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# TRACE METALS IN MALAYSIAN DREDGED MARINE SEDIMENTS

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**ABSTRACT**

Trace metals were accumulated in sediment by anthropogenic and natural processes. The unwanted sediment was removed by dredging activities. The contaminated dredged marine sediments (DMS) were disposed at offshore. This would affect the marine ecosystem and human health. The objective of this study is to evaluate the trace element pollutant in the DMS. In this study, the DMS were retrieved from 4 locations of Malaysia (Lumut, Melaka, Tok Bali and PasirGudang). The samples were analyzed by x-ray fluorescence for trace metals. There were six trace metals detected in all samples (As, Cr, Cu, Ni, Pb and Zn). The evaluation was carried out considering the sediment quality guidelines (SQGs) proposed by National Oceanic and Atmospheric Administration (NOAA) and Florida Department of Environment Protection (FDEP). The highest mean content of as (18 mg/kg) was observed in Melaka and PasirGudang. Melaka had high mean contents of Cr (66.7 mg/kg), Cu (21.7 mg/kg), Pb (40.3 mg/kg) and Ni (22 mg/kg). PasirGudang had high mean content of Zn with 107 mg/kg. Arsenic concentration in the four samples exceeded the ERL and TEL values. Based on the analysis, all the samples need treatment to make it acceptable for reused. From this study, it shown that DMS have different concentration of trace metals and treatment is necessary to preserve environment due to some of the trace metals detected were potentially affect the environment.

**Keywords:** dredging, dredged marine sediments, sediment quality guideline, trace metals, heavy metals and x-ray fluorescence.

## 1. INTRODUCTION

Water discharged from various sources, such as industry, mining, municipal sewage, tourism activities and agriculture have entered the waterways over time. This continuous process would form sediment load in waterways. The sediments often contaminated with organic and inorganic pollutants (Meegoda and Perera, 2001) [1]. Petroleum hydrocarbon, polycyclic aromatic hydrocarbon (PAH) and polychlorinated biphenyls (PCB) are examples of organic pollutants. The inorganic pollutants are mainly trace metals (cadmium, mercury, lead and nickel), nitrates, phosphates and salts (Zoubier *et al*., 2007) [2].

Trace metals in aquatic environment can be absorbed by suspended materials in water and become a part of bottom sediments. Trace metal is one of critical concern by many countries, including Malaysia due to its toxicity, non biodegradable and accumulated in sediment for very long periods. According toBuruaem *et al*. (2012) [3], the trace metals have tendencies to accumulate food chain. There are some trace metals (e.g., manganese, copper, chromium, etc.) is required for metabolic activity in organism. However, it would turn toxic in excess a certain level in organisms. Some other metals even present in low concentration are considered toxic. Sediment polluted with trace metal are often caused by anthropogenic activities such as mining, agricultural, electroplating industries, wastewater treatment plants and many other industries as shown in Figure 1(Govil *et al.*, 2012 and Garbarino *et al*., 1995) [4 and 5] . The trace metals concentration may affect by the microorganisms activities, pH and redox potential.

Dredging activities is needed to remove unwanted material in waterways. The purpose of dredging is to maintain the waterways for shipping, as capital dredging for marine infrastructure development and to remove contaminated sediment (Bert *et al*., 2012) [6]. Sediment that been removed from seabed is known as dredged marine sediments (DMS). The DMS is potentially polluted with various pollutants (e.g. heavy metals and PAH) due to anthropogenic activities. The DMS were commonly disposed at sea (Wakeman, 2001) [7]. The trace metals in the DMS would uptake by aquatic organism through food chain. Therefore it is risky to human health by causing chronic and acute diseases where human are the end consumer of the food chain(Okoro *et al*., 2012 and Nik Ariffin *et al*., 2014) [8 and 9].

The objective of this study is to identify and evaluate the trace metals in the DMS by compared with sediment quality guideline. Since Malaysia did not have sediment quality guideline, the evaluation was carried out considering the sediment quality guidelines (SQGs) proposed by National Oceanic and Atmospheric Administration (NOAA) and Florida Department of Environmental Protection (FDEP)(MacDonald, 1994 and Long and Morgan 1990) [10 and 11].

