MERCURY ACCUMULATION IN SEDIMENTS OF A MANGROVE ECOSYSTEM IN SE BRAZIL

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Abstract. The Hg accumulation in sedimentary environments of a mangrove ecosystem in Sepetiba Bay, SE Brazil, was investigated. These environments include sediments of a mangrove forest, the main tidal creek that drains the forest, and the bare seaward-edge mud flat adjacent to forest and tidal creek. Maximum Hg concentration peaks in sediments from the mud flat (184 ng g^{-1}), tidal creek (98 ng g^{-1}), and mangrove forest (60 ng g^{-1}) correspond to enrichment factors of 6.1, 3.3, and 2.0 above the estimated average background level, respectively. Average inventories of Hg excess (background-corrected) concentrations were substantially different between environments, decreasing from mud flat sediments (5.2 mg m^{-2}) to creek sediments (3.3 mg m^{-2}) to mangrove forest sediments (0.9 mg m^{-2}). Mercury concentration profiles indicated a consistently higher accumulation of Hg in surface layers of mud flat and tidal creek sediments, whereas mangrove forest sediments showed a higher Hg accumulation in root-rich subsurface layers, in agreement with an enrichment of Fe and organic matter contents. While Hg distribution in mud flat and tidal creek sediments appears to be largely affected by contamination, its distribution in mangrove forest substrate appears to be greatly affected by root-sediment interactions. Mercury levels in the study site were comparable to those observed in coastal sediments under moderate Hg contamination at local and regional scales. Results indicate that sedimentary environments surrounding the mangrove forest retain most of the anthropogenic Hg reaching the ecosystem. Since tidal waters have been previously demonstrated as the main source of metals to the site, it is suggested that the Hg retention in mud flat sediments precede and may avoid a higher Hg accumulation in landward environments.

Keywords: intertidal flats, mangroves, mercury, sediment chemistry, Sepetiba Bay

1. Introduction

The biogeochemistry of mangrove sedimentary environments can maintain conditions favorable for retention of anthropogenic inorganic contaminants, particularly heavy metals (Sadiq and Zaidi, 1994; Tam and Wong, 1995; Lacerda, 1998). After reaching mangrove environments these elements tend to form insoluble compounds, mainly as metal-sulfides and metal-organic matter complexes, and adsorb onto the surface of sediment particles, which can result in a substantial accumulation of anthropogenic metals at a given site (Harbison, 1986; Badarudeen *et al.*, 1996; Peters *et al.*, 1997; Clark *et al.*, 1998).

Regardless of the environmental and public health concerns about Hg pollution (e.g., Kawagushi *et al.*, 1999; Klekowski *et al.*, 1999), there are relatively

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few studies on Hg accumulation and behavior in mangrove ecosystems compared to other metal contaminants of potential concern (Lacerda, 1998). Mercury is a ubiquitous contaminant derived from urban and industrial sources that reaches many coastal areas worldwide, such has been observed in Sepetiba Bay, Rio de Janeiro State, SE Brazil. After reaching Sepetiba Bay waters, Hg is distributed following tidal currents creating areas of Hg accumulation along the northeastern bay area (Marins *et al*., 1998). About 35% of the Sepetiba Bay coast is occupied by mangrove ecosystems, mostly distributed from the northern to the eastern bay margins. Although the storage of Hg in mangrove dominated areas of Sepetiba Bay has not been quantified, there is some information on Hg contamination and behavior in the bay's intertidal environments. Quevauviller *et al*. (1992) demonstrated an elevated presence of dimethyl-Hg in anoxic and organic matter-rich mangrove sediments. Marins *et al*. (1997) evaluated the post-depositional remobilization of Hg in sediments under the influence of salt marsh macrophytes and also observed an elevated Hg accumulation in such sediments.

In order to elucidate the relative importance of the Hg accumulation in different sedimentary environments of a mangrove area in Sepetiba Bay, we investigated the Hg storage in a mangrove ecosystem in Itacuruçă, located at the northern bay margin. The studied mangrove ecosystem is comprised of three major depositional environments, including a mangrove forest dominated by *Rhizophora mangle* L., a main tidal creek that drains the forest, and a bare mud flat located in the seaward edge of the forest and the creek (Figure 1). Detailed descriptions of metal sources and emissions to Sepetiba Bay and studies on metal biogeochemistry in its mangrove ecosystems are published elsewhere (Silva *et al*., 1990; Lacerda *et al*., 1988, 1991, 1993b; Marins *et al*., 1998). Previous studies have showed the absence of significant freshwater input to the study site, as indicated by $\delta^{18}O$ analysis of tidal creek waters along two tidal cycles, demonstrating that tidal waters are the main source of the material deposited in the site, including metals (Lacerda *et al*., 1988).

