing
49 sea surface temperatures, ocean water chemistry and sea level change) and anthropogenic
50 factors (over fishing, resource exploitation and urban development)(Kench et al., 2009).
51 Collectively such impacts compromise the suite of ecosystem services afforded by coral
52 reef ecosystems. Among the suite of anthropogenic effects on reef systems coastal
53 pollution has been identified as a particularly pressing concern.

54 Atolls had been considered to be pristine environment. However, anthropogenically
55 induced water pollution on Pacific atolls has been recognised for several decades
56 (Hirshfield et al., 1968; Johannes et al., 1979; Kimmerer and Walsh, 1981). Densely
57 populated and urbanised islands of atolls have been affected by domestic wastewater
58 discharge and municipal solid waste (MSW) dumping (Richmond et al., 2002; Abraham
59 et al., 2004; Vieux et al., 2004; Fujita et al., 2009). To date, studies of reefal water quality
60 have focussed on changes in nutrient loads and the consequent impact on ecological
61 communities. However, there have been few studies of heavy metal contamination of
62 coastal waters and sediments(Burger and Gochfeld, 2000; Miao et al., 2001).

63 In Central Pacific atolls, the shells of large benthic foraminifera are the primary
64 components of sand-size sediments (Collen and Garton, 2004; Yamano et al., 2005).
65 Coral reefs on the ocean side of the islands also act as a natural breakwater (Kayanne et
66 al., 2005). Reef islands are vulnerable to a range of climatic changes, including sea level
67 rise and similar extreme conditions (Mimura, 1999). Consequently, a degradation of coral

68 reefs and a decline in the sand producers, both caused by human activities, will accelerate
69 the onset of serious problems associated with the expected rise in future sea levels.

70 Foraminiferal researchers have reported that anthropogenic nutrification caused a
71 change in benthic foraminiferal communities (Hallock et al., 2003; Uthicke and Nobes,
72 2008; Osawa et al., 2010; Carilli and Walsh, 2012) and large benthic foraminifera were
73 rare or absent in reef flats near densely populated areas (Ebrahim, 2000; Fujita et al.,
74 2009). Large benthic foraminifera are sensitive to environmental stress due to heavy
75 metals (Coccioni, 2000; Frontalini and Coccioni, 2008; Prazeres et al., 2011). Heavy metal
76 contamination is harmful also for fishes, algae and shellfishes, which are important for
77 atoll people as marine resources. This may threaten human health. For these reasons, it is
78 important to comprehensively understand contamination of coastal sediments on reef
79 islands that results from human activities.

80 The objectives of this study are 1) to determine heavy metal concentration in coastal
81 sediments of densely populated, sparsely populated, open dumping and undisturbed
82 natural areas of Fongafale Islet, Tuvalu; 2) to assess the degree of heavy metal
83 contamination using previously proposed contamination indices; and 3) to characterise
84 contamination in coastal sediments using principal component analysis (PCA). The
85 results are also discussed in terms of the implications for ecological processes and
86 construction of reef islands in mid-ocean island settings.

87

88 **2. Materials and methods**

89 *2.1 Study area*

90 The study area was Fongafale Islet (8°31'S, 179°12'E) on Funafuti Atoll, Tuvalu,
91 Central Pacific (Fig. 1). Fongafale Islet is the capital of Tuvalu and is the largest

92 settlement in the country. The islet is ~12 km long and is 0.8 km at its widest point and
93 shares geomorphic features with other atoll islands that experience occasional storms: an
94 oceanward storm ridge, central depression and a lagoon ward beach ridge (Yamano et al.,
95 2007). The islet is low lying with a mean elevation of ~1.5 m above sea level and
96 composed of larger gravel/cobble-size sediments on its ocean margin and finer sand-size
97 materials along its lagoon shoreline. The lagoonal sediments consist of gravel (0–56.4%),
98 sand (42.0–97.9%), and silt and clay (0–10.6%) (Smith, 1995) and the lagoonal sands are
99 composed of large benthic foraminifera (28–54%), coral fragments (33–54%) and shell
100 fragments (10–23%) (Sharma, 2010). With a population of 4,492 people Fongafale
101 represents a densely populated urban settlement (Secretariat of the Pacific Community,
102 2005). The climate at Fongafale Islet is tropical and the average temperature is ~28 °C
103 throughout the year. The annual precipitation is ~3,000 mm.

