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Spatial and Temporal Distribution of Trace Metals (Cd, Cu, Ni, Pb, and Zn) in Coastal Waters off the West Coast of Taiwan

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ABSTRACT

Surface water samples were collected along the west coast of Taiwan during two expedition cruises which represent periods of different regional climatic patterns. Information on hydrochemical parameters such as salinity, nutrients, suspended particulate matter (SPM), and Chlorophyll *a* concentrations were obtained, and dissolved and particulate trace metal (Cd, Cu, Ni, Pb, and Zn) concentrations were determined. Spatial variations were observed and the differences were attributed to (1) influence of varying extents of terrestrial inputs from the mountainous rivers of Taiwan to the coast, and (2) urbanization and industrialization in different parts of the island. Geochemical processes such as desorption (Cd) and adsorption to sinking particles (Pb) also contributed to the variability of trace metal distributions in coastal waters. Results showed temporal variations in chemical characteristics in coastal waters as a consequence of prevailing monsoons. During the wet season when river discharges were higher, the transport of particulate metals was elevated due to increased sediment loads. During the dry season, lower river discharges resulted in a lesser extent of estuarine dilution effect for chemicals of anthropogenic sources, indicated by higher dissolved concentrations present in coastal waters associated with slightly higher salinity.

Key words: Hydrology, Trace metals, Nutrients, Taiwan, Coastal water

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1. INTRODUCTION

Coastal environments are sites where chemical species and terrestrial material are transformed, removed, and/or transported to the ocean (Boyer et al. 2006). Coastal regions are also where more than half of the world's population is located accompanied by extensive industrial and urban wastewater inputs to this region (Smith et al. 2003). Small river systems are highly influenced by seasonally variable climatic patterns which can cause extreme hydrological conditions and have a profound influence in these regions (Wen et al. 2008; Wang et al. 2012). As a consequence, coastal primary productivity can be enhanced during high river flow conditions (Hurst and Bruland 2008; Wang et al. 2012). In order to quantify the material fluxes from land to ocean, and to better understand the influence of human activities on coastal environments, studies on the chemical characteristics of coastal waters are needed (Pohl et al. 2006).

Taiwan is an uplifted mountainous island situated at the edge of the Eurasia and Philippine Sea plates and straddling the tropic of Cancer and has several unique features because of the island's climatic and hydrologic patterns. Because of the convergence of the two major plates, the island is narrow and has very small plains in the east, and most major rivers/streams are located in the western part of Taiwan. With wider plains and more water resources in the west, population density on the island is heavily tilted to that region, with major industrial and urban development occurring in western Taiwan. Taiwan has become one of the world's most powerful economies over the last few decades which has also caused its estuarine and coastal waters to face more environmental stressors (Wen et al. 2008).

The north-south mountain belt (Central Range) on the island of Taiwan has its highest elevation near 4000 m. Therefore, most rivers in Taiwan are mountainous rivers, with slopes of the watershed in the range from 1/700 to 1/34 and, with a few exceptions, having little estuarine characteristics.

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This leads to high suspended particulate matter (SPM) loads in most rivers (Dadson et al. 2003). The high Central Range also has an important influence on seasonal climate and hydrology in the different regions on the island (Chen et al. 2004). In general, most precipitation occurs during May to October, when the southwest monsoon brings plenty of moisture. Between November and April, when the northeast monsoon prevails, precipitation becomes sparse in the western parts of Taiwan due to the presence of the Central Range that blocks strong winds. River discharges reflect the pattern of seasonally variable precipitation which is most obvious in the south. According to statistics provided by the Water Resources Agency (WRA) of Taiwan, during the wet season (May - October), combined regional river discharges account for 63, 78, and 90% of the annual runoffs for the northern, central and southern parts of the nation, respectively.

The chemical composition of coastal waters off Taiwan is therefore strongly affected by river discharges which reflect natural and anthropogenic influences. Four of the major rivers contribute the majority of water and sediments to the coastal zone as judged from their river discharge and sediment yields. The Danshuei (DS) River is under high environmental stress because it flows through the metropolitan Taipei area (6-million population) and receives high domestic wastewater discharge (Jiann et al. 2005; Wen et al. 2008). The Choshuei (CS) River carries very high sediment loads to the coast (Dadson et al. 2003). The Tsengwen (TW) River flows through agricultural plains in southwestern Taiwan and is well known for its barrier islands and lagoon setting near the coast (Liu et al. 2000). The Gaoping (GP) River is a major source of fluvial sediments into the southwestern shelf-slope region off Taiwan (Kao et al. 2006). The GP River is under anthropogenic influence because the second largest city, Kaohsiung (3-million), is in its watershed. Along the western plains of Taiwan, extensive urbanization and industrialization occurs in most watersheds.

The distinctively different and varying hydrological conditions resulting from seasonal climatic patterns, combined with the influence of anthropogenic activities, especially in the western parts of the nation, show spatial and temporal variations in the chemical characteristics of coastal waters. However, results from discrete surface samples represent an integrated result of water mass mixing and source strength of various rivers at different times. To this end, this study aimed to obtain trace metal distributions, along with other relevant hydrochemical parameters, to elucidate the influence of mountainous rivers under varying extents of environmental stressors and changing hydrology. Furthermore, although featuring a strong influence from economic development and mountainous rivers with temporal hydrological conditions, this is the first study to investigate trace metal distributions in the coastal zone in this region, except for a study of Pb and Ag in parts of the study area (Lee et al. 2013).

2. METHODS AND MATERIALS

2.1 Sampling and Sample Processing

Twenty-four near-shore locations along the west coast of Taiwan were sampled during November 19 - 23, 2004 and May 23 - 27, 2005 on board the R/V Ocean Researcher II. The sampling locations (Fig. 1) were generally evenly spaced, except for the central part where it was not possible as there is a wide region of very shallow water due to sand deposition (Liu et al. 2000). When a station is near the mouth of a major river, the station is designated with a D-prefix for easy comparison.

