



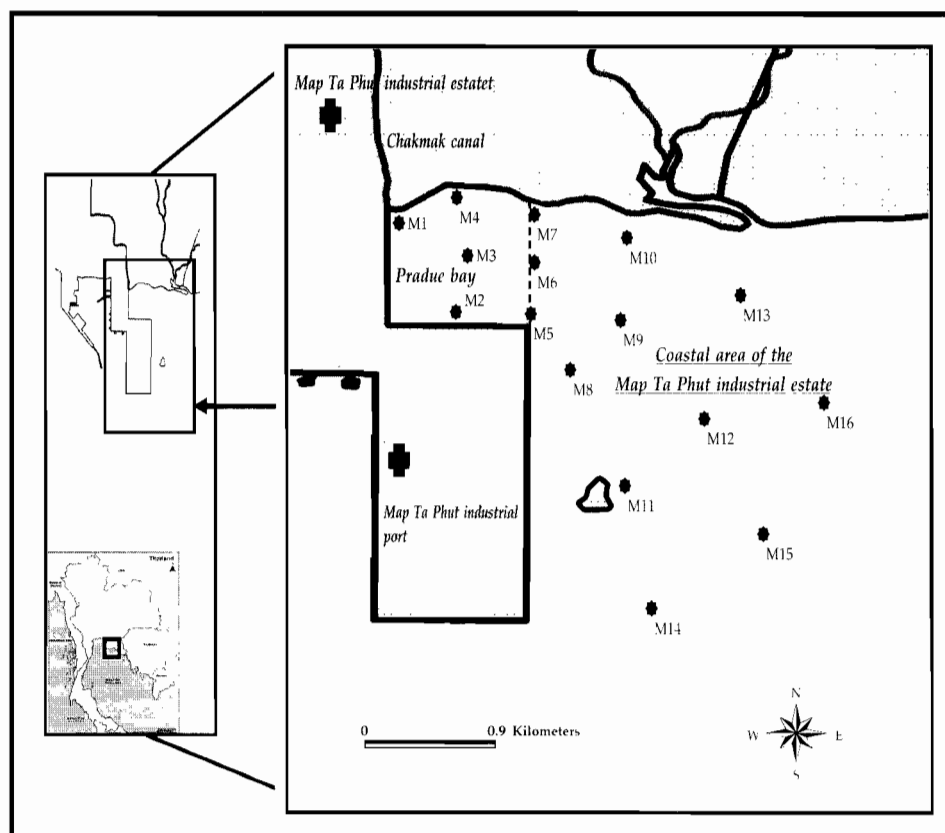


the Chakmak canal that ran through the middle of industrial estate zone. Singkran (2012) reported the basic information of the Pradue bay obtained during the survey of the marine environmental monitoring program of Pollution Control Department (PCD) between 2009 and 2011. Several characteristics of environmental factors were composed of, i.e., the surface area of the bay covered 1.1 km<sup>2</sup>, where the water velocity in the dry and wet seasons were approximately  $\leq 0.01$  m/s, and 0.02 – 0.09 m/s, respectively; the average of water depth was approximately 1.0 – 4.3 m in dry season, and 1.0 – 5.0 m in wet season; the increasing amounts of the sediment thickness found during the year 2009 to 2011 were 11 – 50 cm in November 2009, 14 – 73 cm in January 2010, and 13 – 165 cm in March 2011, respectively.

## MATERIALS AND METHODS

### Sample collection

The sampling sites were designated by a distance gradient, which located from the mouth of Chakmak canal about 0.1-1.1 Km toward coastal area along the Map Ta Phut industrial estate, where the distances were varied approximately from two to four km. Sixteen water samples were collected in August 2011, where seven stations, stations M1 - M7, were from the inner part area of the Pradue bay, and nine stations, stations M8 - M16, were from sites toward coastal area. Likewise, sediment samples were collected from six stations; three samples were from inner part area of the bay (M1, M3, and M6) while the other three samples were from coastal area (M9, M12, and M15). The locations of the sampling stations are summarized and shown in Figure 1.