104 Tuvalu has no factories and sewage treatment plants. Of 639 households on
105 Fongafale, 424 households have buried septic tanks that receive domestic wastewater and
106 163 households have pit toilets with a pour flush (Secretariat of the Pacific Community,
107 2005; Lal et al., 2006). Thus, 92% of the household have access to improved sanitary
108 facilities; however, the bottoms of septic tanks are not sealed against seepage, and are
109 referred to as ‘bottomless’ (Lal et al., 2006; Fujita et al., 2013). Consequently, domestic
110 wastewater leaking from these poorly constructed sanitary facilities was found to be the
111 major pollution source for the lagoonal coast of the densely populated area, the central
112 part of Fongafale Islet (Fujita et al., 2013). An open dumping site is located on the northern
113 end of the road (Fig. 1). MSW is collected by Funafuti Town Council (Merz, 2004) and
114 then is dumped in the site. The primary means of disposal of waste at the site is routine
115 burning.

116

117 *2.2 Experimental design*

118 A field survey was conducted from 20 January to 24 January 2011. The survey
119 focused on the lagoonal coast of the islet and eleven sampling sites were selected (Fig. 1).
120 Sites 1 to 6 were located along the densely populated area; sites 1, 3 and 5 are on the
121 intertidal beach, whereas sites 2, 4 and 6 were in the near shore in water depths of ~1 m.
122 Sites 7 and 8 are on a sparsely populated area, which is located ~5 km north of the main
123 settlement. Sites 9 and 10 are on the open dumping area ~7 km north of the main
124 settlement. Site 11 is near the northern tip of the islet, where there are no nearby
125 inhabitants, and is thus considered to be very close to an undisturbed natural environment.
126 Grounded ships, shipwrecks and seawalls, which can be sources of heavy metal
127 contamination in atolls, were not observed at the sampling sites. During the sampling
128 period, daily observed precipitation was as follows: 20 January, 43.7 mm; 21 January,
129 45.3 mm; 22 January, 73.0 mm; 23 January, 22.9 mm; 24 January, 38.6 mm, which was
130 provided by Tuvalu Meteorological Service.

131

132 *2.3 Water current measurement*

133 A current meter (JFE Advantech, INFINITY-EM) was installed in the nearshore of
134 the central part of the lagoon shoreline in order to determine net flow and velocity
135 patterns in the near shore that may control dispersal of contaminants (Fig. 1). The
136 instrument was mounted 0.2 m above the sediment surface and was programmed to record
137 average velocity and flow direction every 10 min. Current measurements occurred during
138 spring tide conditions between January 20th and 24th. Water depth at the installation site
139 ranged from 0.35 m to 2.48 m during the observation period.

140

141 *2.4 Sampling and chemical analysis*

142 Three sediment samples(<2 mm diameter) were retrieved at each of the 11 sample
143 site, totalling 33 samples. Sediment samples at all of the sites, except sites 1, 3 and 5 were
144 collected from the top 0.1 m of the sediment surface. On the intertidal beaches (Sites 1, 3
145 and 5), holes were dug until grey sand with a hydrogen sulfide odour was reached(Fujita et al.,
146 2013)(generally occurring at ~0.5 m depth) and the grey sand was then sampled. The
147 sediment samples consisted of sand (98.2–99.2%) and silt and clay (0.8–1.8%) and
148 composed of skeletal carbonate grains. Any pellets, fragments of metals or rusts were not
149 contained in the samples.

150 Determination of acid-volatile sulfides (AVS) concentration in the sediments was
151 undertaken in triplicate using an AVS detector tube (Gastec, Model 210H and
152 210L) following the manufacturer's instructions.

153 For heavy metal analysis, 0.5 g of the dried sediment was mineralised with HNO₃.
154 Digestion of the sample was performed in a microwave mineralisation system (Milestone,
155 MLS-1200 MEGA) in five steps: 250W for 5 min; 0 W for 1 min; 250W for 5 min; 400W
156 for 5 min; 600W for 5 min. The final determination of Cr, Mn, Ni, Cu, Zn, Cd and Pb was
157 conducted with external and internal standard method using an inductively coupled
158 plasma mass spectrometer (Yokogawa Analytical Systems, Agilent 7500cs). Detection
159 limits of the seven elements are as follows: 0.3ng g⁻¹ sediment dry weight for Cr, 0.2ng
160 g⁻¹ for Mn, 0.4ng g⁻¹ for Ni, 0.2ng g⁻¹ for Cu, 0.08ng g⁻¹ for Zn, 0.02ng g⁻¹ for Cd and
161 0.02ng g⁻¹ for Pb. The standard error of each three sediment sample was less than 5%.