Rigorous clean sampling procedures were used throughout the sample collection and processing, as described in previous studies (Wen et al. 1996; Wen et al. 1999; Jiann and Presley 2002; Jiann et al. 2009). Polyethylene (PE) and PFA Teflon bottles (Nalgene) were used for sampling, storage and sample processing. All bottles were thoroughly acid washed before use. For water sampling, a sampling bottle (2 L PE) was attached to a titanium rod with a polypropylene holder, and lowered to a ~50 cm depth from the bow of the ship upon arrival at the sampling locations. As soon as the 2 L bottles were filled and retrieved, they were transported to a class-100 clean bench on board ship. Thereafter, seawater samples were filtered immediately through a 0.45 μm pore-sized acid-cleaned capsule filter (Osmonics, Westborough, MA, USA), acidified (pH < 2) by adding 2 mL of sub-boiled nitric acid (Seastar, Baseline grade) into each sample (1 liter). Another aliquot of an unfiltered sample (2 L) was vacuum-filtered through an acid-cleaned, pre-weighed 47 mm polycarbonate (PC, Nuclepore) membrane filter upon collection

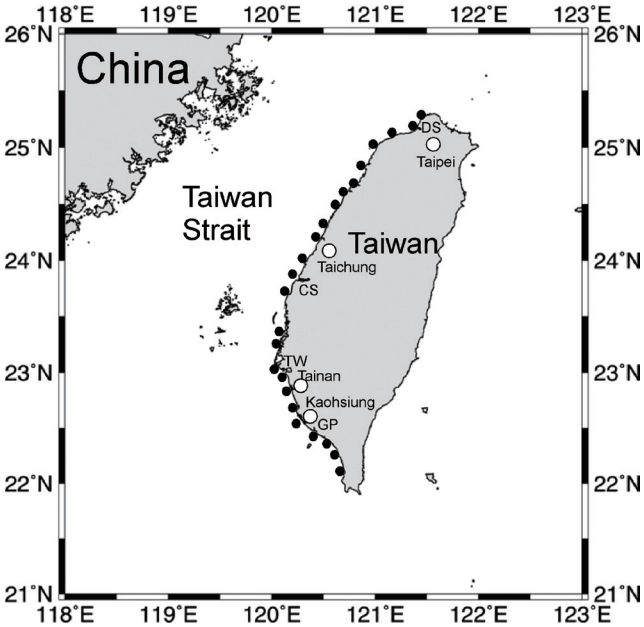

Fig. 1. Map showing major cities and important estuaries (DS, Danshuei; CS, Choshuei; TW, Tsengwen; GP, Gaoping) in Taiwan.

in a clean bench. Filters were stored frozen, and freeze dried, weighed to yield SPM concentrations.

Upon returning to the laboratory, the acidified samples in Teflon bottles were UV-irradiated (8 lamps of 15 W each) for 12 hours, in order to release trace metals from their complexes (Wen et al. 1996; Achterberg et al. 2001). Total dissolved metal concentrations were determined after preconcentrating metals in the filtered, acidified, and UV-irradiated samples onto Chelex-100 columns, as described previously (Jiann and Presley 2002). Particulate metal concentrations were obtained after SPM samples, retained on the filters, were digested using a combination of HNO_3, HF and boric acid (Lauenstein and Cantillo 1998). Measurement of trace metal concentration was performed on a graphite furnace atomic absorption spectrometer (GFAAS, Varian, SpectrAA 880Z), equipped with an auto-sampler and a Zeeman correction system, or a flame AAS (Perkin Elmer AAnalyst 400). The accuracy of the preconcentration and digestion procedures was confirmed by processing seawater and marine sediment standard reference materials (CASS-4 and MESS-3, from the National Research Council of Canada); the recoveries of metals determined for the two reference materials were 91 - 108%. Blanks were determined by processing Milli-Q water (for water samples) and blank filters (for particulate samples) together with the samples. Results showed that blanks were much lower (< 10% of the lowest value) than sample concentrations.

2.2 Supporting Data

At each sampling location, hydrological profiles were obtained with a Seabird CTD (SBE-11, General Oceanics) attached to a rosette sampler. Discrete water samples were collected using 10 L X-Niskin (General Oceanics) bottles, and aliquots of surface waters (2 m) were taken for nutrients (phosphate and silicate), and Chlorophyll *a* measurements. Phosphate and silicate concentrations were determined using the molybdenum blue spectrophotometric method on a flow injection analyzer (Grasshoff et al. 1983; Pai et al. 1990). Samples for Chlorophyll *a* determination were filtered immediately through GF/F filters (25 mm, Whatman), stored at -20°C, and later analyzed on a fluorometer (10-AU-005, Turner), following the method described by Strickland and Parsons (1972). Regional precipitation data and river discharge information are obtained from the Internet provided by the Central Weather Bureau and the WRA of Taiwan (ROC) (Central Weather Bureau 2013; Water Resources Agency 2013).

3. RESULTS

3.1 Hydrology and Nutrients

Coastal waters are direct recipients of terrestrial material that is discharged from rivers and estuaries and mixed with oceanic waters. Therefore, climatic and weather patterns that strongly influence river discharge and continental weathering can affect the amount of material being transported to the ocean. As described earlier (see Introduction) and shown in Fig. 2, the climate in Taiwan shows distinct precipitation and river discharge patterns. The overall trend is that most precipitation occurs between May and October, as was also the case in both 2004 and 2005. However, the

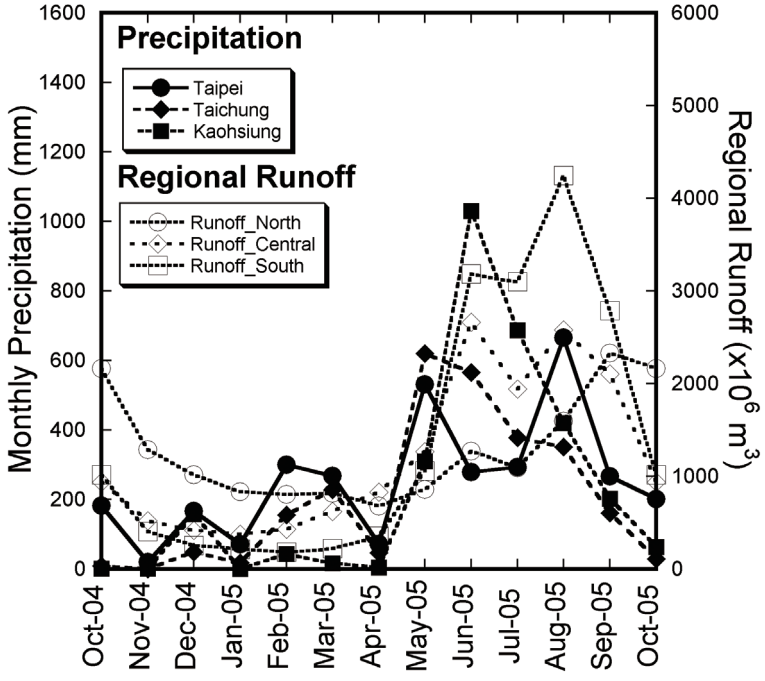

Fig. 2. Monthly precipitation of the four major representative regions in Taiwan during the study period and mean combined river discharges in the north, central and south regions (eastern Taiwan is excluded).

patterns are different in northern (Taipei) and southern Taiwan (Kaohsiung). The dry and wet seasons are clearly separated in southern Taiwan, with precipitation being sparse from November to April (NE monsoon season), and only when SW monsoon prevails from May to October that precipitation is elevated in southern Taiwan. This may have an effect on changing the water chemistry of river waters, as weathering and erosion rates vary with different precipitation rates and river discharge (Dosseto et al. 2006; Moon et al. 2007). As a result of the distinct regional precipitation pattern, the variability of river discharge in each of the rivers in northern Taiwan is less than that in the central and southern parts of the island. Varying precipitation rates are reflected in river discharges; they varied by a factor of 4 to ~66 for individual rivers in May 2005, compared with those in November 2004 (Water Resources Agency 2013). In the days preceding our May 2005 sampling dates, river discharges from rivers in Central Taiwan near 24°N registered > 1500 $m^3 s^{-1}$, resulting from high precipitation rates in this region (Taichung, Fig. 2).

