

Spatio-temporal trends of heavy metals and source apportionment in Tolo Harbour, Hong Kong

Kouping Chen · Jiu Jimmy Jiao

Received: 26 March 2009 / Accepted: 18 August 2009
© Springer-Verlag 2009

Abstract This study provided a picture of the spatial and temporal distributions of Cr, Co, Ni, Cu, Zn, As, Cd and Pb in bottom sediments of Tolo Harbour. The concentrations of the eight heavy metals differed significantly between sites due to the poor tidal flushing in Tolo Harbour. The levels of Cu, Zn, Cd and Pb were generally enriched in sediments from inner Tolo Harbour, while sediments from outer Tolo Harbour (Tolo Channel) had higher levels of Cr, Co and Ni. The redox sensitive element arsenic showed no distinct spatial pattern in Tolo Harbour. The decreasing levels of Cu, Zn, Pb and Cd in sediments with increasing distance from land demonstrated a typical diffusion pattern from land to the direction of sea. Two hot spots of Cu, Zn, Pb and Cd in sediments were located near Tai Po and Sha Tin new town, indicating that Cu, Zn, Pb and Cd were from land-derived sources. The sites with relatively high levels of Cr, Co and Ni in sediments were located in areas close to waste spoil in sea floor. The natural and anthropogenic inputs from Sha Tin and Tai Po to Tolo Harbour were mostly responsible for Cu, Zn, Cd and Pb enrichment in sediments from inner Tolo Harbour. The waste spoil in sea floor was believed to contribute to the Cr, Co and Ni in outer Tolo Harbour. The results of correlation coefficient between the eight heavy metals showed that Cu, Zn, Cd and Pb were strongly positively correlated, and Cr, Co and Ni were also significantly correlated with each other. The

best explanation of strong correlation was their similar source. As, however, is not well correlated with the other seven heavy metals. The average concentrations of Cu and Zn displayed general increasing trends from 1978 to 2006 in Tolo Harbour, while the mean levels of Cr and Pb displayed a substantial decrease from 1978 to 1987, then a slight increase after 1987. No distinct temporal trends of the concentrations of Ni and As were observed from 1978 due to the inconsecutive data. On the other hand, the increasing trends of Cr, Cu, Zn, Cd and Pb were observed since 1996.

Keywords Heavy metals · Sediment pollution · Temporal trend · Spatial distribution · Sources · Tolo Harbour

Introduction

Heavy metals in bottom sediments may be derived from the precipitation of dissolved materials, suspended matter and deposited sediments. The distributions of heavy metals in sediments are an important area of research in environmental studies because it not only reflects the quality of coastal water but also provides useful information on the transportation and fate of pollutants.

Tolo Harbour, an almost land-locked estuary located in the north-eastern territories of Hong Kong (Fig. 1), is most vulnerable to environmental pollution. The harbour is mainly fed by two major rivers (the Shing Mun and Lam Chuen Rivers), which pass through two outskirts towns, Sha Tin and Tai Po, respectively. The main water body of the harbour is approximately 16 km long and 3 km wide. It is connected to the Mirs Bay through Tolo Channel, which is a very narrow channel with a maximum width of only

