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Impact Analysis of Soil Flushing Remediation Technique on Chemical Properties and Metals Contents of Hydrocarbon-contaminated Mudflat in Bodo Creek, Rivers State, Nigeria

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ABSTRACT: Unpolluted, polluted and remediated mudflat samples were collected in Bodo Creek, Rivers State, Nigeria to evaluate the ecological status of the area using impact analysis of sediment remediation by flushing technique on chemical properties and levels of some metals. Results showed that total organic carbon ranged from 21.5 – 23.0 % from the unpolluted to the remediated station, available nitrogen was 2.4 % and 2.5 % for the unpolluted and the polluted/remediated stations, total organic matter was 35 % and 40 % for unpolluted and polluted/remediated stations while available phosphorus ranged from 0.03 to 0.08 mg/kg for remediated to polluted/unpolluted stations. Sodium content ranged from 134 to 137 for unpolluted to polluted/remediated stations, potassium was 1779 mg/kg for unpolluted and 1370 mg/kg for the remediated station, calcium was 224 mg/kg for unpolluted and 296 mg/kg for the remediated station while Pb Cu were below detectable limits. It can be concluded that remediated station is safe ecosystem re-establishment.

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Wetlands are unique ecosystems (Giri *et al*., 2011; Hogarth, 2015). They are a transition ecosystems between terrestrial and marine environments that have interconnection (Clough, 2013). According to Clough (2013), wetlands such as mangrove swamps are usually located in saltwater flooded areas such as coastlines of seas, margins of rivers and creeks, large deltas, estuaries, lagoons etc. These environments are common in tropical and subtropical regions of the world. Wetlands support biologically diverse ecosystems and have vegetation dominated by plants that are adapted to living in water environments. This ecosystem supports high biodiversity because they are rich in nutrients inputs from neighbouring land and water environments. Because of their nutrient rich status, they are highly productive ecosystems and support intricate interactions between its components. Furthermore, they hold significant stock of organic carbon in biomass because of their nutrient-rich status and diverse biotic contents (Alongi, 2014). Nigeria's main wetland, the mangrove swamp is located at the southern part of the country popularly called the Niger Delta which covers a political areas called Rivers, Bayelsa, Akwa Ibom, Delta States etc. is the largest in Africa (Aju and Aju, 2021). The mangrove swamp of Rivers State is fragmented into many formations due the deltaic nature of the region. Bodo, an Ogoni town in Rivers State has one of the major formations in the Niger Delta. This ecosystem is endowed with many bioresources which forms the basis of some socio-economic activities e.g. fishing, hunting, timber harvest, tourism and other forest resources harvest etc.

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of the people of the area (Uddin *et al*., 2013). Beside the rich biotic resources of the area, it has huge deposits of petroleum hydrocarbon (i.e. crude oil) resources; because of which, Bodo has been home to Shell Petroleum Development Company (SPDC) since 1958 when it discovered and started exploiting oil on commercial scale. However, this important ecosystem is facing many threats. Foremost of the threats is petroleum hydrocarbon pollution. For decades now, the environment has experienced severe environmental petroleum hydrocarbon pollution. Crude oil contamination has been a frequent occurrence in the area since the start of crude oil exploitation at Bodo. This condition prompted SPDC to carry out clean-up of the pollution using *in-situ* sediment flushing remediation technique which lasted six (6) months. The objective of the *in-situ* flushing was, to significantly reduce concentration of the pollutant in sediment as this would enable early initiation of natural attenuation over a short time which will render the polluted area cleaned up for revegetation and restoration of the ecosystem. After the remediation exercise in 2018, no report has been made on the status of the areas declared remediated. Hence, the objective of this study is to assess the impact soil flushing remediation treatment on the physicochemical status of Bodo Creek sediment in Rivers State, Nigeria.

MATERIALS AND METHODS

The Study Area: Bodo Mangrove Swamp is located on Bodo flood plain popularly called Bodo Creek (Pegg and Zabbey, 2013). The flood plain consists of four (4) interconnected brackish water channels, namely: Kpador, Koola Seato, Dor Nwezor and Koola Tobsoi. These channels conduct and drain water into and out of Bodo Creek (Zabbey and Babatunde, 2015). This area is beside the Bonny River which connects down to the Atlantic Ocean at Bonny, hence, Bodo mangrove swamp is north of Bonny (Fig. I).