3.1.1 Salinity

Large spatial and temporal variations in coastal water hydrology and physicochemical characteristics of those surface waters off Taiwan's west coast were observed from results of the two cruises conducted 6 months apart (Fig. 3a). Except for the relative low salinity (26) at Station B12 in May 2005, salinity in surface waters ranged from 32.9 to 34.5. Although salinity fell into a relatively narrow range, the average salinity was slightly lower in May 2005 (33.50, 33.81 if data of Station B12 is excluded, to 34.09 in November 2004). This indicates that terrestrial influence in the coastal water was more pronounced in May 2005. The common coastal ranges, relatively similar in salinity, yet with clearly distinguishable differences, also allowed for comparative assessment of the chemical composition in these near-shore waters having different influence from the land.

3.1.2 Suspended Particulate Matter (SPM)

Figure 3b shows the distribution of SPM concentrations along coastal surface waters. In November 2004, SPM concentrations were low (1-2 mg L^{-1}) at the southern locations (B23 - B27, south of 22.5°N), and remained relatively low (< 10 mg L^{-1}) near the TW River, where a large area is relatively flat. The highest SPM concentration (40 mg L^{-1}) was observed at Station B15, just south of the river mouth of the CS River, known for its high sediment yield (Dadson et al. 2003). Low SPM waters in the southern regime are reflecting the stronger open ocean characteristics, as salinity in this region was always higher (> 34) than in coastal waters in regions further north (e.g., in November 2004). The observed high SPM concentrations in coastal waters in the central parts of Taiwan's west coast may be linked to (1) the mountainous characteristics of most rivers in Taiwan, except for those in the south, where most Taiwan's wider plains and agricultural areas are located; (2) re-occurring landslides and mudslides in the central mountainous areas of Taiwan after typhoons or torrential rains that bring large amounts of mud into the rivers and result in high SPM concentrations in those rivers year-round (Kao et al. 2008; Liu et al. 2009). In May 2005, coastal waters contained, in general, even higher SPM than those of November 2004, especially in the central and southern regions. This may be due to (1) a greater extension of river plumes, as indicated by lower salinity; and, (2) higher precipitation (Fig. 2) causing higher sediment loads in the rivers. The highest SPM concentration (293 mg L^{-1}) observed in the May 2005 coastal water was from Station B12, which had a salinity of 26.09 during a time of an extremely high river discharge rate, as mentioned earlier. The level near Station D14 was also high (108 mg L^{-1}) off the CS River. In comparison with the November 2004 samples, in which there was only 1 sample with a SPM concentration, > 24 mg L^{-1}, the observation of high SPM levels from Station B11 - 15 in May 2005 further indicates the extent of the river plume's extension. Coastal waters of the northern Stations (B3 - B6) in May 2005 had lower salinity and higher SPM concentrations relative to those in November 2004, reflecting difference in the seasonal climate pattern and regional runoff between the northern region vs. that of the other regions, as stated earlier (Fig. 2).

3.1.3 Nutrients

As a result of varying river plume extensions between the two sampling times, nutrient concentrations showed large differences. Phosphate concentrations ranged from 0.06 to 2.05 μM in November 2004 and from 0.01 to 0.83 μM in May 2005 (Fig. 3c). Silicate concentrations were 1.64 - 8.01 μM in November 2004 and 1.16 - 41.67 μM in May 2005 (Fig. 3d). With stronger seawater characteristics (higher salinity) observed in the November 2004 samples, it was also found that concentrations of silicate were much lower than those in May 2005. Notably, one particularly high silicate concentration was found to be influenced by greater freshwater extension into the coast (B12, S = 26); it was found that in general silicate concentrations in May 2005 were comparable or higher than those in November 2004, thereby suggesting larger terrestrial fluxes in May 2005. However, a reverse trend was observed for phosphate. This indicates that although the fluxes of freshwater and terrestrial material transported from the island of Taiwan to the Taiwan Strait in May 2005 may be higher than those in November 2004, the influence of anthropogenic inputs to coastal waters near Taiwan in November 2004 was more pronounced. Thus, the distinct patterns of variations in silicate imply fluxes of natural weathering products (Moon et al. 2007), and that of phosphate reflect influence of human