K. Chen (✉)
Center for Hydrosociences Research, Nanjing University,
Nanjing, China
e-mail: kpchen@graduate.hku.hk

K. Chen · J. J. Jiao
Department of Earth Sciences,
The University of Hong Kong,
Hong Kong, China

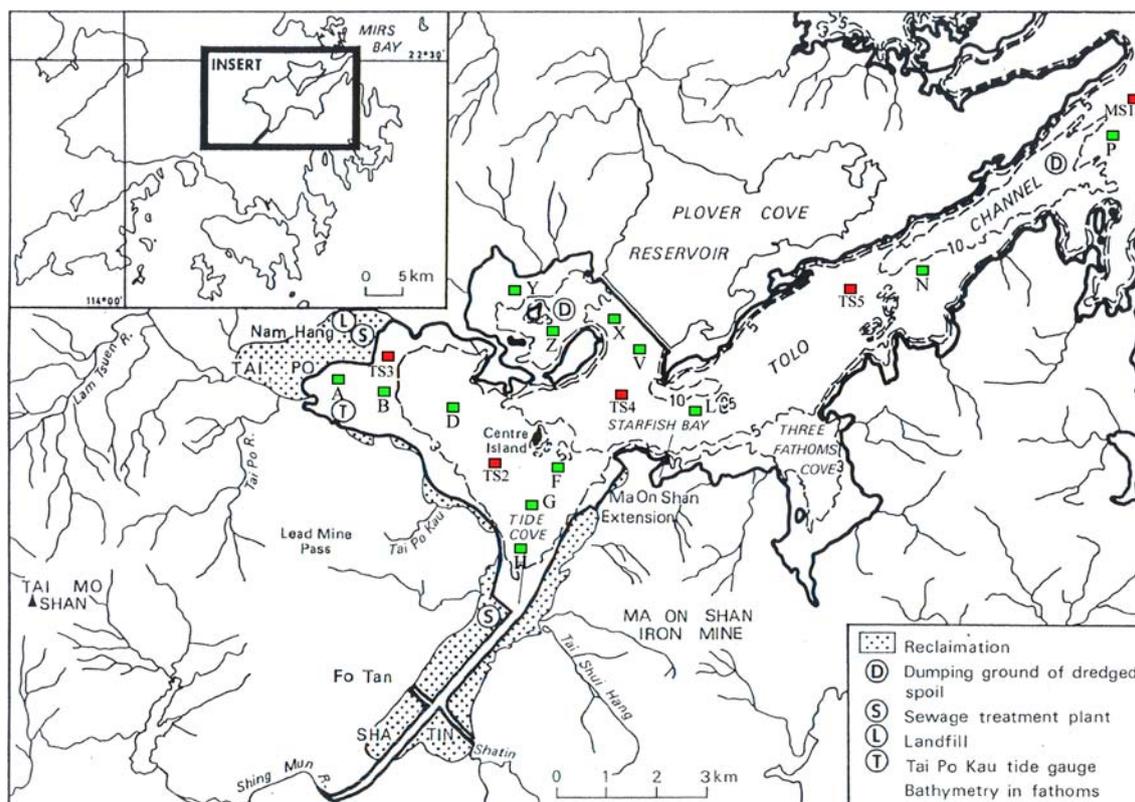


Fig. 1 Sketch map of Tolo Harbour and sampling locations (the base map is from Yim 1984)

1.3 km. The state of metal pollution in the harbour has attracted much public attention since the early 1970s. A few decades ago, the harbour was enclosed by rural areas. Since the middle 1950s, rapid growth of the manufacturing industries, population expansion and rapid urban development around this area has resulted in substantial pollution problems which seriously affected the environment in Tolo Harbour. In 1972, a major housing programme was undertaken by Hong Kong Government aimed at providing adequate housing. Most of the housing to be provided in new towns in New Territories was on land reclaimed from the sea. In the past few decades, land use changes around Tolo Harbour included: a large water reservoir scheme (Plover Cove Reservoir completed in 1968); two new satellite towns (Sha Tin new town and Tai Po new town); a large industrial estate (Tai Po Industrial Estate). Extensive reclamation to acquire land for the new towns and the industrial estate is common around Tolo Harbour.

The dense human population, anthropogenic activities such as reclamation, fish culture and the significantly increased domestic effluent disposal around harbours heavily impact the marine environment (Tanner et al. 2000). Up to now, about 20 papers describe the pollution problems in Tolo Harbour, and Trott and Fung (1973) seem the first to mention the pollution problems about Tolo

Harbour. Of the 20 published papers the state of metal pollution in Tolo Harbour has been assessed by determining metal levels in: (1) water column; (2) bottom sediment; (3) organisms. The objectives of the current study are to (1) investigate the current spatial distribution of heavy metals in sediment of Tolo Harbour and identify the possible pollution sources of heavy metals in Tolo Harbour, (2) study the temporal trend of heavy metal pollution in Tolo Harbour over the past few decades by reviewing the data published since 1978.

Materials and methods

Sample collection

The fieldwork was carried out on board the Electronic & Geophysical Services (Asia) Limited's Class Survey vessel from 18 to 20 November in 2006 and a differential Global Positioning System was used to locate the sampling sites. In total, there were 13 sampling sites in Tolo Harbour distributed from Inner Harbour to Tolo Channel (Fig. 1). All sediment samples were taken with a 0.05-m² van Veen grab. At every sampling site, six replicate grab samples were collected. One grab sample was used for analysis of

heavy metal concentration, total Kjeldahl nitrogen (TKN), total phosphorous (TP) and sediment particle size analysis, and it was stored in a plastic bag for later analysis. Five additional replicate samples were used for analysis of macrobenthos; the samples were sieved on a 0.5 mm sieve, and residues retained on the sieve were transferred into plastic container and fixed with 5% borax-buffered formalin followed by staining with 1% Rose Bengal for later sorting and identification. All sediment samples were stored in an ice box on board.