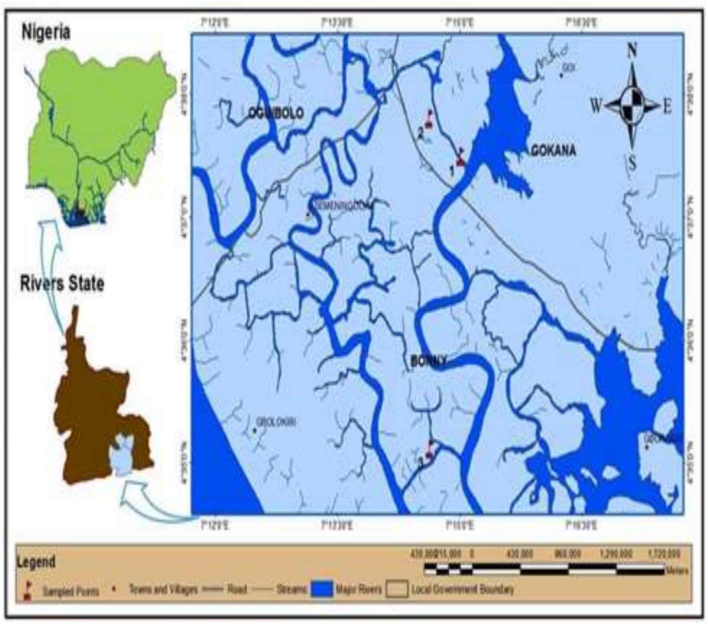

Fig. 1: Map of Gokana Local Government Area, Rivers State showing the study site
Source: Gbaa *et al.* (2020)

This post-remediation assessment was carried out on Kpador Channel. This area lies between latitudes 4^o 58’ 52.2” E to 4^o 63’ 00.5” E and longitudes 7^o 24’ 38.1” N to 7^o 25’ 38.9” N (Fig. I). Within this study area, three (3) sites located on intertidal zone of the Kpador Channel but are separated from each other were by random selected, marked out and used in this study as sample sites for sample collection. Sites 1 and 2 are located on Kpador Creek while site 3 is located 1.5 km away from Kpador Creek but closer to Bonny River. These sites are each separately described as follows.

Sampled Station I: This station is located at latitude 4^o 62’ 49.1” E and longitude 7^o 25’ 02.2” N (Fig. 1). The site was contaminated during the crude oil spill of 2008 and has since after the spill incident remained uncleaned spill till date. As a result of the spill, the site is bare of vegetation cover but has debris of mangrove plants killed by the impact of hydrocarbon pollution (Plate 1a). At the time of this assessment, the sediment of this site had scum of green algal growth which are taken as early colonizers of the ecosystem after destruction of the ecosystem by the petroleum hydrocarbon spill.

Sampled Station II: Station II is located at latitude 4º 63’ 00.5” N and longitude 7º 24’ 38.1” E (Fig. 1). This site was contaminated with petroleum hydrocarbon spill but was declared cleaned by SPDC after sediment flushing remediation with surfactant exercise. The remediation exercise was carried out between September, 2017 and March, 2018 and this was followed with site revegetation. The revegetation exercise was unsuccessful as all seedlings planted died, as a result, this site is void of vegetation cover, but have debris of dead plant materials killed by the petroleum hydrocarbon pollution incident (Plate 1b).

Sampled Station III: Station III is located between latitude 4º 58’ 52.0” E and longitude 7º 24’ 38.8” N (Fig. 1). This site was not impacted by any crude oil pollution of Bodo Creek, therefore it was used as control site in this study. This site had natural growth of mangrove plants which is composed of *Acrostichum aureum*, *Rhizophora racemosa*, *Rhizophora harrisonii*, *Rhizophora mangle* on its shore. These species dominated the vegetation of this site (Plate 1c).