## 2. MATERIALS AND METHODOLOGY

### Sample collection

The disturbed dredged marine sediments were collected from Lumut (Perak), Marina Melaka (Melaka), Tok Bali (Kelantan) and PasirGudang (Johor). The samples were dredged by a trailing suction hopper dredger (TSHD) at a depth of 8-12 m from sea level for Lumut site, and recovered using a backhoe dredger (BHD) at a depth of 3.5-6.5 m and 3.5-5.0 m respectively from sea level at Marina Melaka and Tok Bali (Figure-2). Only

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samples from the PasirGudang site were manually retrieved during low tide from 0 - 10 cm of the sediment surface. The samples were put in sealed plastic bags to prevent moisture loss during transportation and storage.

**Trace metals**

The samples were air dried and kept in an oven at 60 °C for 2 days. The samples were ground and sieved passing 63 µm [4]. The sample for this test was prepared by mixing 9 g fine dried sample with 3 g of wax which based on standard operating procedure of the Philips Axis X-ray digital instrument. Then 10 g from the mixture was pressed under a hydraulic press at 20 N of pressure to get a pellet. The diameter of the pellet was 40 mm and 5 mm of thickness. The pellet was testedwith results in ppm.

**Particle size distribution**

Wet sieving was conducted to determine the particle size distribution of the samples. The soil samples were first mixed with a solution of sodium carbonate and sodium hexamethaphosphate to separate discrete particles of the samples. Next, the soil was soaked with water level just enough to make the soil wet. After that, the sample was put into sieves of different aperture sizes.

**pH value**

The pH value was determined by using a pH meter model Hanna HI8424. The sample was mixed with distilled water and allowed to stand for at least 8 hours. Then the electrode of the pH meter was immersed in the sample suspension.

**Loss on ignition (LOI)**

Loss on ignition (LOI) refers to the mass loss of a combustion residue whenever it is heated in an air or oxygen atmosphere to high temperatures. The mass lost from a soil on ignition is related to the organic matter of the soil sample. For LOI, a dried sample at 50 ˚C was heated to 440 ˚C for not less than 3 hours or until constant mass is achieved.

**Mineralogy**

The X-ray diffraction test was carried out to determine the mineralogical composition of the DMS. Fine dried sample in powder form was used for this test.

**Electrical conductivity (EC)**

The electrical conductivity method was used to measure the soluble salt in the samples. It was determined by using EC meter in unit mili Siemens per centimeter (mS/cm) or desiSiemens per meter (dS/m). The sample was mixed with distilled water with ratio 1:5.

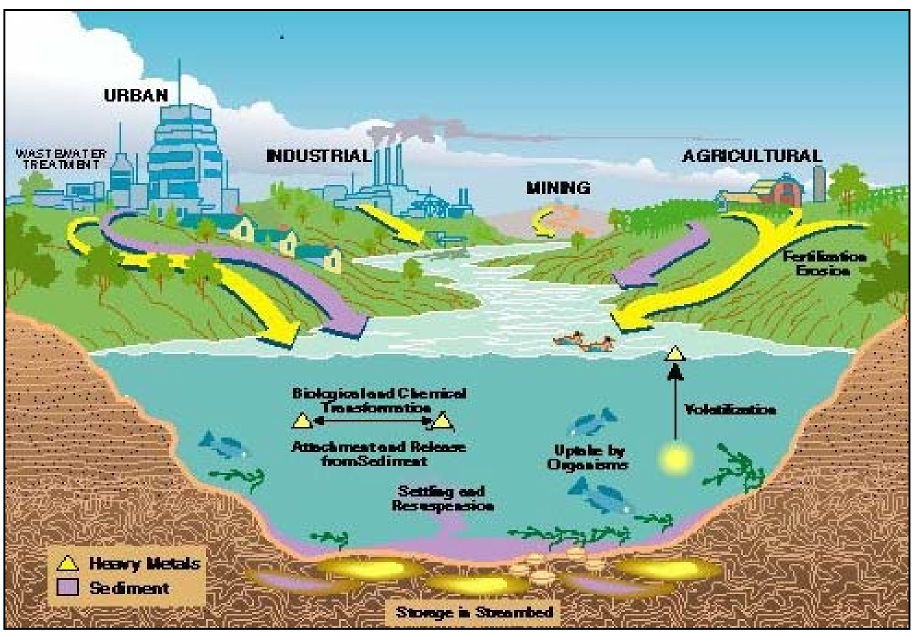


**Figure-1.** Sources and pathway of trace element in sediment (Garbarino *et al.*, 1995) [5].

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**Table-1.** Heavy metals of dredged marine sediments.