Laboratory analysis

Once arriving to the laboratory, all the sediment samples were stored in refrigerator at -20°C before further analysis. The subsamples for metal analysis were oven-dried at 80°C for 2 days, then ground using an agate mortar and pestle to obtain homogenous powder. The prepared samples were digested using an acid digestion (HNO_3 and HF) method and then determined by inductively coupled plasma-mass spectrometer (ICP-MS) (Model VG EX-CELL) at the University of Hong Kong.

Results and discussion

Spatial distributions of heavy metals and source apportionment

The results of eight heavy metal (Cr, Co, Ni, Cu, Zn, As, Cd and Pb) concentrations in bottom sediments collected from the 13 sampling sites in Tolo Harbour are presented in Table 1. Heavy metal concentrations differ significantly among these sites (Figs. 2, 3). The Cr concentration in

Table 1 Heavy metal concentrations (unit: mg kg^{-1}) in Tolo Harbour

Sites	Cr	Co	Ni	Cu	Zn	As	Cd	Pb
A	40.87	13.58	12.56	55.39	436.2	13.14	1.52	140.5
B	37.86	11.38	11.41	51.72	645	14.79	1.28	139
D	47.16	11.43	17.31	52.46	252.4	15.01	0.99	111.3
F	29.7	6.463	11.16	32.35	144.3	8.342	0.62	72.53
G	41.44	7.777	16.36	66.33	250.8	11.22	1.31	90.2
H	18.81	4.578	4.01	18.67	124.6	6.92	0.8	47.67
L	66.72	12.92	27.4	36.56	174.2	11.06	0.78	70.65
N	71.83	16.68	31.05	36.89	162.6	11.07	0.57	59.72
P	73.94	15.91	30.91	28.75	123.2	10.02	0.29	46.02
V	62.54	12.92	26.99	50.41	208.5	11.94	1.05	94.96
X	43.52	10.04	20.12	72.38	188.3	10.61	1.04	83.77
Y	8.39	3.013	10.2	8.19	53.19	2.059	0.23	22.82
Z	76.31	9.992	18.01	10.39	51.52	12.97	0.08	34.45

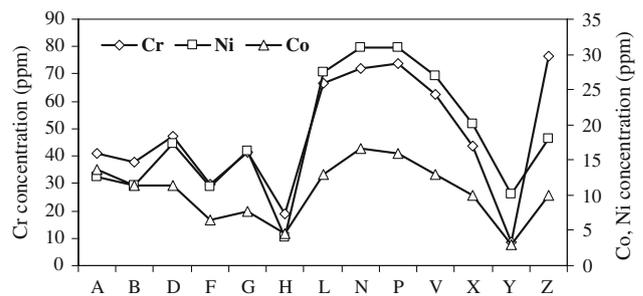


Fig. 2 Spatial distributions of Cr, Co and Ni in Tolo Harbour

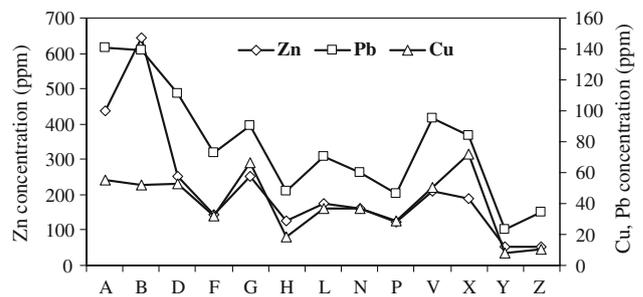


Fig. 3 Spatial distributions of Cu, Pb and Zn in Tolo Harbour

bottom sediments ranges between 8.39 and 76.31 mg kg^{-1} with highest concentration in site Z. The Co and Ni concentrations in bottom sediments range between 3.01 and 16.68 mg kg^{-1} , 4.01 and 31.05 mg kg^{-1} , respectively, with highest concentrations occurring both in site N. It can also be found from Fig. 2 that the general concentrations of Cr, Co and Ni in sediments collected from inner Tolo Harbour (sites A, B, D, H, G and F) show lower levels than those collected from Tolo Channel (sites L, N and P). This means that the main source of high Cr, Co and Ni in Tolo Harbour derives from neither the new towns (Tai Po and Sha Tin) nor Tai Po Industrial Estate. Therefore, it can be inferred that the domestic and industrial effluent discharge from the nearby population is not the main source contributing Cr, Co and Ni in bottom sediments. According to the investigation conducted by Chan (1992) that the Co concentration in sediments collected from Mirs Bay was higher ($7.85\text{--}9.41 \text{ mg kg}^{-1}$) than that in Tolo Harbour ($3.04\text{--}6.89 \text{ mg kg}^{-1}$). Owen and Sandhu (2000) found that Ni concentration varied between 4 and 10 mg kg^{-1} in inner Tolo Harbour, and increased to 17 mg kg^{-1} towards Tolo Channel. Also Environmental Protection Department, Hong Kong Administrative Region (EPD, HKSAR) routinely measures the levels of heavy metals in sediments in Tolo Harbour. Table 2 lists the data of heavy metal concentrations in bottom sediments in the annual report of EPD from 1996 to 2006. The locations of the five stations TS2, TS3, TS4, TS5 and MS17 can be seen in Fig. 1. From Table 2, it can be obviously observed that the mean concentrations of Cr and