**Figure 1.** Sampling sites and location, the Pradue bay and coastal area of Map Ta Phut industrial estate, Thailand.

All plastic-wares and borosilicate glass bottle (500 mL) were acid-cleaned and washed several times with Milli-Q water prior to use. Three water samples were collected by the vertical type Kemmerer water sampler at every sampling site, kept in the double zip lock plastic bags, and subsequently transported in an ice box (<4 C°) to the laboratory at Burapha University, Chon Buri province, Thailand. Water samples were separated into two experiments. The first set was used to determine the total Hg through unfiltered samples, while the second set was used to determine the total dissolved and particulate Hg by pre-weighed 0.45- $\mu$ m pore size cellulose acetate membrane Millipore filters with pre-acid cleaned.

Surface sediment samples were collected using a Grab sampler, and kept in plastic sheets before stored in a cooler box to prevent Hg loss. All sediment samples were freeze-dried, homogenized using a mechanical mortar manually, sieved through a 125- $\mu$ m-mesh size nylon sieve, and were transported to the laboratory.

### Analysis of water and sediment samples

The filtered samples were used to determine the total dissolved Hg, which were determined after oxidized by brominating digestion to inorganic mercury ( $\text{Hg}^{2+}$ ) followed by the reduction to elemental mercury ( $\text{Hg}^0$ ) with the reducing agent  $\text{SnCl}_2$  (pro analysis, MERCK), and then with the addition of hydroxylamine (adapted from 245.7 method, US EPA, 2005). The vaporized mercury ( $\text{Hg}^0$ ) was detected by Cold Vapor Atomic Fluorescence Spectrometry (CVAFS) technique (Merlin Atomic Fluorescence Detector, PS Analytical), under the aeration of the solution with an inert purity argon gas (Ar).

The determination of the total Hg in unfiltered samples was carried out using the same method for the total dissolved Hg. The particulate Hg in aqua-phase (ng/ L unit) was estimated using the calculation of the differences between total Hg and total dissolved Hg concentrations.

The concentration of particulate Hg in solid-phase (g/g unit) was determined by digesting

samples with a mixture of  $\text{HNO}_3/\text{HCl}/\text{H}_2\text{O}_2$  at the ratio of 4:1:2, v/v in a Teflon coated block digestion system (SCP, Digi PREP Jr) at 60°C for 12 h. The Hg concentration was determined by CVAFS technique after dilute digestion with Milli-Q water, while the concentrations of total Hg in the sediment samples were determined using the same method as the particulate Hg.

The suspended particulate matter (SPM) was measured while it was in aliquot samples; where the sample was collected on pre-weighed polycarbonate membrane filters with 0.45 $\mu$ m pore size. Several water quality parameters were observed during the sampling, i.e., salinity, pH, and dissolved oxygen (DO), by the YSI-85 multi probe.

### Analytical quality assurances

The limited detections of total dissolved Hg and particulate Hg were 1.75 ng/L and 0.012 $\mu$ g/g dry weights, respectively, which equilibrated to three times of the standard deviation of the blank. The accuracy of the analytical procedure for particulate Hg and sediment concentrations was assessed by an analysis of sediment certified marine sediment reference material (MESS-3), where the certifications of these MESS-3 samples were agreed with recovered percentage between 97 to 105% for CVAFS after the digestion with acid mixture.

## RESULTS AND DISCUSSION

### General water quality parameters

Several parameters of water quality in the bay toward costal area were observed, i.e., dissolved oxygen (DO) ranging from 6.4 to 7.4, pH level ranging from 6.4 to 7.4 mg/L, and salinity ranging from 32.9 to 33.7 ppt. The observed values were less varied and had no significant difference at the level of  $P > 0.05$  in the areas between the bay toward coastal area, which were possibly resulted from the seawater intrusion into surrounding areas during the sampling time. Meanwhile, concentrations of SPM were obviously higher in the bay area than those along the coastal areas, which ranged from 14.7 to