| Samples | **Heavy metals** (mg/kg) | | | | | |
|---|---|---|---|---|---|---|
| | **Arsenic** (As) | **Chromium** (Cr) | **Copper** (Cu) | **Lead** (Pb) | **Nickel** (Ni) | **Zinc** (Zn) |
| LT | 11.3 | 59.7 | 5.0 | 33.0 | 18.3 | 53.7 |
| MM | 18.0 | 66.7 | 21.7 | 40.3 | 22.0 | 84.3 |
| TB | 16.5 | 39.5 | 6.5 | 35.5 | 12.5 | 38.0 |
| PG | 18.0 | 54.5 | 10.5 | 28.0 | 9.5 | 107.0 |
| Threshold effect level (TEL)[10] | 7.24 | 52.3 | 18.7 | 30.2 | 15.9 | 124 |
| Probable effect level (PEL) [10] | 41.6 | 160 | 108 | 112 | 42.8 | 271 |
| Effect range low (ERL) [11] | 8.2 | 81 | 34 | 46.7 | 20.9 | 150 |
| Effect range median (ERM) [11] | 70 | 370 | 270 | 218 | 51.6 | 410 |


**Figure-2.** Sampling locations in Peninsular Malaysia and the dredgers used.

## 3. RESULTS AND DISCUSSIONS

### Trace metals

In this study, six elements of heavy metal were detected in all samples. The six elements were arsenic (As), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni) and zinc (Zn). The analyzed metals concentration in LT, MM, TB and PG samples are shown in Table 1. The results were converted to mg/kg unit to suite with the sediment quality guidelines.

The highest amount of as was observed in MM and PG samples with 18 mg/kg. TB sample contained 16.5 mg/kg and LT sample contained 11.3 mg/kg. According to Zulkifli *et al.*, (2010) [12], as could derive from leachate of chromated copper arsenate (CCA), which is used to treat and preserve woods. The high concentration of as in marine sediments, can cause acute and chronic toxicity to marine organisms. It was could be transferred via ingestion of particulate matter (As associated with particles), through membrane-facilitated transport or passive diffusion where as dissolved in water Bhattacharya *et al*., 2007[13].

MM sample contained the highest concentration of Cr with 66.7 mg/kg, 59.7 mg/kg for LT, 54.5 mg/kg for PG and 39.5 mg/kg for TB. The possible anthropogenic sources of Cr could be oil spills and ballast cleaning which inadequately increased with the expanding in sea vessel traffic (Zulkifli *et al*., 2010) [12].

Meanwhile, the concentrations of Cu for all samples were 5 mg/kg (LT), 21.7 mg/kg (MM), 6.5 mg/kg (TB) and 10.5 mg/kg (PG) respectively Cu is intimately related to the aerobic degradation of organic matter (Siddiquee *et al*., 2009) [14].

The highest Pb concentration was detected in MM sample (40.3 mg/kg), followed by TB (35.5 mg/kg), LT (33 mg/kg) and PG (28 mg/kg). The sources of Pb in the samples could be attributed to spills of lead from boating activities. Dust from combustion of petrol in automobile cars enhanced the increase of lead content related with traffic road (Saeed *et al*., 2008) [15].

PG contained least amount of Ni compared to the other three samples. The Ni concentration in LT sample was 18.3 mg/kg, MM which indicated the highest concentration 22 mg/kg, TB was 12.5 mg/kg and PG 9.5 mg/kg. According to Zulkifli *et al*., (2010) [12], Ni is used in many industrial and consumer products (e.g. stainless steel, coinage and magnets). These could have been discharged into waterways leading to the sea polluting the sediments.