Table 2 Heavy metal concentrations (unit: mg kg⁻¹) at five stations in Tolo Harbour from 1996 to 2006 by EPD

	TS2	TS3	TS4	TS5	MS17
Cr					
2003–2006	27	24	25	35	35
1999–2003	29	24	26	35	36
1996–2000	27	23	31	32	35
Ni					
2003–2006	17.6	15	17	26	27
1999–2003	18	15	17	24	26
1996–2000	17	14	20	23	25
Cu					
2003–2006	44	43	27	23	16
1999–2003	50	43	27	23	17
1996–2000	44	51	28	21	16
Zn					
2003–2006	206	242	155	130	99
1999–2003	212	211	142	123	95
1996–2000	170	190	140	120	97
As					
2003–2006	8.5	9	8	6.3	6.3
1999–2003	9.6	10.1	8.5	7.2	7.3
1996–2000	8.6	9.1	8.3	7.1	7.6
Pb					
2003–2006	86	102	69	53	42
1999–2003	89	99	68	54	45
1996–2000	79	91	65	53	44

Ni in sediments collected from Tolo Harbour (TS2, TS3, TS4 and TS5) through the 10 years always display low levels than those collected from Mirs Bays (MS17). Furthermore, the levels of Cr and Ni in sediments collected from Tolo Channel (TS5) are always higher than those collected from Inner Tolo Harbour (TS2, TS3 and TS4). These phenomena have again confirmed that the main source of Cr, Ni and Co in Tolo Harbour is not associated with the increased anthropogenic activities in Tai Po and Sha Tin. However, almost no previous research gave a clear explanation of the higher levels of Cr, Co and Ni in Tolo Channel compared to inner Tolo Harbour.

According to Yim (1984), there were two waste spoil sites in sea floor of Tolo Harbour (Fig. 1). The high levels of Cr, Co and Ni in bottom sediments was investigated by EPD from 1996 to 2006; Chan (1992) and this survey all occurred in the vicinity of the two waste spoil sites. Therefore, it is most likely that these waste spoils should be responsible for high levels of Cr, Co and Ni in Tolo Harbour. However, some previous researchers (Lam et al. 1997; Owen and Sandhu 2000; Zhou et al. 2007) simply ascribed the Co, Cr and Ni contamination in Tolo Harbour to anthropogenic pollutants from Tai Po and Sha Tin.

Contrary to Cr, Co and Ni, the levels of Cu, Zn, Pb and Cd in bottom sediments are enriched in inner Tolo Harbour near Tai Po and Sha Tin rather than Tolo Channel (Fig. 3). The Cu concentration in bottom sediments ranges from 8.19 to 72.38 mg kg⁻¹, with highest concentration occurred in site X. The concentrations of Pb, Zn and Cd in bottom sediments range between 22.82 and 140.5, 51.52 and 645, 0.08 and 1.52 mg kg⁻¹, respectively. The maximum concentrations of Pb and Cd both occur in site A. Site B has the highest Zn levels. It can be observed from Table 1 that the general levels of the eight heavy metals in bottom sediment (sites A, B and D) near Tai Po are higher than those (sites H, F and G) near Sha Tin. It should be noted that site H displays the lowest levels of the eight heavy metals comparing to other sites in Inner Harbour. It can be seen from Fig. 1 that site H locates in the outfall of the Shi Mung River and near the reclamation areas. One explanation for the relatively low concentrations of the eight heavy metals in bottom sediments collected from site H is that the mixing of river flow (Shing Mun River) and the coastal currents is not favourable for the deposition of heavy metals from solution (Ip et al. 2007). Also the reduced local water area due to land reclamation along Shing Mun River is obvious and this in turn modifies the tidal range and currents. Park and Park (1998) documented that a reduction in the area of an enclosed water body due to land reclamation may increase wave amplitude and current velocities. According to Zhao et al. (2006) when the tidal current is increased, the sediments will be re-suspended. This may explain why the eight heavy metals in sediment collected from site H display the lowest levels comparing to other sites in Inner Harbour.