162

163

164 2.5Indices of sediment contamination

165 Three separate indices were used to examine the contaminant level of sediments. The
166 indices have been widely used for the assessment of heavy metal contamination in
167 various environments such as coastal, estuarial, river, lake and agricultural sediments
168 (Hakanson, 1980; Tomlinson et al., 1980; Amin et al., 2009; Bhuiyan et al., 2010; Varol,
169 2011). In the interpretation of sediment contamination, choice of background
170 concentration values plays an important role. While many previous studies used the
171 average values of shale or crustal abundance as a reference baseline, the best alternative is
172 proposed to compare concentrations between contaminated and mineralogically and
173 texturally comparable, uncontaminated sediments (Covelli and Fontolan, 1997; Rubio et
174 al., 2000; Sakan et al., 2009). There were no data on background concentrations for the
175 investigated Fongafale coastal lagoon sediment. As noted previously, shells of large
176 benthic foraminifera are the primary components of sand-size sediments in Central
177 Pacific (Collen and Garton, 2004; Yamano et al., 2005). In fact, the lagoonal sand
178 compositions are similar to those at the northern tip of the islet (Sharma, 2010) which is
179 considered to be very close to the undisturbed natural environment. Thus, the background
180 values in the present study were calculated from the mean concentrations of heavy metals
181 at site 11.

182 2.5.1 Contamination factor (CF)

183 The *CF* is the ratio obtained by dividing the concentration of each heavy metal in the
184 sediment ($C_{heavy\ metal}$) by the concentration in background ($C_{background}$):

$$185 \\ 186 CF = \frac{C_{heavy\ metal}}{C_{background}} \\ 187$$

188 CF values were interpreted as follows: low contamination, $CF < 1$; moderate contamination,
189 $1 < CF < 3$; considerable contamination, $3 < CF < 6$; and very high
190 contamination, $CF > 6$ (Hakanson, 1980).

191

192 2.5.2 Pollution load index (PLI)

193 The PLI was determined as the n th root of the product of the n CF :

194

195

$$196 \quad PLI = \left(\prod_i^n CF_i \right)^{1/n}$$

197

198 Values of $PLI > 1$ imply that heavy metal pollution exists. Otherwise, if $PLI < 1$, there is no
199 heavy metal pollution (Tomlinson et al., 1980).

200

201 2.5.3 Geoaccumulation index (I_{geo})

202 The I_{geo} is defined as the following equation:

203

204
$$I_{geo} = \log_2 \left(\frac{C_n}{1.5 B_n} \right)$$

205

206 Where C_n is the concentration of heavy metal assayed in sediment samples and B_n is the

207 background concentration of the heavy metal (n). The value of 1.5 is the background

208 matrix correction factor due to lithospheric effects (Müller, 1979; Bhuiyan et al., 2010;

209 Varol, 2011). A scale of I_{geo} values indicated differences in level of pollution where: $I_{geo} < 0$

210 denotes unpolluted sediments; unpolluted to moderately polluted when $0 < I_{geo} < 1$;

211 moderately polluted when $1 < I_{geo} < 2$; moderately to heavily polluted when $2 < I_{geo} < 3$;

212 heavily polluted when $3 < I_{geo} < 4$; heavily to extremely polluted when $4 < I_{geo} < 5$; extremely
213 polluted when $I_{geo} > 5$ (Müller, 1979).

214

215 *2.6 Principal component analysis*

216 PCA (Hopke, 1983) was performed on the normalised data of heavy
217 metal concentration in the sediment to describe the association of the sediment samples,
218 using a statistical package (SPSS Japan, PASW[®] Statistics 18). Factor loadings of the
219 original variables (the seven heavy metals) were computed and then a principal
220 component biplot was generated to display graphically relationships between the
221 sampling sites and relationships between the variables. PCA has been widely used in
222 analysis for source tracing of heavy metal contamination in sediment (De Bartolomeo et
223 al., 2004; Bhuiyan et al., 2010; Varol, 2011).