2. Material and Methods

Sediment samples were collected in the mangrove forest, mud flat, and tidal creek environments of the Itacuruçá mangrove ecosystem in August 1997. Four sediment cores were sampled from each environment by means of acid-cleaned acrylic tubes (9 cm i.d.), during low tide. Sediment cores were sampled nearly 30 m apart from each other, along a transect parallel to the coastline in the mud flat, as well as in the mangrove forest fringe (Figure 1). Cores were also sampled nearly 30 m apart from each other along the creek length (Figure 1).

Sediment cores were sectioned in 0–5, 5–10, 10–20, and 20–30 cm intervals. Sediment samples were wet-sieved through a 1 mm mesh sieve to remove shell fragments, live mangrove roots (abundant a few centimeters below the mangrove forest sediment surface), and mangrove detritus prior to all analysis. In order to

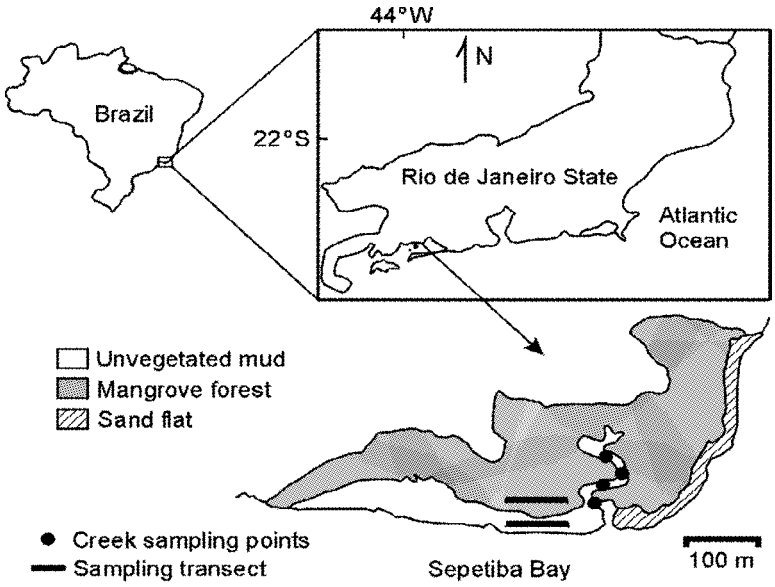

Figure 1. Location of Itacuruçá mangrove ecosystem, tidal creek sampling points, and sampling transects at the mangrove forest and unvegetated mud flat.

characterize the sedimentary environments, dried (50 °C for 48 hr) sediment sub-samples were used to determine the content of Fe, fine sediments, and organic matter. Organic matter content was determined gravimetrically after combustion at 450 °C for 24 hr, while the content of fine (silt-clay) sediments was determined after sieving through a 63 μm mesh sieve. Total Fe concentrations were analyzed by conventional flame atomic absorption spectrophotometry, following a digestion in a nitric-hydrochloric-fluoridric acid solution (Fiszman *et al.*, 1984). Dried (50 °C for 48 hr) sediment sub-samples were also used to determine the Hg content. Total Hg concentrations were analyzed by cold vapor atomic absorption spectrophotometry, following a digestion in an aqua regia solution and a reduction with $SnCl_2$ (Marins *et al.*, 1998). Standard reference sediments (NIST SRM 2704) were analyzed in parallel with the samples in order to estimate the Hg analytical recovery. Reference sediment analysis showed a precision of 10%, as indicated by the relative standard deviation of six replicates. This analysis showed that Hg was nearly quantitatively recovered, with an average recovery of 97%.