PG showed the highest amount of Zn with 107 mg/kg, followed by MM (84.30 mg/kg), LT (53.70 mg/kg) and TB (38 mg/kg). The PG sampling site could have receive anthropogenic Zn input from industrial activities, such as electrical and electronic factories, ports, coal-generated power plants and transportation. According to Zulkifli *et al*., 2010 [12], agricultural activities also could contribute to Zn contamination in sediments.

Particle size has significant effect on the heavy metal content in sediment. Heavy metals stick with fine particles and enriched in organic matter. This supports the finding, where the heavy metals were detected in all samples which dominant with fine particles (Table-2). However, in controlling the distribution of trace metals in the sediment, organic matter content is more important factor compared to particle size. The different mobility of

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each sediment fraction may influence the trace metal distribution accumulated in sediments.

**Table-2.** Properties of dredged marine sediments.

| Experiment | Results | | | |
|---|---|---|---|---|
| | LT | MM | TB | PG |
| **Particle Size Distribution (%)** | | | | |
| Gravel (76.2 ≥ gravel >2 mm) | 3 | 3 | 5 | 2 |
| Sand (2≥ sand >0.075mm) | 15 | 9 | 20 | 18 |
| Silt (0.075≥ silt >0.002mm) | 4 | 20 | 15 | 54 |
| Clay (≤ 0.002mm) | 78 | 68 | 60 | 26 |
| **Loss on ignition (%)** | 6.33 | 9.39 | 4.28 | 8.38 |
| **pH value** | 8.30 | 8.32 | 8.51 | 8.35 |
| **Electrical conductivity (dS/m)** | 7.72 | 6.74 | 8.10 | 11.29 |
| **Mineralogy (%)** | | | | |
| Quartz ($SiO_2$) | / | / | / | / |
| Halite (NaCl) | / | / | / | / |
| Montmorillonite ($Na_{0.3}(AlMg)_2Si_4O10OH_2.6H_2O$) | / | / | / | / |
| Kaolinite ($Al_2Si_2O_5(OH)_4$) | / | / | / | / |
| Calcite ($CaCO_3$) | / | / | / | / |
| Illite(K,H3O)$Al_2Si_3AlO10(OH)_2$) | / | / | / | / |

/ detected

According to Pinheiro *et al*., 1999 and Liu and Gonzalez 1999[16 and 17], organic matter immobilizes Pb and it can affect the Pb concentration. The organic matter may influence the site of deposition of metals due to its ability of adsorption and this leads to strong correlation between it and metal elements.

According to Pakzad *et al,* (2014) [18], high content of illite is one of the factors causing the high Zn concentration in the sediments. The higher amounts of clay particle can result in more concentration of Zn. Organic matter and clay minerals are the factors controlling adsorption of Cu. The decrease of clay minerals also results in the lower concentration of Cu. It is absorbed by clay particles. Salinity has caused the lower concentration of Zn and Cd (Pakzad *et al,* 2014) [18].

Tack (2010) [19], found that fines content in sediment has higher tendency for metal sorption compared to coarse particles. Contaminating metals tend to adhere to the fine particles in aquatic sediments, due to their greater relative surface area Herut and Sandler, 2006 [20]. Sediment parameters (mineralogy, texture), metal characteristics, pH, organic matter and oxidation–reduction potential are important parameters controlling the accumulation and the availability of heavy metals in the sedimentBastami *et al*., 2015[21].

Several factors, including the mineralogical and chemical compositions of suspended material, anthropogenic influences, deposition, sorption, enrichment in organism, and various physicochemical characteristics can influence the distribution of heavy metals Singh *et al.,* 2005 and Jain *et al*., 2007[22 and 23].

**Assessment according to sediment quality guidelines (SQGs)**

The probable toxic effect of the dredged marine sediments on the benthic organisms due to the heavy metal load had to be evaluated. This is to avoid serious adverse effects on the ecosystem of the disposal site especially at open sea water. It is a common approach to compare the pollutants concentration with sediment quality guidelines (SQGs). The evaluation was carried out by referring to the sediment quality guidelines (SQGs) proposed by National Oceanic and Atmospheric Administration (NOAA) and Florida Department of Environmental Protection (FDEP) due to Malaysia did not have sediment quality guideline (MacDonald, 1994 and Long and Morgan 1990) [9 and 10]. In order to protect the benthic communities, these guidelines indicate concentrations of specific pollutants within sediments. Two sets of guidelines are commonly used, effect range low (ERL)/effect range median (ERM) and threshold effect level (TEL)/ probable effect level (PEL) approaches. The main difference is that the TEL/PEL approach incorporates effects plus no effects data whereas the ERM/ERL method is based on effects data only. Low range values (ERL or TEL) represent concentrations below which adverse effects upon sediment dwelling fauna would be expected infrequently. Upper range values (ERM or PEL) represent concentrations above which adverse effects are likely to occur. The