224

225 **3. Results and discussion**

226 *3.1 Water current in the shallow water of the lagoon*

227 Results of the current meter deployment in the nearshore, which included spring tide
228 conditions, showed no clear current direction dependence was found. Currents were not
229 modulated by the flood and ebb tides (Fig. 2), in contrast to the observations and model
230 estimates of Damlamian (2008). This was because our observation was made in the
231 nearshore. Mean current speed was 0.02 ms^{-1} and peaked at $\sim 0.10 \text{ ms}^{-1}$. The current
232 patterns and magnitude of velocity suggest contaminants would only be transported for a
233 small proportion of a tidal cycle (dependant on entrainment potential).

234

235

236 3.2 AVS and heavy metals in the coastal sediments

237 Table 1 presents the results of the analysis of the 33 sediment samples (11 sites in
238 triplicate). AVS concentrations ranged from 0.024 to 0.133 mgg⁻¹ in the densely
239 populated area. In this densely populated area, AVS was detected at both the intertidal
240 beach zone (sites 1, 3 and 5) and nearshore zone (sites 2, 4 and 6), and is likely present
241 due to the leaking domestic waste water through coastal sediment on the beachface. Such
242 a pollution mechanism was previously identified by Fujita et al. (2013). In contrast to the
243 densely populated zone, AVS were not detected in the sparsely populated, open dumping
244 and undisturbed natural areas. In the non-populated areas there is no pollution mechanism
245 that could generate AVS at the coast. In the sparsely populated area domestic water
246 leakage is likely to be significantly less and of insufficient concentration to be detected.

247 The heavy metal concentrations in the coastal sediments of the atoll were much
248 lower than those in the coastal, estuarial, bay, river, lake and agricultural sediments in
249 other environments (except coral reefs) (Hakanson, 1980; Tomlinson et al., 1980; Urase et
250 al., 2006; Amin et al., 2009; Bhuiyan et al., 2010; Varol, 2011). The heavy metal
251 contamination shows considerable variation in both the concentration and presence of
252 different heavy metals. The area with the highest concentration was the densely populated
253 area, followed by the open dumping, sparsely populated and undisturbed natural areas.
254 The accumulation order of heavy metals in each area was Zn > Pb > Cu > Cr > Mn > Ni >
255 Cd for the densely populated area, Ni > Cu > Mn > Zn > Cr > Pb > Cd for the open
256 dumping area, Zn > Ni > Cr > Pb > Mn > Cu > Cd for the sparsely populated area and Ni
257 > Zn > Cr > Pb > Mn > Cu > Cd for the undisturbed natural area (Table 1).

258

259 3.3 Assessment of sediment contamination

Table 2 presents the *CF* and *PLI* results. The highest *CF* values for all of the heavy metals studied (except Ni) were found in sediments of the densely populated area. The *CF* values for Cr, Mn, Cu, Zn, Cd and Pb in the densely populated area were >6, except Cr and Mn at site 1, Ni at all sites and Zn at site 4. This indicates “very high contamination” by these heavy metals. In the sparsely populated area, the *CF* values for all of the metals studied indicated “considerable contamination” or “moderate contamination”. The highest *CF* value for Ni was found at site 9 (the open dumping area), which suggests “moderate contamination”. The *CF* values for Mn and Cu in the open dumping area denote a “very high contamination”, those for Cr, Zn and Pb indicate “considerable contamination” or “moderate contamination”, and those for Cd show “very high contamination” or “moderate contamination”. The total *CF* values show contamination was highest in the densely populated area, followed by the open dumping and sparsely populated areas.

The *PLI* ranged from 6.0 to 12.5, 1.8 to 3.1 and 3.9 to 4.5 for the densely populated, sparsely populated and open dumping areas, respectively. These values indicate that all the sediments of the three areas are contaminated with the heavy metals studied.

Table 3 presents *I_{geo}* values of the heavy metals. The highest *I_{geo}* values of heavy metals studied (except Ni) were found in the sediments of the densely populated area. The highest *I_{geo}* value for Cu in the densely populated area denotes an “extremely polluted,” values for Zn, Cd and Pb indicate “heavily to extremely polluted”, value for Mn shows “heavily polluted” and that for Cr denotes “moderately to heavily polluted”, whereas value for Ni indicates “unpolluted.” The highest *I_{geo}* value for Ni was found at site 9 (the open dumping area), which shows “unpolluted to moderately polluted.” The total *I_{geo}* values indicate that pollution was the highest in the densely populated area, followed by the open dumping and sparsely populated areas.