Yim (1984) documented that natural inputs of Pb and Zn derived from the breakdown of sulphide minerals such as galena and sphalerite were ubiquity in the mineralised volcanic rocks in the Tai Mo Shan area (Fig. 1). These sulphide minerals were also present in the alluvial boulders and cobbles of the Lam Tsuen Valley, which located upstream of Tai Po. The mean concentrations of Pb and Zn found in the stream sediments were both higher than the sea floor sediments of Tolo Harbour (Lee 1981). Transportation of Pb and Zn in solution by the Lam Tsuen River into the harbour via Tai Po was confirmed by the study of stream water samples by Lee (1981). The highest levels of Pb and Zn in sediments collected from sites A and B should be attributed to the enrichment of the two metals in the bottom sediments of Lam Tsuen River. The enriched levels of Pb and Zn in bottom sediments of Lam Tsuen River were mainly due to the natural input from the sulphide minerals. In addition to the natural input of heavy metals from the sulphide minerals, anthropogenic sources of heavy metals associated with urban and industrial activities were also important inputs to marine sediments, which should also be

responsible for the highest Pb and Zn levels in sites A and B. Rapid urbanisation and industrialisation from 1950s around Tolo Harbour resulted in large quantities of both solid and liquid chemical wastes and domestic effluent enriched in heavy metals to enter the harbour (Wong et al. 1980; Yim and Leung 1978; Owen and Sandhu 2000; Zhou et al. 2007). Copper sulphate, extensively used to treat diseases in fishes cultured in floating cages, may contribute high copper concentration observed in this area (Chu et al. 1990).

The eight heavy metals observed in sites V and X locating west of Plover Cove Dam show abnormally high levels in regard of their locations. The reversed phenomena are best explained by the dumping of dredged spoil and wastes originating from the inshore areas of Sha Tin and Tai Po and construction of the Plover Cove Reservoir (Yim and Leung 1978).

Generally, there is a decline in Cu, Zn, Pb and Cd with distance from Tai Po new town and its associated industrial estate (Fig. 3). The four heavy metal concentrations were higher in the landward sites, decreasing gradually seaward. The levels of the four heavy metals (Cu, Zn, Pb and Cd) are 55.39, 436.2, 140.5 and 1.52 mg kg⁻¹, respectively, in site A near Tai Po. These values decline to 36.56, 174.2, 70.65 and 0.78 mg kg⁻¹, respectively, in site L and drop further to 28.75, 123.2, 46.02 and 0.29 mg kg⁻¹ in site P, which is located in Tolo Channel. The spatial distribution patterns of the four heavy metals are supported by previous research in Tolo Harbour (Chu et al. 1990; Lam et al. 1997; Owen and Sandhu 2000). It has been documented by Chu et al. (1990) that higher concentrations of metals, especially Cu, Zn and Cd, were found in sediments from inner Tolo Harbour (20–150, 25–180, 3–7 mg kg⁻¹) than in sediments from Tolo Channel (5–25, 25–35, 3–4 mg kg⁻¹). Owen and Sandhu (2000) reported similar spatial patterns of Cu, Pb and Zn in Tolo Harbour. It seemed that most of the sites with higher Cu, Pb, Zn and Cd concentrations were found closer to the landward region, implying that the contamination of the four heavy metals might be derived from the land source. Li et al. (1986) noted that currents in Tolo Harbour were weak, with speeds of 0.2–0.25 m s⁻¹ in Tolo Channel and velocities of only a few cm s⁻¹ occurring in Tolo Harbour. Consequently, tidal flushing and pollutant dispersal are expected to be weak, thus favouring deposition of heavy metals within Tolo Harbour before reaching Tolo Channel. Therefore, the concentrations of the four heavy metals were largely determined by the distance to the element sources. This explains why the Cu, Pb, Zn and Cd concentrations in site A were the highest, while the levels of Cr, Co and Ni in sediments collected from Tolo Channel were higher than those from inner Tolo Harbour near Tai Po and Sha Tin (Fig. 2). From Table 2, it could also found that the levels of Cr and Ni in sediments collected from Tolo Channel (TS5) were always higher than

those collected from Inner Tolo Harbour (TS2, TS3 and TS4) from 1996 to 2006. Yim (1984) documented that there were two waste spoil sites in sea floor of Tolo Harbour (Fig. 1). It was found that the sediments with higher Cr, Co and Ni levels were located near the waste spoil sites. Therefore, it was likely that the waste spoils should be responsible for higher levels of Cr, Co and Ni in Tolo Channel. The data of arsenic concentration was not discriminating enough to judge the spatial pattern through Tolo Harbour.