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adverse effects could occasionally occur with the pollutants concentration between ERM-ERL and TEL-PEL.

The distribution of metals in these ranges is shown in Figure 3-8. The comparison with ERL/ERM guidelines showed that arsenic (As) exceeded the ERL and below ERM values indicated occasionally adverse effect (Figure-4). The chromium (Cr) (Figure-5), copper (Cu) (Figure-6), lead (Pb) (Figure-7) and zinc (Zn) (Figure-9) in all samples exhibited concentration below ERL indicating rarely adverse effects to marine organisms. Nickel (Ni) concentration in MM sample is greater than ERM value which expects occasional adverse effect to marine organisms (Figure-8).

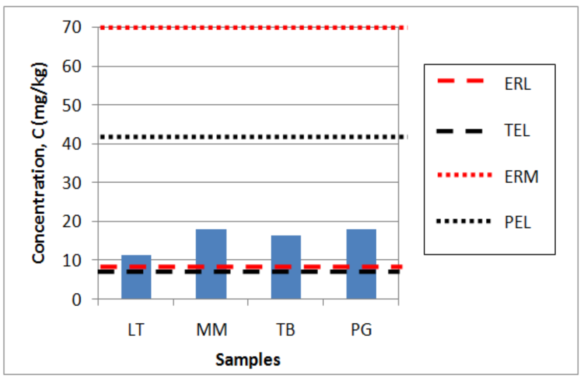


**Figure-3.** Arsenic concentrations in DMS.

The comparison of As, Cr, Cu, Pb, Ni and Zn concentrations with TEL/PEL values are shown in Figure-4-9. It is shown that for the TEL/PEL approach as concentration (Figure-4) for all samples were between TEL and PEL value. This indicated the possible effect range within which adverse effects occasionally occur. For element Cr, only TB sample below TEL, which biological effects are not expected to occur (Figure-5). The Cr concentration of LT, MM and PG samples were between the TEL and PEL value.