284 The assessment results by using the three indices of *CF*, *PLI* and *I_{geo}* indicate that
285 heavy metal contamination of sediments existed in each of the three areas examined. Each
286 index consistently indicates that the densely populated area had greatest levels of
287 contamination, followed by the open dumping and sparsely populated sectors of the islet.

288

289 *3.4 Characterisation of sediment contamination using principal component analysis*

290 PCA was performed to compare the heavy metal composition between the sediments
291 and to identify noteworthy elements in the contaminated areas. Kaiser–Meyer–Olkin and
292 Bartlett’s results were 0.60 and $p < 0.05$, respectively. This indicates that PCA would be
293 effective in the data set (Kaiser and Rice, 1974; Varol, 2013). Two PCs with eigenvalues
294 > 1 that account for a cumulative variance of 81.1% were obtained. The first PC,
295 accounting for 58.3% of the total variance, correlated with Cr, Zn, Cu, Pb and Cd (factor
296 loading > 0.70). The second PC, which accounted for 22.8% of the total variance,
297 correlated with Mn and Ni (factor loading > 0.70).

298 The dimensionality of the original seven chemical variables was reduced to two
299 factors (Fig. 3). The principal component biplot of the first and second factors shows the
300 spatial distribution of the sediment samples in the four areas. In the first and fourth
301 quadrants, the samples of the densely populated area exhibited relatively high
302 concentrations of Cr, Zn, Cu, Pb and Cd. The samples of the open dumping area were
303 located in the second quadrant and exhibited relatively high concentrations of Mn and Ni.
304 The samples of these two areas were clearly separated from site 11 (the undisturbed
305 natural area), whereas the samples of the sparsely populated area were located in the third
306 quadrant near the site 11, at which site the samples exhibited relatively low heavy metal
307 concentration.

308 Automobile can be one of pollution sources in the densely populated area. Murakami
309 et al. (2009) reported that Ni, Cu and Zn were derived from traffic activities. Ni
310 concentrations in the densely populated area were however less than those in the sparsely
311 populated and open dumping areas. Thus, it is quite unlikely that automobile is major
312 pollution source. The likely sources of the heavy metal contaminants are various types of
313 grey wastewater, which have been summarised by Eriksson et al. (2002). Naturvårdsverket
314 (1995), Christova-Boal et al. (1996) and Surendran and Wheatley (1998) found
315 wastewater originating from laundry contained the following heavy metals: $0.029 \mu\text{gL}^{-1}$
316 for Mn, $0.058\text{--}0.322 \mu\text{gL}^{-1}$ for Cu, $0.09\text{--}0.44 \mu\text{gL}^{-1}$ for Zn, $0.00063 \mu\text{gL}^{-1}$ for Cd and
317 $0.033 \mu\text{gL}^{-1}$ for Pb; and discharge from kitchen sinks contained: $0.072\text{--}0.13 \mu\text{gL}^{-1}$ for Cr,
318 $0.031\text{--}0.075 \mu\text{gL}^{-1}$ for Mn, $0.050\text{--}0.26 \mu\text{gL}^{-1}$ for Cu, $0.096\text{--}1.8 \mu\text{gL}^{-1}$ for Zn,
319 $0.00052 \mu\text{gL}^{-1}$ for Cd and $0.005\text{--}0.14 \mu\text{gL}^{-1}$ for Pb (Naturvårdsverket, 1995; Surendran and
320 Wheatley, 1998). Based on this reported data it is inferred that domestic wastewater from
321 households on Fongafale Islet contains at least Cr, Mn, Cu, Zn, Cd and Pb as heavy metal
322 elements.