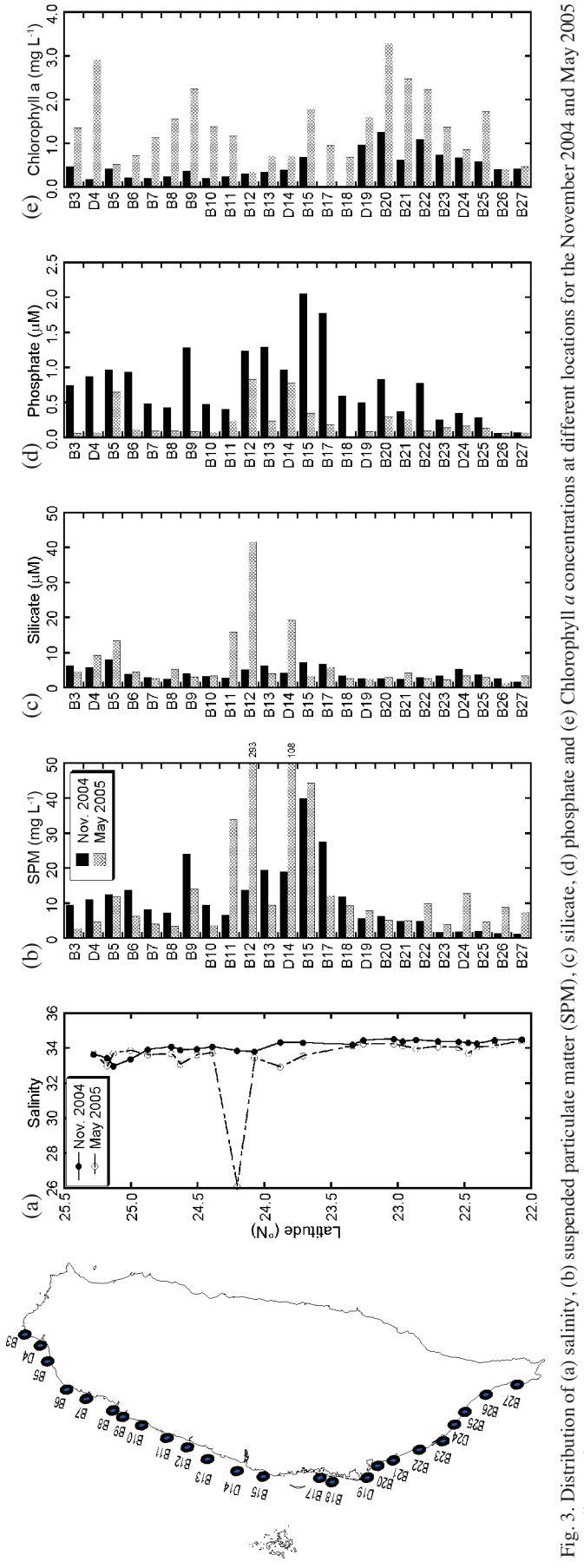

Fig. 3. Distribution of (a) salinity, (b) suspended particulate matter (SPM), (c) silicate, (d) phosphate and (e) Chlorophyll *a* concentrations at different locations for the November 2004 and May 2005 sampling cruises.

activities (Smith et al. 2003). This allows an assessment of the varying extent of natural and anthropogenic influences in coastal waters off Taiwan. Phosphate, as an indicator of environmental stress, has been reported to be exported at high concentrations from the Taipei metropolitan through the DS River Estuary to the coast (Wen et al. 2008).

3.1.4 Chlorophyll *a*

Distinct variations in Chlorophyll *a* concentrations were found both spatially and temporally in coastal waters (Fig. 3e), with concentrations ranging from 0.18 to 1.25 μg L^{-1} in November 2004, and from 0.34 to 3.28 μg L^{-1} in May 2005. In November 2004 Chlorophyll *a* concentrations in coastal waters displayed a pattern that showed a peak in the central-south area (Stations B17 - D19 near the TW River), and decreased toward both the north and south. Other elevated Chlorophyll *a* concentrations were sparsely observed. In contrast, Chlorophyll *a* concentrations in the May 2005 coastal waters showed no obvious spatial distribution patterns in which the levels were relatively close between northern and southern regimes. However, most high values are associated with larger rivers, where higher nutrient supplies are provided. Although May is in the early stage of southwest monsoon season, increased river discharges at this time of year appeared to stimulate phytoplankton growth. There was no obvious relationship between nutrient levels and Chlorophyll *a* concentrations at different locations.

3.2 Dissolved Metal Concentrations

As seen in the hydrochemical results above, in which large spatial and temporal variations were found within every parameter, variability was also observed for trace metals. Due to varying river discharges and seasonally varying precipitation, different and time-varying trace metal fluxes are transported from the rivers to the coast. Therefore, trace metal concentrations present in coastal waters depend on the seasonality of river discharge and the biogeochemical processes occurring in estuaries.

Considering all data from the two sampling times, dissolved trace metal concentrations in near-shore coastal waters were comparable with other regions of the world (Table 1), for example, coastal areas near the Celtic Sea (Cotté-Krief et al. 2002), Pearl River Estuary (Wang et al. 2012), eastern Gotland (Pohl et al. 2006), Gulf of Farallones (Hurst and Bruland 2008), Baja California (Sañudo-Wilhelmy and Flegal 1996), Hawaii (Bienfang et al. 2009), and the Gulf of Thailand (Censi et al. 2006), as well as continental shelves (the Gulf of Mexico and East China Sea) receiving large terrestrial inputs (the Mississippi and Changjiang Rivers, respectively) (Jiann et al. 2009; Wen et al. 2011). Concentration ranges observed were 27 - 289 pM for Cd, 1.32 - 66.26 nM for Cu, 2.39 - 86.87 nM for Ni, 0.08 - 0.44 nM for Pb, and 0.65 - 66.79 nM for Zn. The lower trace metal concentrations observed in this study are comparable with or slightly higher than those found in offshore and continental shelf waters (Table 1), suggesting that terrestrial sources of chemicals discharged from Taiwan had limited impact in its surrounding seas. As shown in Figs. 4a - e, "hot spots" for each element seem to be different and very few locations are characterized as major sources of all the elements determined in this study. The lowest concentrations were observed strictly in the southernmost locations.

3.3 Particulate Metal Concentrations

Figures 4f - j show the results of particulate metal

Table 1. Comparison of trace metal concentrations (pM for Cd and nM for the other elements) in coastal waters in different regions.

Region	Cd	Cu	Ni	Pb	Zn	Ref.
Celtic	53 - 368	2.95 - 6.64	1.69 - 9.08	0.158 - 0.431		Cotté-Krief et al. 2002
Pearl River	60 - 150	0.3 - 1				Wang et al. 2012
Eastern Gotland	100 - 120	8.2 - 8.9		0.02 - 0.2	6.5 - 10.1	Pohl et al. 2006
Gulf of Farallones	100 - 900	1 - 7			0.5 - 3.5	Hurst and Bruland 2008
Baja California	54 - 170	0.95 - 1.91	3.30 - 4.15			Sañudo-Wilhelmy and Flegal 1996
San Francisco Bay	193 - 594	1.1 - 10.6	4.5 - 16.3		0.6 - 11.1	van Geen and Luoma 1993
Hawaii		1.03 - 48.71	3.35 - 8.63	0.02 - 0.55	1.41 - 48.98	Bienfang et al. 2009
Gulf of Tailand		7.23 - 32.80	4.41 - 71.58		7.11 - 26.51	Censi et al. 2006
Gulf of Mexico	87 - 187	1.4 - 18.3	2.6 - 18.8			Wen et al. 2011
East China Sea	36 - 287	0.87 - 8.66	2.66 - 6.04			Jiann et al. 2009
W Coast off Taiwan	27 - 289	1.32 - 66.26	2.39 - 86.87	0.08 - 0.44	0.65 - 66.79	This study