Correlations between the eight heavy metals

The correlation coefficient of the eight heavy metals is shown in Table 3. It can be seen from Table 3 that the three heavy metals Cr, Co and Ni, which show higher levels in sediments from outer Tolo Harbour (Tolo Channel) than those from inner Tolo Harbour, are significantly correlated (Cr and Ni: 0.86; Cr and Co: 0.83; Ni and Co: 0.81). The strong positive correlation between Cr, Co and Ni indicates that they mostly came from the same source, while the four heavy metals Cu, Zn, Pb and Cd, which show decreasing levels with distance from Tai Po and Sha Tin, are also strongly correlated (Zn and Pb: 0.90; Cd and Pb: 0.90; Cu and Pb, 0.77; Cu and Cd: 0.84; Cu and Zn: 0.58), indicating that these heavy metals probably have similar provenance. The redox sensitive element As, showing no spatial pattern in Tolo Harbour, is not correlated significantly with the other seven metals, indicating different source or different chemical character with other seven metals. The correlation coefficient of the eight heavy metals and the spatial distribution pattern of these metals presented above supported each other.

Trend of heavy metal level from 1978 to 2006

Metal concentrations in sediments of Tolo Harbour from 1978 to 2006 are summarised in Table 4. Generally, it is very

Table 3 Pearson correlation coefficients among eight heavy metals in Tolo Harbour (*n* = 13)

	Cr	Co	Ni	Cu	Zn	As	Cd	Pb
Cr	1.00							
Co	0.83**	1.00						
Ni	0.86**	0.81**	1.00					
Cu	0.07	0.33	0.16	1.00				
Zn	-0.12	0.27	-0.21	0.58	1.00			
As	0.57	0.65	0.29	0.56	0.60	1.00		
Cd	-0.21	0.14	-0.21	0.84**	0.77**	0.49	1.00	
Pb	-0.04	0.33	-0.12	0.77**	0.90**	0.71	0.90**	1.00

** Correlation is significant at the 0.01 level

Table 4 Temporal variation of heavy metal concentrations (unit: mg kg⁻¹) in Tolo Harbour from 1978 to present

Sources	Cr	Ni	Cu	Zn	As	Cd	Pb
Yim and Leung (1978)	38 (6–74)		26 (10–48)	137 (53–216)	14 (2–36)		108 (60–177)
EPD (1987)	0–50		<50	0–200		0.21–0.80	<50–100
EPD (1995)	24–49	10–24	15–59	100–>150			>60
Owen and Sandhu (2000)	14–30	4–17	21–84	69–270			48–144
EPD (1996–2000)	29.6 (16–42)	19.8 (10–39)	32 (10–97)	143.4 (67–270)	8.1 (4.5–13)	0.28 (<0.1–0.6)	66.4 (36–110)
EPD (1999–2003)	30 (18–40)	20 (15–26)	32 (17–50)	156.6 (95–212)	8.54 (7.2–10.1)	0.36 (0.1–0.6)	71 (34–130)
EPD (2003–2006)	32.2 (20–41)	21 (8–31)	34.3 (18–60)	183.2 (75–380)	7.95 (5.5–11)	0.45 (0.2–0.8)	77.5 (44–130)
This study (2006)	47.6 (8.4–76.3)	18.3 (4–31)	40.0 (8.19–72.38)	216.5(51.5–645)	10.7 (2.1–15.0)	0.81 (0.08–1.52)	78.0 (22.8–140.5)
Ratio of increment (%)	25.3	NA	14.0	58.0	NA	NA	–27.8

difficult to compare metal concentrations determined by different previous researchers. This is mainly due to two important factors: (1) the size fraction analysed; (2) the analytical method used. For example, Yim and Leung (1978) analysed the sediments of size fraction smaller than 170 μm while Owen and Sandhu (2000) analysed the bulk sediments. Yim and Leung (1978), Owen and Sandhu (2000) determined metal concentrations in sediments by atomic absorption spectrophotometry, while both heavy metal concentrations in sediments by EPD, HKSAR and the heavy metals in the current study were determined by ICP-MS. The data of the present study and EPD were based on analysis of bulk sediments. Both the size of the sediment fraction and the method of chemical analysis of the current study are identical to those in the previous studies by EPD. Theoretically EPD and current study based on analysis of bulk sediments should yield lower concentrations than those by Yim and Leung (1978) that focused on smaller fractions. In spite of this reason, the general increasing trends of Cu and Zn from 1978 to 2006 can be still observed from Table 4. For example, in 1978 the mean level of Zn was 137 mg kg⁻¹, while to 2006 it increased to 216.5 mg kg⁻¹. According to Owen and Sandhu (2000), the development of Sha Tin and Tai Po new towns had a major impact on the water quality of Tolo Harbour through input of sewage and reclamation. Also over the past few decades the hydrodynamic conditions of Tolo Harbour has been physically altered by man through construction of dam at Plover Cove and reclamation to establish new towns. The temporal increasing trends of Cu and Zn from 1978 to 2006 could be attributed to a generalised increment of inputs from both domestic and industrial sources associated with increased urban and industrial activities. It can be seen from Table 4 that Cr and Pb displayed a substantial decrease from 1978 to 1987, then increased slightly afterwards. Apart from analysis of bulk sediments which should yield lower concentrations in 1987, another important reason for the significant decrease of Pb