323 A number of factors can explain the spatial differences in heavy metal concentration
324 along the shoreline of Fongafale. It has been reported that the distribution of heavy metals
325 can be related to sediment grain size, organic content and redox regime (Kalantzi et al.,
326 2013). The sediment samples predominantly consisted of skeletal carbonate sand as stated
327 earlier. Thus, there is no difference in sediment grain size along the shoreline. Dean et al.
328 (2007) reported that humic substances have a high content of oxygen-donor ligand, which
329 form complexes with heavy metals, particularly Cu, Zn and Pb. In this manner, the
330 organo-metallic complexes precipitate. Organic acids also form complexes with heavy
331 metals (Essington, 2003). Imai et al. (2002) reported that dissolved organic matter in

332 effluent from sewage treatment plants and human-waste treatment plants contains aquatic
333 humic substances. Thus, domestic wastewater leaking from poorly constructed sanitary
334 facilities can be inferred to also contain humic substances. Since a septic tank consists of
335 two compartments: one for settling and one for anaerobic treatment (Lal et al., 2006),
336 leaking domestic wastewater can also contain organic acids, as a result of fermentation.
337 Furthermore, the organo-metallic complexes co-precipitate with sulfide in anoxic
338 conditions (Sutherland et al., 2007). As mentioned above, AVS was detected in the coastal
339 sediments of the densely populated area and negative redox potential was observed in the
340 coastal water (Fujita et al., 2013). Collectively, the factors above provide reasons why the
341 sediments in the densely populated area contained relatively high concentrations of heavy
342 metals, when compared with the undisturbed natural area.

343 Results in this study have similarities with previous investigation of heavy metal
344 contamination. For example, Prechthai et al. (2008) reported that Cr, Mn, Ni, Cu, Zn, Cd
345 and Pb were contained in leachate from open dumping sites for MSW in Thailand, and
346 Mn, Zn and Cd showed relatively high mobility potential. The three heavy metals were
347 found to be prominent heavy metals in sediments associated with the open dumping area
348 in the present study. According to the report by Merz (2004), the composition of MSW
349 from households in Funafuti Atoll is as follows: biodegradable (52.4 wt%), papers (10.4
350 wt%), metals (steel cans, aluminium cans, refrigerators, car bodies, roofing irons and
351 copper pipes: 9.8 wt%), glass (9.5 wt%), plastics (9.3 wt%), construction and demolition
352 scraps (3.2 wt%), textiles (2.2 wt%), potentially hazardous MSW (dry cells, alkaline
353 batteries, button batteries, vehicle batteries, solar power systems and used containers: 0.6
354 wt%) and others (2.5 wt%). Given the constituents of MSW, Mn and Ni can be derived
355 from batteries, since batteries generally include heavy metals such as Mn, Ni, Zn and

356 Cd. Thus, batteries dumped can be argued to be a factor in the relatively high
357 concentrations of Mn and Ni compared with the other areas.

358 Our findings have significant implications for considerations of
359 pollution/contamination in atoll settings. To date, water pollution such as nutrification has
360 been the major focus of studies on the impact of domestic wastewater discharge. However,
361 our results show that domestic wastewater can also lead to significant contamination of
362 heavy metals in coastal sediments in reefal settings. Consequently, careful attention
363 should be paid to coastal sediment contamination of heavy metals in Pacific atolls,
364 particularly in densely populated urban settings where waste disposal and the lack of
365 wastewater treatment are common. The implications of this study are profound. Both
366 deteriorating water pollution and heavy metal contamination may have detrimental (but
367 unquantified) effects on reef ecological health. Such deterioration can impact the
368 availability of food resources, human health and the rate of carbonate sediment
369 production that is critical for the development and maintenance of reef landforms (Kench
370 et al., 2009).

371

372 **4. Conclusions**

373 Anthropogenic nutrification of coastal water near densely populated area of atolls is
374 well-recognised. The present study demonstrates for the first time that domestic
375 wastewater leaking from poorly constructed sanitary facilities, evident by an assessment
376 of contamination factor, pollution load index and geoaccumulation index, results
377 insignificant contamination of coastal sediments by heavy metals such as Cr, Zn, Cu, Pb
378 and Cd. With respect to the open dumping area, Mn and Ni, which can be derived from
379 disposed batteries, were identified as noteworthy elemental contaminants in comparison

380 with observations in the other areas. We conclude that heavy metal contamination in
381 coastal sediment of the Pacific atolls may be a significant issue for benthic sedimentary
382 environments in heavily populated areas. Given the prevalence of similar dumping and
383 sanitary facilities in atoll nations in the Pacific careful attention should be paid toward the
384 impacts of these activities on the health of coral reef ecosystem.