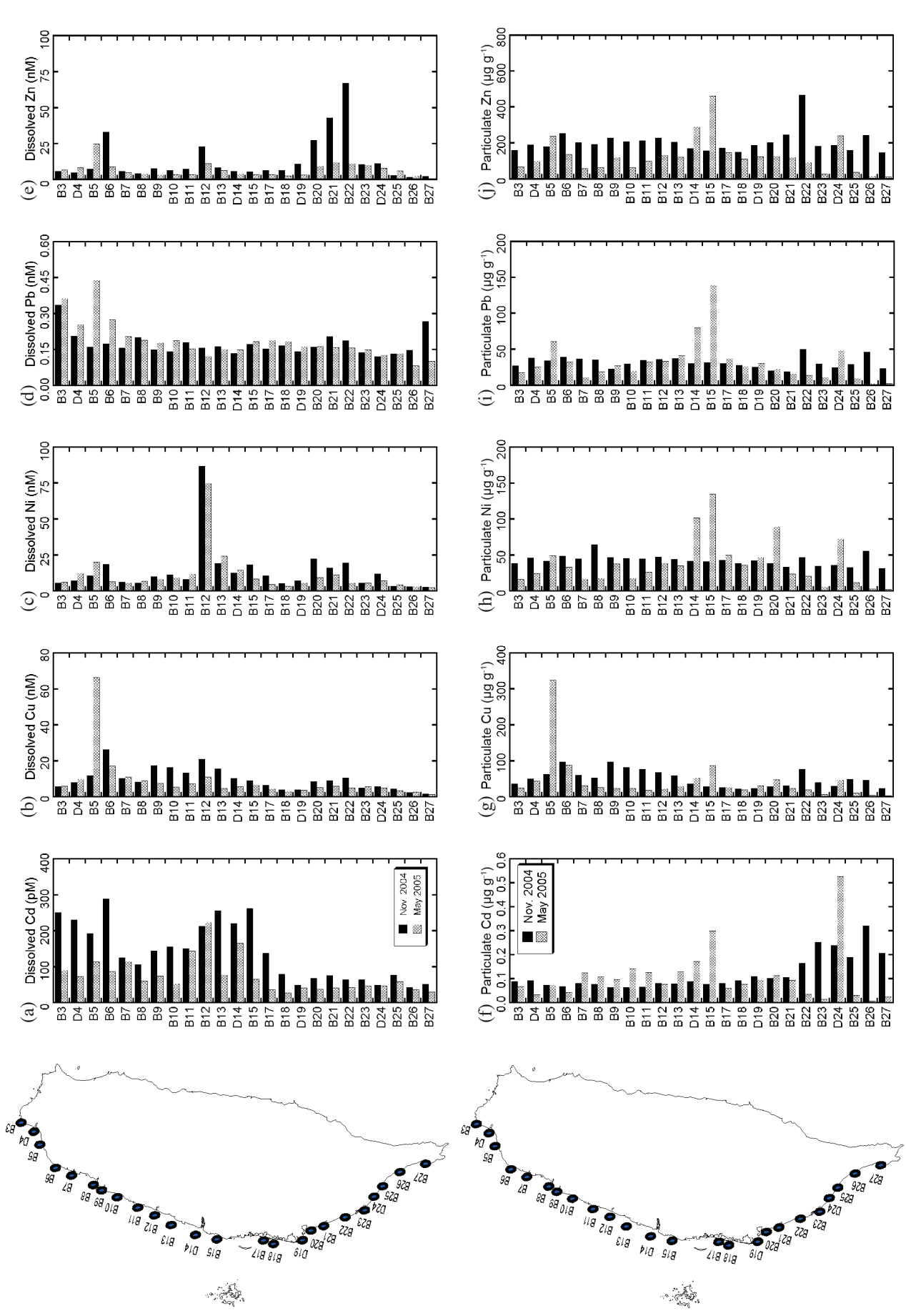

Fig. 4. Distribution of dissolved (a) Cd, (b) Cu, (c) Ni, (d) Pb and (e) Zn and particulate (f) Cd. (g) Cu, (h) Ni, (i) Pb and (j) Zn concentrations at different locations for the November 2004 and May 2005 sampling cruises.

concentrations (in SPM) in waters collected in November 2004 and May 2005. Large variations in particulate metal concentrations were observed, both temporally and spatially, for each of the elements studied. Particulate metal concentrations for the metals determined in this study at the two sampling times, respectively, were: Cd, 0.064 - 0.319 and 0.006 - 0.527 μg g^{-1}; Cu, 21.85 - 97.47 and 3.2 - 323.63 μg g^{-1}; Ni, 31.48 - 74.47 and 1.98 - 134.90 μg g^{-1}; Pb, 18.35 - 50.41 and 1.93 - 193.03 μg g^{-1}; Zn, 147.0 - 464.2 and 10.7 - 460.5 μg g^{-1}. When comparing particulate metal concentrations among sampling locations on the coast of western Taiwan, it is clear that at certain locations (e.g., Stations B5, D14, B15, and D24 in May 2005; Station B22 in November 2004) particulate metal concentrations were higher (> 2 times of the average of a cruise) for almost all elements determined. These locations are associated with known industry-intensive regions in Taiwan, or with rivers having higher freshwater discharges. Because Chlorophyll *a* concentrations in the coastal waters off Taiwan were not high, most particulate matter is believed to be mineral matter transported from the rivers, except for very low values at the southern locations in May 2005. This indicates that the SPM in waters from the southern locations were mostly composed of biogenic material, with their metal concentrations much lower than common crustal ranges. Further evidence is provided from particulate Fe concentrations determined in these SPM samples, which also showed low concentrations (~3000 μg g^{-1}, not shown) relative to those of common mineral material.

4. DISCUSSION

4.1 Terrestrial and Anthropogenic Influence

The intensity of terrestrial influence on water characteristics in coastal waters off Taiwan was seen in several parameters determined in this study. At locations near the major rivers of Taiwan where relatively larger terrestrial material was transported by these rivers, higher SPM and silicate concentrations and lower salinity were observed. Some exceptions to this coupling of SPM and silicate concentrations were found, mainly in northern locations, which are probably the result of estuarine modification, leading to different behavior of dissolved constituent (silicate) and particulate material (SPM). In contrast to lower salinity and higher SPM and silicate concentrations indicate the relative extent of terrestrial influence; the influence of anthropogenic inputs from Taiwan to its coastal waters is seen from results of phosphate and trace metal (Cu, Ni, Zn) concentrations. At locations near where there are major rivers (e.g., D4, B9, B10, D14, D19, D24), population centers (e.g., D4, B12, B22, D24) or intensive industrialization (B5, B6, B12, B22), phosphate and/or trace metal concentrations were found to be relatively elevated, compared to those at adjacent locations.

4.2 Sources of Trace Metals

Trace metal levels in coastal waters off western Taiwan in general were not higher than those in other regions in the world (Table 1), probably owing to a strong dilution effect of ocean waters. However, as shown in Fig. 4, a few "hot spots" can be identified to have elevated trace metal (Cu, Ni, and Zn) concentrations in coastal waters that had relatively constant salinity, except for one data point (station B12 in May 2005, salinity was 26.09). These hot spots are centered near station B5 - B6, B12, and B20 - B22. Station B5 - B6 are located off the coast near the Hsinchu high-tech industrial park and other heavily industrialized areas, where high metal discharges had led to the occurrence of "green oysters," caused by extremely high Cu concentrations in oyster tissue, in this coastal region (Lin and Hsieh 1999). Station B12 is located near Taichung Harbor that is also adjacent to "Taichung industrial park" hosting various industries, established since 2002. In the southwestern region, dissolved trace metal concentrations showed strong gradients with the peaks at or near Stations B20 - 22, an area known for having trace metal pollution concerns in the past (Han and Hung 1990; Jeng et al. 2000) originating from a metal scraping business carried out in its watershed (B20), and from the City of Tainan (B21) and Kaohsiung Harbor (B22). Estuarine waters collected in February 2009 in this region showed very high Cu (450 nM) and Ni (1000 nM) concentrations (Ho 2011), likely leading to the observed elevated Cu, Ni, and Zn concentrations in this region.