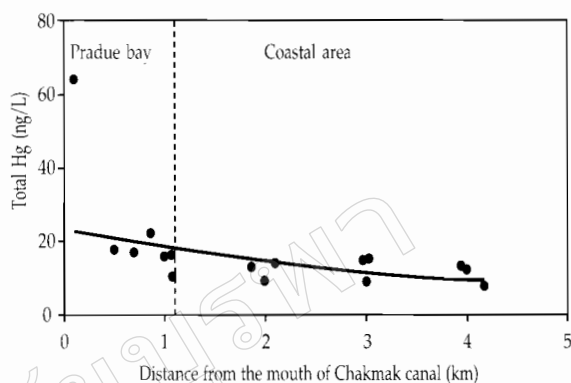
106.0 mg/L. The maximum SPM concentration was found in the area near the mouth of Chakmak canal (station M1), and decreased rapidly in the middle of the bay and further seaward. The influence of SPM loaded from land was observed in the bay. The high concentrations in the bay area were possibly resulted from direct suspended particle sources, river bank erosion and/or a resuspension of the bed sediments during high water discharge condition (Lawson et al., 2001). This result agreed with the report of PCD (2010) (unpublished data), which showed that the high total suspended solid and total solid loaded from land drainage in the Chakmak canal were ranged from 12 to 121 mg/L, and 198 to 9,168 mg/L, respectively.

### Total mercury

The distributions of total Hg concentrations from un-filtered water samples are summarized as shown in Figure 2. The concentrations of total Hg were ranged from 7.18 to 63.77 ng/L, which were lower than that of Thai Coastal Water Quality Standard (100 ng/L) (PCD, 1997). The highest concentration was found in the area near the mouth of Chakmak canal (stations M1), which could be due to the effect of industrial water discharged from the Map Ta Phut industrial estate through the Chakmak canal into the Pradue bay (Panutrakul, 2010). Then, the Hg concentrations decreased rapidly in the middle area of the bay and became constant in the seaward area. The decrease in concentrations in parallel with the distance was possibly following upon a high dilution effect of seawater that led to less contamination of Hg in the middle area of the bay.

It was found that the concentrations in the bay areas when compared to the total Hg concentration in this study to other aquatic systems were lower than those of the worldwide estuaries areas that reported in heavy and moderate polluted areas, i.e., the Scheldt estuary, Belgium, where the concentrations were 14.4-272 ng/L (Leermakers et al., 2001), and the Rio de Janeiro coastal lagoon estuaries, Brazil, where the concentrations were 37 - 285 ng/L (Lacerda and

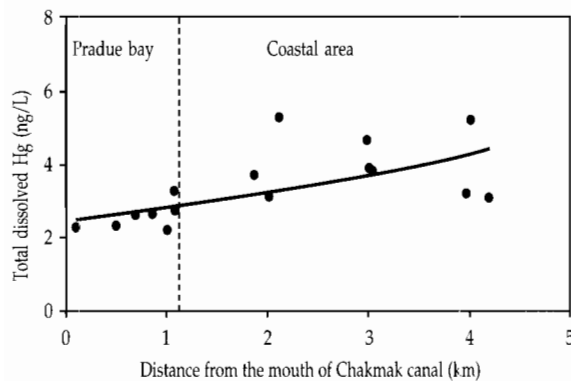
Goncalves, 2001). However, these concentrations were in the same range with the Newark bay, United States, where the concentrations ranged from 12.1 to 77.2 ng/L (Balcom, 2008), which were influenced by inputs of urban sewage loading.



**Figure 2.** Total Hg concentration (ng/L) in the Pradue bay toward coastal area

### Total dissolved mercury

The distributions of total dissolved Hg concentrations from filtered water samples are summarized in Figure 3. The concentrations of the total dissolved Hg were ranged from 2.30 to 5.29 ng/L, which it was gradually increased along the coastal area, whereas it was contrast to a decreasing pattern of the concentrations of the total Hg in the bay. Leermakers et al. (1995) reported that the interaction of Hg bound to organic solid-phase under a long residual time of distribution might promote the degradation of organic particulate matter and released Hg into solution. These could support the enrichment of the total dissolved Hg in seawater of coastal area. The percentages of the total dissolved Hg were correlated with the concentrations of the total Hg in the bay that ranged from 3.6 to 20.5 %, which were lower than those in the coastal area that ranged from 25.5 to 44.4 %. Results of the study of Leermakers (2001) suggested that Hg was mostly bound to the particulate phase whereas very few percentage of total dissolved Hg found in the estuarine and coastal water mass but increased in the marine water mass.