Plate 1: Sample station (***a***) polluted station, (***b***) Remediated station (***c***) Unpolluted station
Source: Gbaa *et al.* (2020)

Samples Collection and Analyses: One 5 m x 5 m sample plot each, was mapped out at the polluted, remediated and unpolluted stations. By means of soil auger, triplicate sediment samples were collected by random within the area at 0 – 15 cm of sediment depth. Samples for each station were placed in separate polyethene bag, labelled and kept in ice-parked cooler to preserve them in their natural conditions at the time of collection. Each sample was divided into two parts: one part was taken to Plant Anatomy and Physiology Research Laboratory, University of Port Harcourt for analysis of Total Organic Carbon (TOC), Available Nitrogen (AN) and Available Phosphorus (AP) contents and the other half was used for metals (Na, lead, cadmium, iron, zinc and copper) contents analysis at AnalConcept Laboratory, Port Harcourt.

Parameters of the Study and Analytical Methods: Total organic carbon (TOC) and available nitrogen (AN) were determined using Walkley and Black

(1934) and Kjeldahl methods respectively. Carbon-nitrogen ratio (C/N) was obtained by calculation through dividing sample TOC with its Available Nitrogen content as expressed in equation (1) while Organic Matter (OM) was calculated using the formula of Estefan *et al.* (2013) stated as equation (2).

$$C/N = {}^{TOC}/_{AN} \quad (1)$$

$$OM\ (\%) = 1.724 \times TOC \quad (2)$$

Where C/N = Carbon nitrogen ratio; OM = Organic Matter

Available Phosphorus (AP) was determined by colorimetry after chemical reduction of ammonium molybdate with ascorbic acid and its concentration measured with flame photometer (Sherwood model 410). Exchangeable Sodium (ES) was obtained after extraction with ammonium acetate solution and its concentration measured by flame photometry with flame photometer (Sherwood, 410 model) while the concentration of other metals (lead, cadmium, iron, zinc and copper) were analysed after acid digestion of samples with nitric acid (HNO_3) and the concentration of each metal obtained from the digest using atomic absorption spectrophotometer, AAS (GBC Avanta model).

Analyses and Presentation of Data: Means and standard error mean (SEM) were calculated from data collected. Results were statistically analyzed using Minitab 17 and One-way Analysis of Variance (ANOVA) was used to test significant difference(s) between means of sampled stations at $P < 0.05$. Statistical significance of differences between means of assessment variables of sampled stations were determined with Tukey's HSD test. Results for Total Organic Carbon (TOC), Available Nitrogen (AN), Carbon-nitrogen Ratio (C/N), Total Organic Matter (TOM) and Available Phosphorus (AP) are described in charts which bar represents sample mean, error bar for SEM and alphabetical superscripts were used to represent levels of statistical difference between means at $P < 0.05$ while results of metals content are presented in composite table.

RESULTS AND DISCUSSION

Total Organic Carbon (TOC): Sediment samples were collected from the stations for analysis and determination and compare TOC contents. Result obtained are displayed in Fig. 2. Polluted station had TOC of 23.13%, 21.48% for the remediated station and 21.43% for the unpolluted station (Fig. 2). Comparative analysis of this result showed 1.65% decrease in TOC between polluted/remediated stations, 1.7% decrease between polluted/unpolluted station and 0.05% between remediated/unpolluted station and no significant statistical difference between stations (Fig. 2). Although there was no significant difference between station TOCs, however, TOC was higher in the polluted station than the others. The higher TOC of the polluted station could have resulted from possible input of organic carbon from the hydrocarbon spill incident that was not cleaned. Furthermore, lower TOC of the remediated station implies that the flushing remediation was efficacious as it restored TOC to same concentration as the unpolluted station in line with Gbaa *et al.* (2020) and Jez *et al.* (2021) which evaluated soil flushing remediation technology.