Maximum enrichment factor (EF) was calculated by the ratio between maximum Hg concentration peak in a sediment core and the average background value for the site. The inventory of Hg excess concentrations was calculated through the product: $Hg_{xs} \times \rho \times t$, where Hg_{xs} is the excess (background-corrected) Hg concentration (ng g^{-1}), ρ is the sediment density (g cm^{-3}), and t is the thickness of sediment-depth interval (cm), for each sample (e.g., Cundy and Croudace, 1995). The total Hg_{xs} inventory of a sediment profile was calculated through the sum of Hg_{xs} inventories of each depth interval. Sediment density was estimated by

weighing a known volume of sediment subsamples, after drying at 50 °C for 48 hr; it ranged in 0.50–1.24, 0.62–1.20, and 0.45–1.22 g cm^{-3} in sediments from the mud flat, tidal creek, and mangrove forest, respectively. Averages of low concentrations found in the deeper layers (20–30 cm depth) of sediment cores were used to estimate the Hg background concentration of the study site. Average background concentration for the site measured 30 ng g^{-1}, as indicated by background values of the twelve individual cores, which ranged between 17 and 48 ng g^{-1}. These values are close to or lower than background levels previously observed in sediments of Sepetiba Bay and other coastal environments in the Rio de Janeiro State (20–60 ng g^{-1}; Lacerda *et al.*, 1993a; Marins *et al.*, 1997, 1998; Wasserman *et al.*, 2000; Machado *et al.*, in press).

3. Results and Discussion

3.1. Mercury vertical distribution in sediments

Total Hg concentrations in sediments are presented in Figure 2. Depth profiles indicated a pronounced decrease in Hg concentration from surface to subsurface layers of mud flat sediments. There was no substantial trends in Hg distribution in mangrove forest sediments, except for a slight increase in Hg concentration at the 10–20 cm depth. Creek sediments also had a decrease in Hg concentration with increasing depth, but was less accentuated than that in mud flat sediments, displaying a smooth profile. Since there are various anthropogenic sources of Hg in the Sepetiba Bay basin area (e.g., from oil-fired electric power generation and metallurgical activities; Marins *et al.*, 1998), a considerable degree of Hg contamination in the study site can be expected. Although mud flat sediment profiles, and to a lesser extent tidal creek sediment profiles, suggest a consistent record of anthropogenic Hg input, the lower concentrations and a greater Hg accumulation in subsurface layers of mangrove forest sediments do not appear to be directly linked to an anthropogenic load.

Diverse factors may influence the distribution of anthropogenic metals in aquatic sediments, such as sediment chemical and textural composition, and diagenetic processes (Salomons and Förstner, 1984). Data on sediment composition are shown in Figure 3. In general we found that silt-clay content decreased with increasing depth in mud flat sediments, with the opposite observed in mangrove forest sediments. No pattern related to sediment depth was observed for silt-clay content in tidal creek sediments. Organic matter content was greater in the upper surface layer (0–5 cm) of mud flat sediments, whereas the opposite occurred in tidal creek sediments. No general trend in organic matter content was observed for mangrove forest sediments; the highest content of organic matter was found at the surface and in the 10–20 cm depth layers. Iron concentrations were similar in all layers of the mud flat sediments, with the exception of a slight rise in the surface layer. In

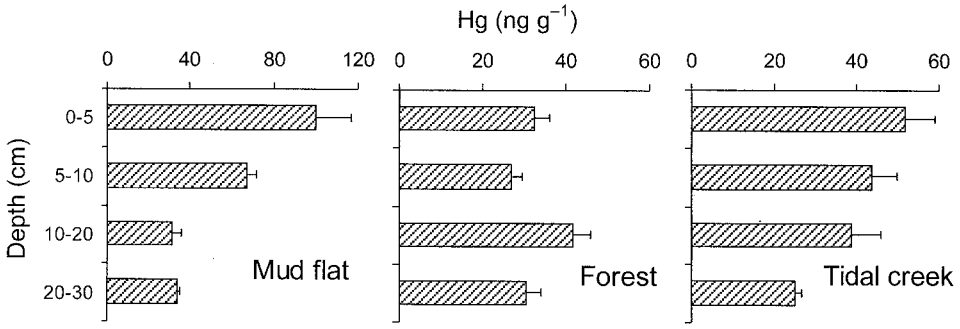

Figure 2. Mercury concentrations in sediments from studied mud flat, mangrove forest and tidal creek. Bars indicate average values (± SE) of four cores. Note the scale differences between sampling stations.