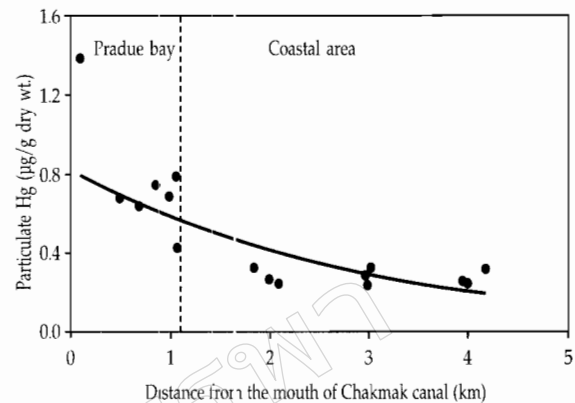
**Figure 3.** Total dissolved Hg (ng/L) concentration in the Pradue bay toward coastal area.

The total dissolved Hg concentrations in the bay toward coastal area were lower than those reported in heavy impacted area of industrial discharge stream in the Ria de Aveiro estuary, Portugal, which were ranged from 1.5 to 13.8 ng/L (Pato et al., 2010). On the contrary, total dissolved Hg concentrations were in the same order as that found in the high sewage discharge areas, such as the Ochlockonee estuary, United States, which were ranged from 1.0 to 4.0 ng/L (Guentzel et al., 1996), and the Newark bay, United States that were ranged from 0.59 to 3.14 ng/L (Balcom et al., 2008).

### Particulate mercury

It was found that concentrations of particulate Hg in the solid-phase ( $\mu\text{g/g}$  unit) were ranged from 0.23 to 1.38  $\mu\text{g/g}$  of dry weight (see in Figure 4). The highest concentration was found in the area near the mouth of Chakmak canal. Then, the concentrations were decreased rapidly in the middle of the bay, and became constant in the coastal area. These concentrations were in the same range to those found in the Chao Phraya River estuary, which were ranged from 0.18 to 2.45  $\mu\text{g/g}$  of dry weight (Sirirattanachai, 2001) and in Bangpakong River estuary, which were ranged from 0.54 to 2.20  $\mu\text{g/g}$  of dry weight (Thongra-ar, 2001). These two rivers are the main rivers where the water is drained into the upper part of the Gulf of Thailand, and serve as carriers that load the

suspended matter derived from freshwater runoff passing through agricultural, industrial, and urbanized areas.

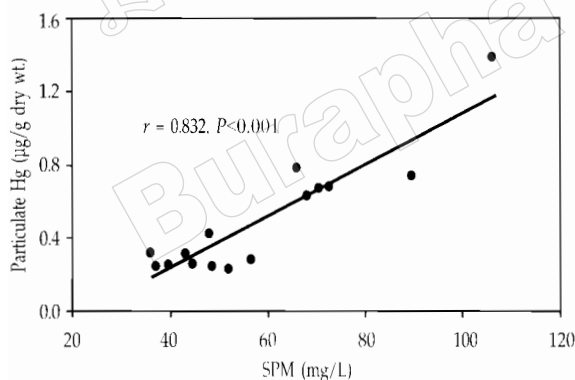


**Figure 4.** Particulate Hg concentration ( $\mu\text{g/g}$  dry weight) in the Pradue bay toward coastal area.

The percentage of particulate Hg in aqua-phase (ng/L unit) was ranged from 4.08 to 61.47 ng/L, which was related to the total amount of Hg in the bay area that was ranged from 82.6 to 96.4 %, while it was higher than those amounts in the coastal area, which were ranged from 55.6 to 75.6 %. The concentrations of the total Hg in the bay were influenced by the bounding suspended matters, which were observed as the major fraction from the industrial discarded matters. This result was agreed with the vertically well-mixed water of estuary zones where the concentrations of the particulate Hg were generally account for more than 90 % of the total Hg (Baeyens et al., 1998; Lacerda and Goncalves, 2001; Laurier et al., 2003). It was shown in the previous study by Liu et al. (2011) which reported that the total Hg concentrations had a major impact by a coal-fired power plant in surface seawater of the coastal area of Xiamen sea, China, which were ranged from 2.5 to 98 ng/L, with a mean value of 55 ng/L found near the discharge outlet, 11 ng/L found at other sampling points, and 12 ng/L found at the reference point. It was also found that high concentrations of total Hg were observed in the water near the discharge outlet from the plant, which was observed as the

particulate Hg appeared in the main fraction (>90 %) of total Hg.