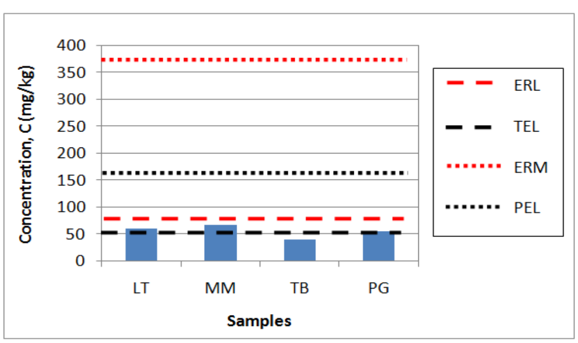


**Figure-4.** Chromium concentration in DMS.

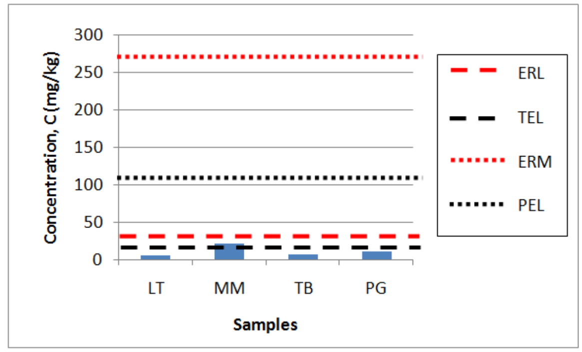


**Figure-5.** Copper concentration in DMS.

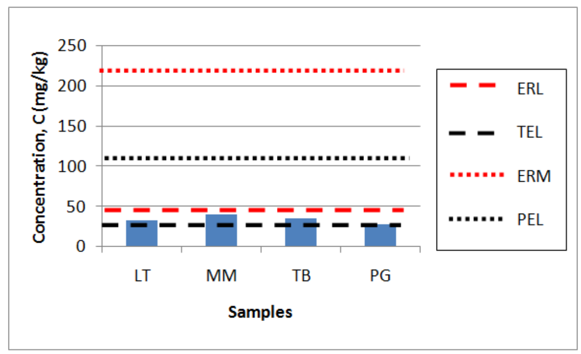


**Figure-6.** Lead concentrations in DMS.

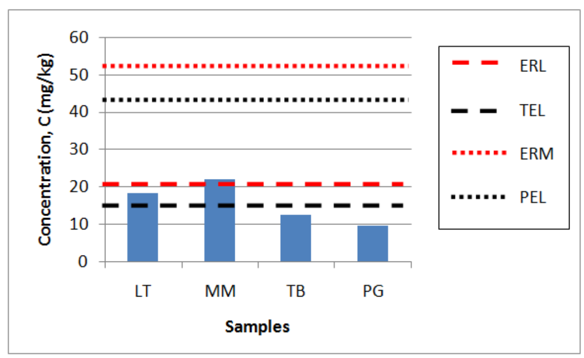


**Figure-7.** Nickel concentration in DMS.

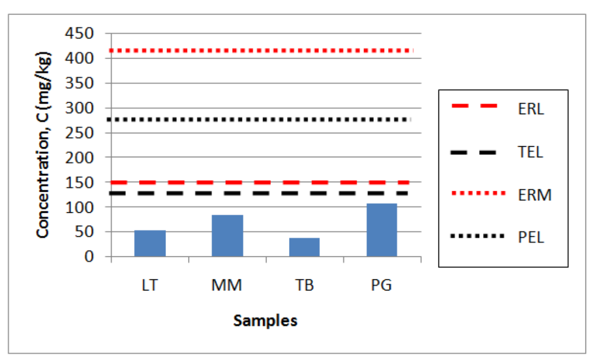


**Figure-8.** Zinc concentration in DMS.

Cu concentration in all samples were below TEL, adverse effects rarely occur except MM, between the TEL and PEL which indicated adverse effects occasionally occur (Figure-5). The concentration of Pb in PG sample was below than TEL (Figure-6), the minimal effect range within which adverse effects rarely occur and the possible effect range within which adverse effects occasionally occur for LT, MM and TB.

The Ni concentration in LT and MM were between the TEL and PEL values, while TB and PG below the TEL value (Figure-7). Figure-8 shows Zn concentration in all samples was below TEL value. It indicated adverse effects rarely occur for element Zn in all samples.

By comparing the heavy metals concentration with the SQG, the LT sample was contaminated with As, Cr, Pb and Ni. The MM sample was contaminated with five of six heavy metals; As, Cr, Cu, Pb and Ni. The As and Pb were pollutant in TB sample and PG sample was polluted with As and Cr.

## 4. CONCLUSIONS

The DMS sampled from Lumut, Melaka, Tok Bali and PasirGudang were found contained six trace metals; As, Cr, Pb, Zn, Ni and Cu. All the trace metals concentration in the four of DMS was below than ERM and PEL. However, As, Cr, Pb and Ni in Lumut DMS exceed the TEL values. This indicated that pretreatment is needed to make the Lumut DMS acceptable for reuse. Arsenic and nickel concentration in Melaka DMS was exceeded both guideline, ERL and TEL. The concentration of Cr, Cu and Pb in Melaka was also higher than TEL limits. The Melaka DMS are acceptable for reuse after pretreatment. The DMS of Tok Bali contained two trace metals (As and Pb) that higher than ERL and TEL. The DMS need treatment to reduced the as and Pb concentration thus make the DMS acceptable for reuse. The PasirGudang DMS was high concentration of as and Cr. The As exceeded ERL and TEL value and Cr exceeded the TEL value. To make it acceptable for reuse, pretreatment should be applied to reduce the as and Cr concentration. From this study, it shown that DMS have different concentration of trace metals and treatment is necessary to preserve environment due to some of the trace metals detected were potentially affect the environment.

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