For Cd concentrations, there was clear variability in waters between the northern and southern regimes of Taiwan's west coast in which dissolved Cd concentrations were much higher in waters from the northern regime (Fig. 4a). This feature is distinctively different from the "spotty" high concentrations found for Cu, Ni, and Zn. This unique distribution pattern observed over a large area along the west coast off Taiwan could be attributed to desorption of Cd from SPM transported by mountainous rivers. In general, SPM concentrations were higher in the northern regime (Wei et al. 2009) and Cd tends to desorb from particles' surfaces in coastal waters when high Cl concentrations from seawater are present (Stumm and Morgan 1996), thus resulting in low particulate Cd concentrations (Fig. 4f).

Distribution patterns of Pb are distinctively different in coastal waters off Taiwan in which the concentrations were relatively constant at most sampling locations (except at Station B3 - B6, B26 - B27 with relatively low SPM concentrations, Figs. 4d, i). This can be explained by the particle-reactive nature of Pb, whereby Pb tends to adsorb onto sinking particles occurring in the upper parts of estuaries (Jiann et al. 2005; Jiann and Wen 2009; Tanguy et al. 2011), leaving low and constant dissolved Pb concentrations to be transported to the coastal region.

Distributions of individual trace elements (Fig. 4) show

that the sources of specific trace metals appear to be location dependent. There are poor correlations between pairs of hydrochemical parameters and trace metals. This suggests that, for the elements determined in this study, there were little co-emission of multiple chemicals. It is especially evident for Cd and Pb, as their distributions are largely controlled by different geochemical processes.

4.3 Temporal Variations of Trace Metal Concentrations in Coastal Waters

Owing to the distinctively different climatic patterns that lead to significantly different discharge rates in Taiwan's mountainous rivers (Chen et al. 2004), especially in the southwestern parts of Taiwan, the chemical composition in waters transported to the coasts also show temporal variability as shown in Table 2 where the results are compared for the two cruises. For each parameter, comparison between the two sampling times was made by calculating a ratio between November 2004 and May 2005 using average values. Results of the location with extreme salinity value (26, Station B12 in May 2005) are excluded in averages calculation for both sampling times so that comparative assessment can be made based on repeatedly sampled locations. Among the parameters reported in this study, average SPM, silicate and Chlorophyll *a* concentrations were higher in coastal waters in May 2005 when river discharge rates were higher. In contrast, average phosphate, Cd, Ni and Zn concentrations were higher in November 2004, while those for Cu and Pb have similar values for the two periods. The relatively low ratio (2004/2005) for Cu (1.07) is attributed to an extremely high value observed in May 2005 (Station B5, 66 nM), which would have been 1.55 if that high value is left out. For Ni, the ratios are nearly identical for both cruises, whether results from Station B12 are included or not to derive the average concentrations, because dissolved Ni concentrations at Station B12 were much higher than the other locations at both sampling times. The higher SPM concentrations observed in May 2005 resulted from high river flow reaching further into coastal waters, while higher silicate concentrations were caused by weathering rates increasing as precipitation rates increased. At the same time, a higher river discharge led to diluted chemicals of anthropogenic sources, thus lowering their concentrations, as seen for phosphate and some trace metals (Cd, Cu, Ni, and Zn) in the May 2005 samples. Higher SPM concentrations, resulting from higher river discharges, directly contributed to higher particulate metal loads (and export to the coast) in May 2005 because average metal concentrations in SPM were relatively similar between the two sampling periods except for Zn (Table 2). The ratio of average phosphate concentrations between the 2004 and 2005 samples was substantially higher (3.9) than the other parameters of anthropogenic origin (< 2.1). This could have been caused by higher

Table 2. Average and range for parameters determined in this study and the ratio between values for November 2004 and May 2005. Results of Station B12 were excluded when average values were derived because of the extreme salinity (26) found in May 2005.

	Dissolved[a]			Particulate[a]			Concentration in SPM[b]		
	November 2004	May 2005	2004/2005	November 2004	May 2005	2004/2005	November 2004	May 2005	2004/2005
SPM	10.82 (1.21 - 39.96)	14.43 (2.68 - 107.60)	0.75						
Phosphate	0.73 (0.06 - 2.05)	0.19 (0.01 - 0.77)	3.91						
Silicate	4.05 (1.64 - 8.01)	5.25 (1.16 - 19.17)	0.77						
Chlorophyll *a*	0.50 (0.18 - 1.25)	1.40 (0.41 - 3.28)	0.36						
Cd	136 (42 - 289)	68 (27 - 164)	2.02	8.3 (2.2 - 27.1)	20.1 (0.5 - 164.2)	0.41	0.12 (0.06 - 0.32)	0.11 (0.01 - 0.53)	1.12
Cu	9.26 (1.69 - 26.31)	8.69 (1.32 - 66.26)	1.07	8.58 (0.44 - 36.29)	12.05 (0.37 - 90.09)	0.71	49.22 (21.85 - 97.47)	43.54 (3.20 - 323.63)	1.13
Ni	10.36 (2.61 - 22.32)	8.40 (2.39 - 24.15)	1.23	7.96 (0.65 - 27.83)	16.97 (0.25 - 186.95)	0.47	42.41 (31.48 - 64.47)	37.74 (1.98 - 134.90)	1.12
Pb	0.173 (0.121 - 0.335)	0.188 (0.082 - 0.439)	0.92	1.62 (0.13 - 5.96)	4.15 (0.07 - 41.32)	0.39	31.15 (18.35 - 50.14)	31.27 (1.93 - 139.03)	1.00
Zn	12.57 (1.65 - 66.79)	6.47 (0.65 - 24.86)	1.94	31.9 (2.7 - 95.0)	47.7 (1.2 - 475.7)	0.67	203 (147 - 464)	123 (11 - 461)	1.65

Note: a: Concentration units are mg L^{-1} for SPM, μM for phosphate and silicate, μg L^{-1} for Chlorophyll a, pM for Cd, and nM for Cu, Ni, Pb and Zn.
b: Concentrations units are all in μg g^{-1} dry weight.

biological uptake as indicated by higher Chlorophyll *a* concentrations in May 2005. However, these Chlorophyll *a* concentrations were not exceptionally high and its biological uptake may not have affected trace metal concentrations to a significant extent relative to that of phosphate.