the mangrove forest sediment profiles, Fe concentrations varied little, showing an increase only at a depth of 10–20 cm. Iron levels increased slightly with increasing depth in the tidal creek sediments. Previous studies in the same mangrove forest also found higher total Fe concentrations at subsurface sediment layers (Silva *et al.*, 1990; Lacerda *et al.*, 1991), although levels of organic matter were highest only at surface layers (Lacerda *et al.*, 1991). There was no apparent association between Hg concentration and other parameters, except for organic matter, silt-clay, and Fe in the 0–5 cm layer of mud flat sediments, and for organic matter and Fe at 10–20 cm depth in mangrove forest sediments. In fact, the distribution and the diagenetic behavior of Hg in coastal sediments may be related to organic matter content (Lindberg and Harriss, 1974; Mucci and Edenborn, 1992) and Fe and Mn redox behaviors (Gagnon *et al.*, 1997; Bloom *et al.*, 1999).

The coincidence between peaks of Hg and other constituents of mangrove forest subsurface sediments (Figures 2 and 3) may indicate an influence of sediment composition on Hg distribution in sediment horizons affected by mangrove rhizospheres. A strong effect of macrophyte roots on Hg depth distribution has been shown in salt marsh sediments of Sepetiba Bay, where a post-depositional remobilization and depletion of Hg in subsurface layers oxidized by the oxygen release from roots was observed (Marins *et al.*, 1997). However, the presence of a subsurface peak of redox-sensitive elements like Fe, and associated elements (e.g., Hg; Gagnon *et al.*, 1997), may indicate the presence of a subsurface redox boundary (Mucci and Edenborn, 1992; Cundy and Croudace, 1995). Subsurface sediment oxidation can be a characteristic of sediments colonized by mangrove trees due to oxygen release from roots, as appears to be the case of the study site (Silva *et al.*, 1990), which may be enough to affect both the distribution and geochemical partitioning of heavy metals (Lacerda *et al.*, 1993b; Clark *et al.*, 1998). On the other hand, although the root tissue decay is possibly the best explanation for the subsurface organic matter enrichment in the mangrove forest substrate, it is unexpected that the organic matter derived from the generally metal-poor mangrove tissues