and Cr from 1978 to 1987 was the strictly cessation of direct discharge of iron mine tailing from Ma On Shan Mine to Tolo Harbour since 1976 (Yim and Leung 1978). The general trends of Ni, As and Cd concentrations from 1978 to present were not evident due to the inconsecutive data in some years. Considering the inconsecutive data, it is difficult to draw a satisfactory conclusion on the temporal trend of heavy metal concentration in Tolo Harbour from 1978 to present. However, from 1996 to 2006 the levels of heavy metals in Tolo Harbour were consecutive (Table 2) and comparison can be made to display the temporal trend over this period. Figure 4 shows the average concentrations of Cu, Zn, Pb, Cr, Ni, As and Cd in bottom sediments within Tolo Harbour from 1996 to 2006. It can be observed from Fig. 4 that Cu, Zn, Pb, Cr and Cd display an overall increasing trend over this period, while the average levels of Ni and As over this period changed irregularly, and it is not adequate to establish valid temporal trend for Ni and As based on the present database.

Conclusion

The spatio-temporal distribution of eight heavy metal concentrations in sediments of Tolo Harbour and their possible sources are examined in the present study. Of the eight heavy metals investigated, Cu and Zn displayed general increasing levels from 1978 to 2006 in Tolo Harbour, while Cr and Pb displayed significant decrease from 1978 to 1987, then increased slightly afterwards. No distinct temporal trends of Ni, As and Cd in Tolo Harbour can be observed from 1978 due to the inconsecutive data. Considering data continuity, the temporal trends of heavy metals in Tolo Harbour from 1996 to 2006 were evaluated. In general, the average levels of Cr, Cu, Zn, Cd and Pb in Tolo Harbour displayed increasing trends, while Ni and As did not show obvious temporal trends over this period.

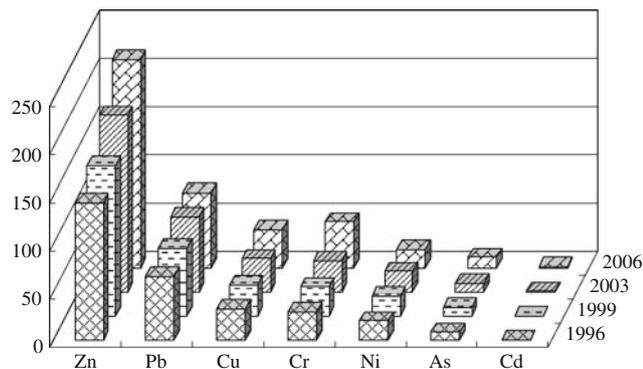


Fig. 4 Temporal trends of heavy metal concentrations from 1996 to 2006 in Tolo Harbour

As for the current spatial distribution, according to the present investigation, two hot spots are identified within Inner Harbour near Tai Po and Sha Tin with regard to Cu, Zn, Pb and Cd. Sediments from Tolo Channel have relatively low concentration in the four heavy metals. The levels of the four heavy metals showed general decreasing trend with increasing distance from land. This implicates that the four heavy metals are land-derived and are mainly accumulated in the Inner Harbour without reaching Tolo Channel due to the poor tidal flushing and water exchange capacity of Tolo Harbour. Both natural input from sulphide minerals in Tai Mo Shan, iron mine tailing from Ma On Shan and anthropogenic sources such as domestic and industrial sewage from the nearby population should be responsible for Cu, Zn, Pb and Cd enrichment in sediment from Inner Harbour.

The levels of Cr, Co and Ni in sediments from inner Tolo Harbour are relatively low comparing to outer Tolo Harbour, indicating that these metals are not from land-derived source. The three heavy metals are enriched in sediments from sites N, P X and Z, which are located near waste spoil sites in sea floor of Tolo Harbour. It is believed that the high levels of the three metals in sediments from outer Tolo Harbour should be attributed directly to the waste spoil in sea floor of Tolo Harbour.