5. CONCLUSIONS

Because of the effects of dilution by ocean waters aided by strong currents in the Taiwan Strait, most coastal waters off western Taiwan did not appear to be affected by human activities, which is surprising. Still, temporal and spatial variations in trace metal distributions and hydrochemical parameters were found during two expeditions along the western coast of Taiwan. Developments of various industries and urbanization in different regions on the island led to differences in trace metal concentrations in coastal waters, as Cu, Ni, and Zn levels showed variability in coastal waters from different regions. The high sediment loads transported from mountainous rivers to Taiwan's coastal waters led to higher dissolved Cd concentrations resulting from desorption processes. In contrast, dissolved Pb concentrations were relatively constant over large areas owing to Pb removal within estuaries. Temporal variability was observed for hydrochemical parameters with greater transport of silicate and SPM, resulting from higher river discharges during the wet season. Concentrations of elements from anthropogenic sources (P, Cu, Ni, and Zn) were more elevated in coastal waters off Taiwan during the dry season.

Situated in the region with shifting monsoons over time, climate patterns in Taiwan show large spatial and temporal variability, resulting in the fluctuation of regional precipitation and river discharge rates. This leads to time-dependent river inputs of terrestrial and anthropogenic material into near-shore waters off the west coast of Taiwan. Consequently, among the chemical determined in this study, there exist very few correlations or relationships among parameters, suggesting water composition in the study area are influenced by too many differences in constitute elements, and insufficient commonality among the temporally variable sources of various chemicals.

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APPENDIX A

Information of sampling location and results of hydrochemical parameters for near-shore coastal waters collected off the west coast of Taiwan.

	Latitude (°N)	Longitude (°E)	Water [Depth (m)]	Sampling (Date)	Salinity	SPM (mg L^{-1})	Phosphate (μM)	Silicate (μM)	Chlorophyll *a* (μg L^{-1})
				November 2004					
B3	25.28	121.46	28	2004/11/19	33.63	9.37	0.74	6.24	0.46
D4	25.18	121.38	20	2004/11/19	33.43	11.04	0.87	5.70	0.18
B5	25.13	121.23	18	2004/11/19	32.96	12.54	0.96	8.01	0.42
B6	25.00	120.99	21	2004/11/19	33.36	13.79	0.93	3.83	0.21
B7	24.87	120.90	31	2004/11/19	33.91	8.18	0.48	2.84	0.20
B8	24.70	120.79	25	2004/11/20	34.08	7.16	0.43	2.38	0.23
B9	24.63	120.73	15	2004/11/20	33.90	23.98	1.28	4.04	0.36
B10	24.51	120.64	23	2004/11/20	33.93	9.43	0.47	3.23	0.20
B11	24.39	120.52	15	2004/11/20	34.09	6.50	0.40	2.64	0.23
B12	24.20	124.43	15	2004/11/20	33.85	13.65	1.23	5.09	0.31
B13	24.07	120.32	13	2004/11/20	33.79	19.47	1.29	6.16	0.34
D14	23.88	120.20	17	2004/11/20	34.34	18.82	0.96	4.19	0.39
B15	23.71	120.14	11	2004/11/20	34.32	39.96	2.05	7.14	0.68
B17	23.34	120.08	13	2004/11/23	34.20	27.41	1.77	6.67	NA*
B18	23.26	120.04	19	2004/11/23	34.46	11.77	0.59	3.34	NA
D19	23.03	120.05	11	2004/11/23	34.51	5.68	0.49	2.49	0.96
B20	22.96	120.14	16	2004/11/23	34.38	6.27	0.83	2.59	1.25
B21	22.86	120.17	13	2004/11/23	34.47	4.74	0.37	2.36	0.62
B22	22.70	120.23	13	2004/11/23	34.37	4.84	0.77	2.90	1.08
B23	22.54	120.28	21	2004/11/23	34.36	1.61	0.25	3.37	0.73
D24	22.47	120.41	56	2004/11/23	34.31	1.82	0.35	5.28	0.67
B25	22.40	120.50	18	2004/11/23	34.27	1.98	0.28	3.67	0.58
B26	22.27	120.63	22	2004/11/23	34.45	1.22	0.06	2.52	0.41

APPENDIX A

(Continued)

	Latitude (°N)	Longitude (°E)	Water [Depth (m)]	Sampling (Date)	Salinity	SPM (mg L^{-1})	Phosphate (μM)	Silicate (μM)	Chlorophyll *a* (μg L^{-1})
November 2004									
B27	22.07	120.68	21	2004/11/23	34.50	1.21	0.07	1.64	0.42
May 2005									
B3	25.28	121.48	31	2005/5/23	33.702	2.68	0.06	4.53	1.35
D4	25.18	121.37	25	2005/5/23	32.988	4.60	0.07	9.17	2.91
B5	25.13	121.23	15	2005/5/23	33.70	11.78	0.65	13.33	0.52
B6	25.00	120.99	26	2005/5/23	33.87	6.20	0.11	4.40	0.72
B7	24.87	120.91	36	2005/5/24	33.63	4.10	0.09	2.51	1.12
B8	24.70	120.79	25	2005/5/24	33.69	3.38	0.09	5.16	1.56
B9	24.63	120.75	14	2005/5/24	33.08	14.04	0.08	3.02	2.24
B10	24.51	120.65	24	2005/5/24	33.62	3.64	0.07	3.40	1.38
B11	24.34	120.53	15	2005/5/24	33.73	33.74	0.24	15.83	1.17
B12	24.21	120.44	11	2005/5/24	26.09	292.65	0.83	41.67	0.34
B13	24.07	120.33	12	2005/5/24	33.45	9.38	0.23	3.89	0.71
D14	23.88	120.21	16	2005/5/25	32.92	107.60	0.77	19.17	0.71
B15	23.71	120.15	11	2005/5/25	33.56	44.34	0.34	3.20	1.78
B17	23.34	120.09	15	2005/5/25	34.13	12.12	0.18	5.83	0.94
B18	23.25	120.05	19	2005/5/25	34.23	9.24	0.01	2.50	0.68
D19	23.02	120.06	12	2005/5/25	34.25	7.74	0.08	2.38	1.60
B20	22.96	120.14	15	2005/5/25	34.12	5.05	0.29	2.89	3.28
B21	22.86	120.18	13	2005/5/25	33.97	5.03	0.26	4.17	2.47
B22	22.69	120.24	11	2005/5/25	34.08	9.79	0.09	2.51	2.23
B23	22.54	120.29	19	2005/5/25	34.03	3.85	0.14	2.18	1.37
D24	22.47	120.42	45	2005/5/26	33.72	12.70	0.16	3.31	0.86
B25	22.40	120.31	16	2005/5/26	34.05	4.61	0.13	2.91	1.72
B26	22.27	120.63	24	2005/5/26	34.14	8.83	0.06	1.16	0.41
B27	22.07	120.69	22	2005/5/26	34.41	7.36	0.07	3.36	0.46

Note: NA: not available.