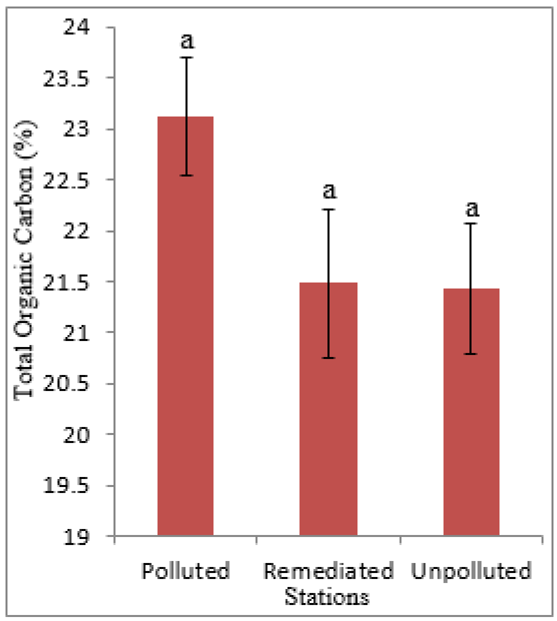

Fig. 2: Total organic carbon contents of sampled stations

Available Nitrogen (AN): Available nitrogen, AN contents of the sampled stations are presented in Fig. 3. Nitrogen content was slightly higher in the polluted and remediated station compared with the unpolluted station. This suggests that the pollution increased nitrogen. AN increased by 0.22% and 0.25% between polluted/unpolluted and remediated/unpolluted station respectively. However, there was no significant statistical difference in AN contents between the sampled sites (Fig. 3). The observed increase AN between the unpolluted and the polluted stations varies with the findings of John *et al.* (2016) which investigated the impact of crude oil on soil nitrogen dynamics on wetland ultisol of the Niger Delta, Nigeria. The likely chemistry to explain the observed increase could be that there was release of nitrogen into sediment from the crude oil.

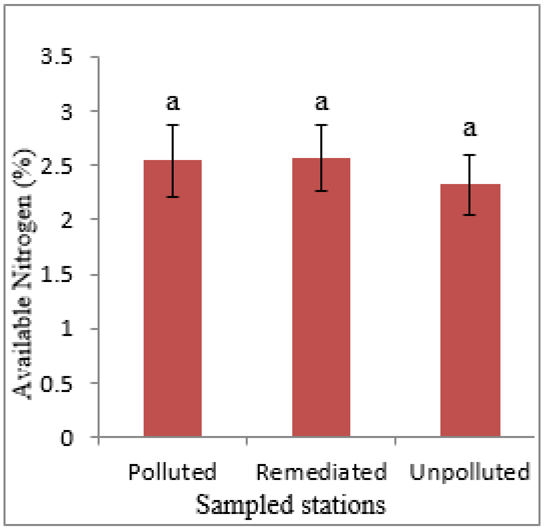

Fig. 3: Available nitrogen of sampled stations

Carbon/Nitrogen Ratio (C/N): The general C/N results for the sampled stations are presented in Fig. 4. C/N results were generally high for all stations. The polluted and unpolluted sites have C/N ratios of 9.51% and 9.47% respectively while the remediated site has 8.53%. This implies that pollution increased C/N ratio while remediation decreased it. There were no significant statistical differences between C/N ratios between sites. This means that the petroleum hydrocarbon contamination and its remediation by soil flushing had no significant effects on sediment TOC and AN.

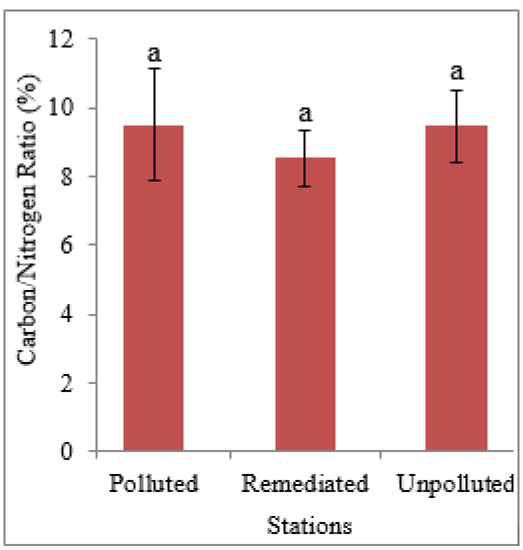

Fig. 4: Carbon/nitrogen of sampled stations

Total Organic Matter (TOM): Details of total organic matter are shown in Fig. 5. Total organic matter was highest (i.e. 40.19±2.86%) on the polluted station, 36.38±2.60% and 36.43±1.81% for the remediated and the unpolluted stations respectively. However, there was no significant statistical difference in TOM between stations. Total organic matter content is one of the important soil/sediment properties which plays role soil chemical interactions between soil particles and organic pollutant. Results obtained from this assessment showed approximate increase in TOM of the polluted station over the unpolluted station. This increase is in line with and was explained by Mendez *et al.* (2017) to result from hydrocarbon pollutants which adhered to sediment particle were retained by the smallest particle group.