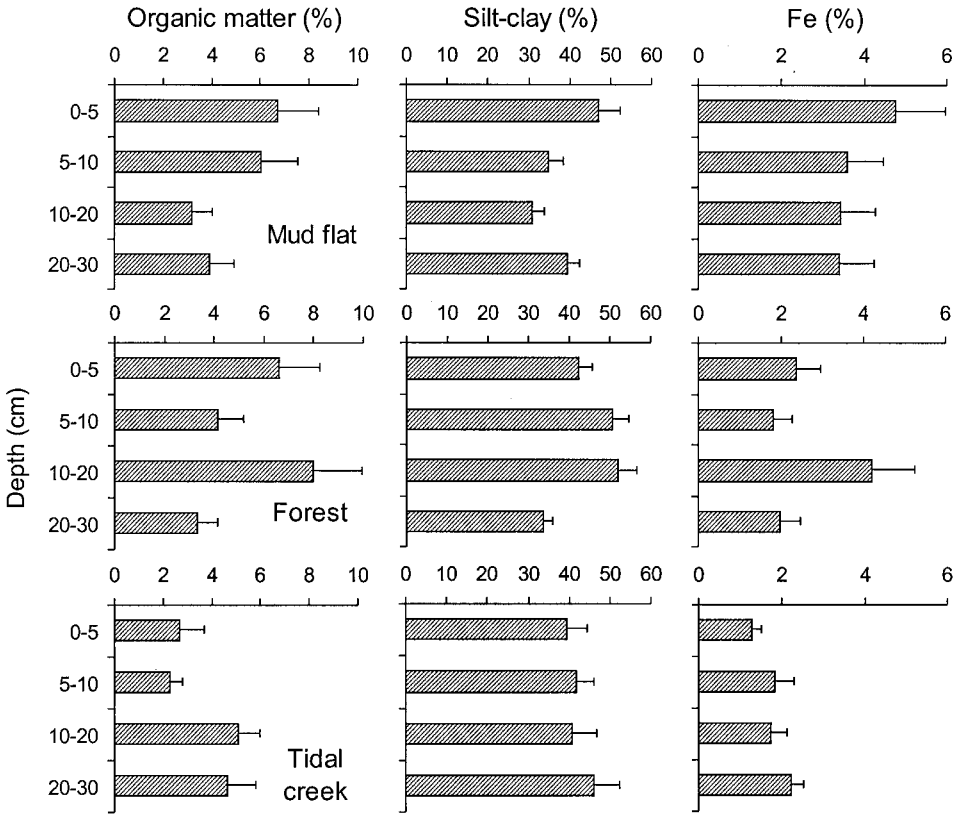

Figure 3. Organic matter, iron, and silt-clay contents in sediments from studied mud flat, mangrove forest and tidal creek. Bars indicate average values (± SE) of four cores.

(Peters *et al.*, 1997; Lacerda, 1998) can contribute substantially to sediment Hg accumulation. In the studied mangrove forest, the total *R. mangle* biomass contains less than 1% of the total storage (including sediment and vegetation burdens) of seven other heavy metals (Silva *et al.*, 1990).

3.2. Mercury accumulation in sediments

Table I summarizes data on the degree of Hg accumulation in sediments of the study site. All datasets present a high spatial variability within the environments, as seems to be typical of mangrove environments (e.g., Harbison, 1986; Tam and Wong, 1995). Higher Hg concentrations were observed in mud flat sediments, where the maximum EF (6.1) was nearly three-times and two-times higher than that of sediments from the mangrove forest (2.0) and tidal creek (3.3), respectively (Table I). A strong sediment heterogeneity within the environments was demonstrated (Figure 3), indicating the relevance of an inventory-based approach, which is dependent on both metal concentration and accumulation rates (Frignani *et al.*,

TABLE I

Maximum Hg concentration peaks, maximum Hg enrichment factors (EFs), and total excess Hg concentration (Hg_{xs}) inventories in different sedimentary environments. Values are averages of four replicate sediment cores, with the range of data from individual cores between parenthesis

	Mud flat	Mangrove forest	Tidal creek
Maximum concentration (ng g^{-1})	110 (54–184)	39 (22–60)	72 (55–98)
Maximum EF	3.7 (1.8–6.1)	1.3 (0.7–2.0)	2.4 (1.8–3.3)
Total Hg_{xs} inventory (mg m^{-2})	5.2 (0.6–7.8)	0.9 (0–2.6)	3.3 (1.1–6.6)

1997), to evaluate the Hg storage in sediments. Total Hg_{xs} inventories also demonstrated a higher Hg accumulation in mud flat (5.2 mg m^{-2} in average) and creek sediments (3.3 mg m^{-2} in average) than in mangrove forest sediments (0.9 mg m^{-2} in average). Sediment cores sampled from the mangrove forest showed concentration ranges from below the estimated average background level (30 ng g^{-1}) to slightly above it, resulting in only 33% of Hg_{xs} in mangrove forest sediments above the uncertainty associated with the average background level ($\pm$9 ng g^{-1}), in contrast to proportions of 75 and 58% in mud flat and creek sediments, respectively. This supports the assumption that Hg levels in mangrove forest sediments are not directly linked to an anthropogenic influence, and suggests an anthropogenic contribution to the Hg enrichment observed in mud flat and tidal creek surface sediments.

As presented in Table II, this study found maximum Hg concentrations in sediment cores of similar magnitude to that of previous analysis of Hg contamination in Sepetiba Bay area and bottom sediments of Rio de Janeiro State continental shelf and coastal lagoons. A stronger contrast is demonstrated between our results and those of mangrove sediments from Guanabara Bay (also located in Rio de Janeiro State), where the past Hg input from a chlor-alkali plant (Wasserman *et al.*, 2000) and from a refuse landfill and many other urban and industrial sources (Machado *et al.*, in press) resulted in Hg concentrations in sediments one or two orders of magnitude higher than those observed in Sepetiba Bay (Table II). Greater Hg exposure levels are likely to occur in mangrove areas due to strong contamination point sources, such as chlor-alkali plants (e.g., Meyer *et al.*, 1998) and refuse landfills (e.g., Clark *et al.*, 1998). However, most of previous studies on Hg contamination in mangrove areas indicate an influence of smaller point sources and/or diffuse sources, resulting in Hg levels in sediments lower than or within the same magnitude of our study site (e.g., Ragsdale and Thorhaugh, 1980; Da Silva *et al.*, 1996; Klekowski *et al.*, 1999; Perdomo *et al.*, 1998). Major Hg sources in Sepetiba Bay are of a diffuse nature (e.g., Marins *et al.*, 1998; Lacerda, 1998), resulting in long term, rather than acute contamination of its ecosystems.