APPENDIX B

Dissolved and particulate trace metal concentrations of near-shore coastal waters sampled off the west coast of Taiwan.

	Cd		Cu		Ni		Pb		Zn	
	Dissolved (pM)	Particulate (g g^{-1})	Dissolved (nM)	Particulate (μg g^{-1})	Dissolved (nM)	Particulate (μg g^{-1})	Dissolved (nM)	Particulate (μg g^{-1})	Dissolved (nM)	Particulate (μg g^{-1})
November 2004										
B3	250.8	0.088	5.64	36.56	5.35	38.45	0.335	26.85	5.89	157.1
D4	230.8	0.092	7.90	49.98	7.18	45.62	0.205	38.04	4.64	189.7
B5	191.3	0.074	11.85	62.89	10.58	41.18	0.160	34.01	7.25	177.2
B6	288.9	0.066	26.31	97.47	18.48	48.18	0.174	38.77	32.76	252.6

APPENDIX B
(Continued)

	Cd		Cu		Ni		Pb		Zn	
	Dissolved (pM)	Particulate (g g^{-1})	Dissolved (nM)	Particulate (µg g^{-1})	Dissolved (nM)	Particulate (µg g^{-1})	Dissolved (nM)	Particulate (µg g^{-1})	Dissolved (nM)	Particulate (µg g^{-1})
November 2004										
B7	124.6	0.080	10.26	59.98	5.94	44.76	0.157	36.75	5.57	202.3
B8	105.6	0.076	8.14	51.46	5.57	64.47	0.200	34.98	4.16	190.6
B9	144.6	0.064	17.30	96.14	9.84	46.79	0.148	22.90	7.59	226.5
B10	155.3	0.064	16.23	81.60	11.06	45.54	0.142	29.14	6.43	205.8
B11	149.6	0.065	13.24	77.21	8.20	44.29	0.179	34.43	7.09	211.0
B12	213.1	0.080	20.88	67.80	86.87	47.07	0.155	35.52	23.00	225.7
B13	255.7	0.079	15.63	58.42	19.18	44.07	0.161	36.98	8.42	203.7
D14	220.2	0.089	10.18	36.39	12.47	41.13	0.133	30.13	5.74	169.4
B15	261.8	0.076	8.98	29.07	18.09	40.87	0.172	30.92	5.32	155.4
B17	138.3	0.080	6.52	25.69	10.35	42.74	0.153	30.17	6.20	169.7
B18	79.9	0.091	3.97	21.85	5.19	38.32	0.166	28.00	6.25	148.3
D19	48.7	0.110	3.81	23.47	7.26	42.18	0.142	24.94	10.55	186.1
B20	68.0	0.101	8.48	29.11	22.32	38.14	0.160	19.77	27.34	201.8
B21	74.8	0.105	9.00	30.45	16.03	32.85	0.204	18.35	42.84	244.0
B22	63.6	0.164	10.63	77.08	19.34	46.44	0.188	50.14	66.79	464.2
B23	63.9	0.252	4.91	40.11	5.32	34.17	0.138	29.76	10.52	181.3
D24	48.2	0.239	5.66	29.20	11.91	35.74	0.121	24.06	11.07	187.1
B25	75.5	0.189	4.26	48.92	3.23	32.57	0.130	28.51	2.75	158.2
B26	42.2	0.319	2.44	45.89	2.79	55.50	0.146	45.75	1.65	242.7
B27	50.5	0.207	1.69	23.02	2.61	31.48	0.267	23.12	2.36	147.0
May 2005										
B3	90.2	0.067	6.00	24.36	5.92	15.65	0.363	17.04	6.72	66.5
D4	72.4	0.032	9.77	43.12	12.16	24.15	0.254	25.12	8.05	97.9
B5	112.5	0.074	66.26	323.63	19.87	49.19	0.439	60.47	24.86	237.6
B6	86.3	0.042	17.09	87.58	6.17	33.10	0.274	32.14	8.84	136.1
B7	113.8	0.123	11.01	30.82	5.67	16.23	0.204	10.65	4.67	56.1
B8	59.8	0.107	8.90	25.30	6.65	17.80	0.189	18.47	3.85	60.2
B9	74.4	0.096	7.32	22.76	8.03	37.30	0.178	27.03	3.31	119.2
B10	52.8	0.142	5.31	23.30	8.95	17.67	0.186	19.50	3.51	61.4
B11	143.6	0.126	7.17	17.21	11.50	26.12	0.153	32.16	3.55	95.6
B12	223.1	0.076	10.98	22.00	74.76	38.52	0.121	33.38	11.15	131.2
B13	76.5	0.128	4.64	28.42	24.15	35.36	0.152	40.68	6.15	120.8
D14	164.4	0.171	5.63	53.21	14.33	101.98	0.149	79.58	3.95	289.1
B15	65.2	0.298	6.57	86.26	8.44	134.90	0.182	139.03	3.52	460.5
B17	36.1	0.060	4.40	25.52	4.51	49.45	0.187	36.77	3.40	145.0
B18	26.9	0.077	2.88	19.17	3.34	35.48	0.184	26.05	2.61	109.5
D19	41.0	0.095	3.47	30.62	5.65	46.90	0.162	29.93	3.06	122.6
B20	37.0	0.113	5.27	47.28	9.30	89.22	0.161	22.71	9.24	125.1

APPENDIX B
(Continued)

	Cd		Cu		Ni		Pb		Zn	
	Dissolved (pM)	**Particulate (g g^{-1})**	**Dissolved (nM)**	**Particulate (μg g^{-1})**	**Dissolved (nM)**	**Particulate (μg g^{-1})**	**Dissolved (nM)**	**Particulate (μg g^{-1})**	**Dissolved (nM)**	**Particulate (μg g^{-1})**
					May 2005					
B21	40.3	0.093	5.92	23.48	11.01	23.68	0.158	15.70	11.86	118.8
B22	41.9	0.035	4.78	19.31	5.60	20.22	0.156	13.55	10.87	89.9
B23	46.7	0.014	5.48	6.81	5.35	5.35	0.149	10.50	9.95	25.4
D24	46.3	0.527	4.73	45.94	7.02	72.36	0.126	48.29	7.96	238.4
B25	58.5	0.030	3.39	10.26	4.12	11.36	0.132	9.47	5.93	34.8
B26	36.2	0.006	2.64	3.95	3.06	2.63	0.082	2.32	2.42	14.4
B27	29.9	0.023	1.32	3.20	2.39	1.98	0.102	1.93	0.65	10.7