It was observed that there were high variations in the decreasing of particulate Hg in the bay area toward the coastal area. The decreasing in concentrations along the distances from the mouth of Chakmak canal was probably due to low Hg concentrations mixed with marine particulate matters that affected the total Hg that decreased in the middle of the bay. These observations suggested that the association of Hg with particulate matters on the particulate solid-phase (Coquery and Cossa, 1995; Laurier et al., 2003; Pato et al., 2008), which were probably due to the increasing of particulate Hg fraction that related to the increase of SPM, which was observed as a significant positive correlation (see in Figure 5) between particulate Hg and SPM concentrations where the values of  $r = 0.832$  and  $P < 0.001$ . These findings of trace metal adsorptions with SPM indicated that there were the high surface coating and enhancement in the water column (Lofts and Tipping, 1998; Bibby and Webster-Brown, 2006). The profusion of particulate Hg associated with the SPM concentrations was similar to the patterns of those observed in the Sepetiba bay tributaries, Brazil (Paraquetti et al., 2004).

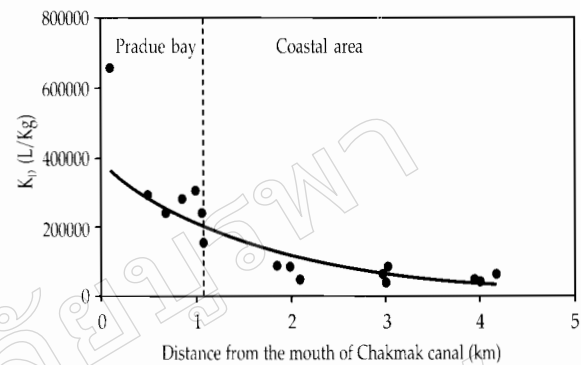


**Figure 5.** The relation between particulate Hg and SPM concentrations

### The partitioning coefficient (KD)

The KD was explained as the ratio of relative affinity of Hg that was dissolved and in particulate phases, which was presented as liter per kilogram

(L/kg) from that total dissolved Hg (ng/L) and in the particulate (ng/kg) concentrations (Coquery et al., 1997). The values of  $K_D$  were varied in two orders of magnitude from low values at  $0.4$  to  $0.8 \times 10^4$  L/kg in the coastal area, to high values at  $1.5$  to  $6.5 \times 10^5$  L/kg in the boundary of the bay (see in Figure 6).



**Figure 6.**  $K_D$  values (L/kg) in the Pradue bay toward coastal area.

The values of  $K_D$  were high in the bay area and decreased when passed into coastal area. The high  $K_D$  values in the bay were agreed with the study of Sirirattanachai (2001), which was studied in the Chao Phraya River estuary, Thailand where the value was about  $3.24 \times 10^5$  L/kg. The comparative increase of  $K_D$  values in the bay was paralleled with an increase of SPM concentrations in this area. Leemakers et al. (1995) reported that  $K_D$  value in the Scheldt estuary, Belgium was about  $2.0 \times 10^5$  L/kg in the lower estuary, which was influenced from physical sorption/desorption processes, whereas the  $K_D$  values increased to about  $1 \times 10^6$  L/kg in the upper estuary, which were influenced from precipitation/oxidation processes of sulfide ions. Therefore, these results suggested that the high  $K_D$  value in the bay was possibly followed upon the physical sorption/desorption processes on particulate phase (Lacerda and Goncalves, 2001; Paraquetti et al., 2004), and then decreased when it was transported into the coastal area.

It was found that  $\log K_D$  values in the bay area and along the coastal area were ranged from

5.18 to 5.82, and 4.94 to 4.96, respectively. The log  $K_D$  values in the bay area were relatively high when compared with the dynamic of high water mixing from other estuaries, i.e., the Loire River estuary, France, where the value was  $5.5 \pm 0.13$ , the Seine River estuary, France, where the value was  $5.9 \pm 0.13$  (Coquery et al., 1997), and the Scheld estuary, Belgium, where the values were from 4.60 to 6.15 (Leermakers et al., 2001).