TABLE II

Comparison between maximum Hg concentrations found in sediment cores from this study and Hg concentration ranges available in the literature for other coastal environments at local (northern and northeastern Sepetiba Bay) and regional (Rio de Janeiro State) scales

Environment	Hg (ng g^{-1})[a]	Source
Sepetiba Bay intertidal sediments	22–184	This study
Sepetiba Bay mangrove forest sediments	43–278	Quevauviller *et al*. (1992)
Sepetiba Bay salt marsh sediments	56–177	Marins *et al*. (1997)
Sepetiba Bay bottom sediments	17–163	Marins *et al*. (1998)
Sepetiba Bay bottom sediments	23–135	Wasserman *et al*. (in press)
Continental shelf bottom sediments	30–220	Lacerda *et al*. (1993a)
Coastal lagoons bottom sediments	120–440	Lacerda and Gonçalves (2001)
Guanabara Bay mangrove sediments	50–3000	Wasserman *et al*. (2000)
Guanabara Bay mangrove sediments	35–890	Machado *et al*. (in press)

[a] Concentrations determined in total sediments, except for those reported by Lacerda and Gonçalves (2001), which were determined in the <63 μm sediment fraction.

Along six transects across an intertidal zone in Australia, Harbison (1986) observed a trend of decreasing Zn, Cu, and Pb concentrations in sedimentary environments following the order: mangrove forest sediments > mud flat sediments > tidal channel sediments. Although Morrisey *et al*. (2000) observed higher concentration of Zn and Pb, but not for Cu, in mud flats located between mangrove forests and the main channel at one of their studied sites in New Zealand, there was no clear pattern of metal distribution among different shore heights (mangrove forest height > mud flat height > channel height) of their study area. However, since tidal inundation can affect the metal spatial variation in sediments (Mackey and Hodgkinson, 1995), it may be expected that the trend of Hg accumulation in the Itacuruçá mangrove ecosystem follows the order of tidal inundation (mud flat-channel-mangrove forest), as observed here.

A higher metal concentration in mud flat sediments than in adjacent landward mangrove forest sediments has been previously observed in Sepetiba Bay (Lacerda *et al*., 1993b). If this is a frequently observed trend, the accumulation of harmful metals in mud flats outside mangrove forests should be accounted for by monitoring contamination and the exploitation of resources in such areas. The current use of Sepetiba Bay shellfish for human consumption, identified as accumulators of metal contaminants (Carvalho *et al*., 1991), derived from mud flats surrounding mangrove forests may need special attention in relation to Hg even in sites without heavy Hg contamination (Kawagushi *et al*., 1999).

4. Conclusions

Sediment profiles showed a more or less evident increase in Hg concentration from low (or background) levels at lower sediment depths to pronounced levels at surface layers in mud flat and tidal creek sediments, or slight subsurface peaks in mangrove forest sediments. Mud flat and tidal creek sediments seem to have a Hg distribution largely affected by contamination, whereas Hg distribution appears to be greatly affected by root-sediment interactions in mangrove forest sediments. The study site showed Hg levels similar to those observed in coastal sediments affected by diffuse Hg sources at local (Sepetiba Bay) and regional (Rio de Janeiro State) scales. This indicates that Hg concentrations (and possibly its storage) within the Itacuruçá mangrove area may reflect the general degree of Hg contamination in Sepetiba Bay, as may be expected because of the diffuse nature of Hg input in this area. Our results suggest that sedimentary environments surrounding the mangrove forest are retaining most of the anthropogenic Hg load in these ecosystems. Since tidal waters were characterized as the main source of heavy metal input in the study site (Lacerda *et al*., 1988), the Hg retention in mud flat sediments seems to precede and can avoid a higher Hg accumulation in adjacent tidal creek and mangrove forest environments.

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