385

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393

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568 **Tables**

569 Table 1. Concentrations of AVS and heavy metals in the coastal sediments along

570 Fongafale Islet.

571

	Densely populated						Sparsely populated		Open dumping		Natural
	1	2	3	4	5	6	7	8	9	10	11
AVS (mg g ⁻¹)	0.102	0.047	0.034	0.133	0.024	0.075	<0.002	<0.002	<0.002	<0.002	<0.002
Cr (ng g ⁻¹)	140.0	168.0	240.0	174.5	240.0	258.5	87.6	56.4	120.0	72.0	26.2
Mn (ng g ⁻¹)	56.0	86.4	81.6	85.1	156.0	88.6	42.0	28.8	87.6	140.0	13.1
Ni (ng g ⁻¹)	53.0	74.4	67.2	67.6	100.8	84.9	132.0	100.8	216.0	170.0	81.8
Cu (ng g ⁻¹)	160.0	372.0	276.0	250.9	564.0	563.1	38.4	21.6	78.0	290.0	9.6
Zn (ng g ⁻¹)	580.0	576.0	948.0	87.3	1440.0	1015.4	204.0	61.2	192.0	35.0	34.9
Cd (ng g ⁻¹)	3.9	11.2	6.7	4.4	3.7	3.3	0.9	0.7	3.5	0.6	0.4
Pb (ng g ⁻¹)	680.0	564.0	588.0	240.0	324.0	267.7	67.2	34.8	25.2	99.0	20.7

572

573

574 Table 2. Contamination factors (*CF*) and pollution load indices (*PLI*) of heavy metals for
 575 coastal sediments of the densely populated, sparsely populated and open dumping
 576 areas.

577

Site	<i>CF</i>							<i>PLI</i>
	Cr	Mn	Ni	Cu	Zn	Cd	Pb	
1	5.3	4.3	0.6	16.7	16.6	9.7	32.8	7.5
2	6.4	6.6	0.9	38.8	16.5	27.6	27.2	10.9
3	9.2	6.2	0.8	28.8	27.2	16.6	28.4	10.8
4	6.7	6.5	0.8	26.1	2.5	10.8	11.6	6.0
5	9.2	11.9	1.2	58.8	41.3	9.2	15.6	12.5
6	9.9	6.8	1.0	58.7	29.1	8.2	12.9	10.3
7	3.3	3.2	1.6	4.0	5.8	2.2	3.2	3.1
8	2.2	2.2	1.2	2.3	1.8	1.8	1.7	1.8
9	4.6	6.7	2.6	8.1	5.5	8.6	1.2	4.5
10	2.8	10.7	2.1	30.2	1.0	1.5	4.8	3.9

578

579

580 Table 3. Geoaccumulation indices (I_{geo}) of heavy metals for coastal sediments of the
 581 densely populated, sparsely populated and open dumping areas.

582

Site	Cr	Mn	Ni	Cu	Zn	Cd	Pb
1	1.8	1.5	-1.2	3.5	3.5	2.7	4.5
2	2.1	2.1	-0.7	4.7	3.5	4.2	4.2
3	2.6	2.1	-0.9	4.3	4.2	3.5	4.2
4	2.2	2.1	-0.9	4.1	0.7	2.8	2.9
5	2.6	3.0	-0.3	5.3	4.8	2.6	3.4
6	2.7	2.2	-0.5	5.3	4.3	2.5	3.1
7	1.2	1.1	0.1	1.4	2.0	0.6	1.1
8	0.5	0.6	-0.3	0.6	0.2	0.2	0.2
9	1.6	2.2	0.8	2.4	1.9	2.5	-0.3
10	0.9	2.8	0.5	4.3	-0.6	0.0	1.7

583

584

585 **Figure captions**

586 Fig. 1. Maps of the study area. a) Tuvalu, b) Funafuti Atoll and the other atolls in Tuvalu,
587 c) Funafuti Atoll, d) observation sites in Fongafale Islet. The distribution of houses is
588 based on a QuickBird satellite image that was taken in 2006, e) the open dumping
589 area and f) the densely populated area.

590 Fig. 2. Current distribution in the shallow water of the lagoon.

591 Fig. 3. Principal component (PC) biplot of the first and second PCs for the sampling sites
592 and variables.

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595 Fujita et al._Fig. 2

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600 Fujita et al._Fig. 3

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