### Mercury in sediment

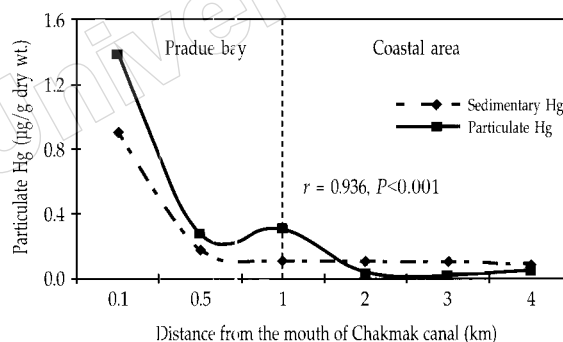
The concentration of Hg in surface sediments from six sampling sites of the bay toward industrial estate were ranged from 0.09 to 0.90  $\mu\text{g/g}$  dry weight. The highest concentration of Hg was found at the area that connected with Chakmak canal (station M1), and was considered as high concentration when compared with other sites from the middle of the bay areas toward the coastal areas (stations M3 to M15) with a mean value of 0.10  $\mu\text{g/g}$  dry weights. Among Hg concentrations, which was proposed by the marine and coastal sediment quality guideline for Thailand (PCD, 2006), it was found that several concentrations did not exceed the low range effect, but only concentration at the area station M1 that connected with Chakmak canal exceeded the median range effect.

The Hg concentrations in sediment were within the ranges of those reported in other industrialized and development of the Asian countries, such as the Haihe River estuary, China, where it was in the range of 0.06 to 8.77  $\mu\text{g/g}$  dry weights (Shi et al., 2005); Hong Kong coast, where it was in the range of 0.05 to 8  $\mu\text{g/g}$  dry weights (Zhou et al., 2007); Mangrove wetlands, China, where it was in the range of 0.002 to 0.90  $\mu\text{g/g}$  dry weights (Hung et al., 2009); the Gaoping estuary, Taiwan, where it was in the range of 0.07 to 2.15  $\mu\text{g/g}$  dry weights (Hung et al., 2009); the Jiuzhou bay, China, where it was in the range of 0.8 to 64  $\mu\text{g/g}$  dry weights (Wang et al., 2009); the Pearl River estuary, China, where it was in the range of 0.001 to 0.20  $\mu\text{g/g}$  dry weights (Shi et al., 2010); Hong Kong coast, where it was in

the range of 0.16 to 0.47  $\mu\text{g/g}$  dry weights (Liang et al., 2011); and the Vemnanad wetland, India, where it was in the range of 0.01 to 4.23  $\mu\text{g/g}$  dry weights (Rammasamy et al., 2012).

### Correlation between particulate Hg and sediment

The correlation of particulate Hg and sediment concentrations along the distance from the mouth of Chakmak canal toward coastal area are summarized in Figure 7. The highest concentration in sediment was increased in relation to the particulate Hg in the area that connected with Chakmak canal, and Hg concentrations decreased simultaneously in the middle of the bay area toward the coastal area. The concentration of the particulate Hg was strongly positive correlated with concentrations of the sediments where the values of  $r = 0.936$  and  $P < 0.001$ . The results showed that most of Hg contamination in the Pradue bay sediments was associated with particulate phase.



**Figure 7.** Particulate mercury concentrations versus sedimentary phase in the Pradue bay toward coastal area.

It was observed that the deposition of particulate Hg in sediments was increased in a high amount in the bay area. The study of Singkran (2012) demonstrated the water depth and velocity in Chakmak canal along with those values in the bay area by using the integration of two-dimensional hydrodynamic models of CCHE2D software package. The prediction of the CCHE2D was proposed and



results were supported the facts that the construction of Map Ta Phut industrial estate port, which expanded into the bay, obstructed the natural route and current direction in the bay along the coastal area and served as a major cause of deposition pattern and an increasing of sedimentation in the bay. These factors affected the scavenging of Hg where it was deposited into the bay sediment.

In conclusion, the different patterns of total dissolved and particulate Hg distributions were observed in the bay toward coastal area. The concentrations of total Hg in this study were not exceeded the Thai Coastal Water Quality Standard. However, the results suggested that the influence of SPM loaded from the land drainage into the bay controlled the distribution and partitioning of Hg. The  $K_D$  values were related to the physical sorption/desorption processes that might control Hg particle-water interaction in this zone. The variations of Hg in particulate fraction and sediments were relatively decreased in the middle of the bay toward coastal area. These observations were influenced by natural changes in the bay, which were a major cause to the high degrees of particulate Hg to deposit and accumulate in the bay sediment.

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