Acknowledgments This study was supported by the grant from the Research Grants Council of Hong Kong (HKU 7028/06P). The authors wish to thank Shengyan Tian, Robin Liu and Phil Xu for assistance in field sampling.

References

Chan HM (1992) Heavy metal concentrations in coastal seawater from Tolo Harbour, Hong Kong. In: Morton B (ed) *The marine flora and fauna of Hong Kong and southern China III*. Hong Kong University Press, Hong Kong, pp 621–628
 Chu KH, Cheung WM, Lau SK (1990) Trace metals in bivalves and sediments from Tolo Harbour, Hong Kong. *Environ Int* 16:31–36

EPD (1988) *Marine water quality in Hong Kong. Results for 1987* from the marine monitoring program of the Environmental Protection Department. Monitoring Section, Water Policy and Planning Group, Environmental Protection Department, Hong Kong Government, Hong Kong
 EPD (1996) *Marine water quality in Hong Kong. Results for 1995* from the marine monitoring program of the Environmental Protection Department. Monitoring Section, Water Policy and Planning Group, Environmental Protection Department, Hong Kong Government, Hong Kong
 EPD (2000) *Marine water quality in Hong Kong. Results for 1996 to 2000* from the marine monitoring program of the Environmental Protection Department. Monitoring Section, Water Policy and Planning Group, Environmental Protection Department, Hong Kong Government, Hong Kong
 EPD (2003) *Marine water quality in Hong Kong. Results for 1999 to 2003* from the marine monitoring program of the Environmental Protection Department. Monitoring Section, Water Policy and Planning Group, Environmental Protection Department, Hong Kong Government, Hong Kong
 EPD (2006) *Marine water quality in Hong Kong. Results for 2003 to 2006* from the marine monitoring program of the Environmental Protection Department. Monitoring Section, Water Policy and Planning Group, Environmental Protection Department, Hong Kong Government, Hong Kong
 Ip CCM, Li XD, Zhang G, Wai OWH, Li YS (2007) Trace metal distribution in sediments of the Pearl River Estuary and the surrounding coastal areas, South China. *Environ Pollut* 147(2):311–323
 Lam MHW, Tjia AYW, Chan CC, Chan WP, Lee WS (1997) Speciation study of chromium, copper and nickel in coastal estuarine sediments polluted by domestic and industrial effluents. *Mar Pollut Bull* 34(11):949–959
 Lee S (1981) *Lam Tsuen River Survey*. Environmental Protection Agency Report. No.20/40/18 II, Hong Kong Government
 Li CW, Lee JHW, Cheung YK (1986) Mathematical model study of tidal circulation in Tolo Harbour, Hong Kong: development and verification of a semi-implicit finite element scheme. *Proc Instn Civ Eng* 81(2):569–592
 Owen RB, Sandhu N (2000) Heavy metal accumulation and anthropogenic impacts on Tolo Harbour, Hong Kong. *Mar Pollut Bull* 40(2):174–180
 Park JW, Park SS (1998) Hydrodynamic modeling of tidal changes due to land reclamation in an open-ended harbor, Pusan, Korea. *J Environ Sci Health A* 33(5):877–890
 Tanner PA, Leong LS, Pan SM (2000) Contamination of heavy metals in marine sediment cores from Victoria Harbour, Hong Kong. *Mar Pollut Bull* 40(9):769–779
 Trott LB, Fung AYC (1973) Marine pollution in Hong Kong. *Mar Pollut Bull* 4(1):13–15
 Wong MH, Ho KC, Kwok TT (1980) Degree of pollution of several major streams entering Tolo Harbour, Hong Kong. *Mar Pollut Bull* 11:36–40
 Yim WWS (1984) Geochemical mapping of bottom sediments as a aid to marine waste disposal in Hong Kong. *Conserv Recycl* 7(2–4):309–320
 Yim WWS, Leung WC (1978) Sedimentology and geochemistry of sea-floor sediments in Tolo Harbour, Hong Kong—implications for urban development. In: Whiteside PGD (ed) *The role of geology in urban development*. Geol Soc Hong Kong Bull 3:493–510
 Zhao ZD, Lian JJ, Shi JZ (2006) Interactions among waves, current, and mud: numerical and laboratory studies. *Adv Water Resour* 29:1731–1744
 Zhou F, Guo HC, Liu L (2007) Quantitative identification and source apportionment of anthropogenic heavy metals in marine sediment of Hong Kong. *Environ Geol* 53:295–305