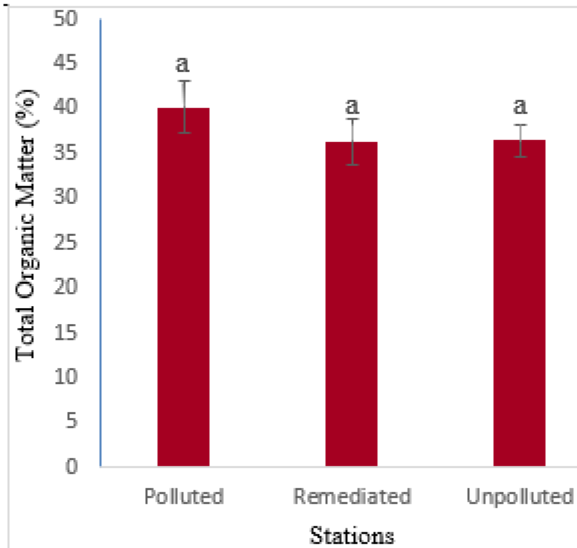

Fig. 5: Total organic matter of sampled stations

Available Phosphorus (AP): Sampled stations available phosphorus, AP results are displayed in Fig. 6. The unpolluted station had the highest AP content (i.e. 0.089 mg/kg) which is closed followed by the polluted station with 0.082 mg/kg and the least AP, 0.035 mg/kg from the remediated station. There are no significant statistical difference between station AP contents (Fig. 6).

The observed reduction in AP between the polluted and remediated stations can be attributed to processes such as immobilization of the mineral by organic component of the flushing agent or adsorption to fine clay particles, washing away of the mineral in runoff or leaching down soil profile after precipitation (Prasad and Chakraborty, 2019).

These processes decrease phosphorus availability in soil and sediment. The reduction in AP of the remediated station could have also resulted from formation of phosphate complexes with organic compounds of the soil flushing agent used in the remediation treatment.

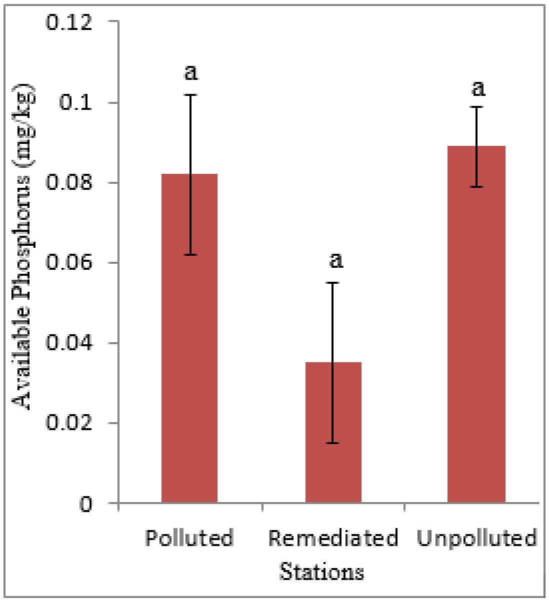

Fig. 6: Available phosphorus of sampled stations

Metals Content (mg/kg): In general, soil flushing had varied and more significant influence on metals content than the other sediment properties such as TOC, TOM, AN, and AP because soil-washing agents such as inorganic acids, bases, salts, synthetic surfactants and synthetic chelators enhance soil metals' desorption and removal from soil which may have adverse effects on soil. The results for metals in sediment across all sites in Bodo mangrove wetland are shown in Table 1. Sodium, Na content of the remediated station was 137.53 mg/kg and 134 kg for the polluted and unpolluted stations. As shown in Table 1, there was no significant statistical difference between the polluted and unpolluted stations but between the remediated station and the others (Table 1). The higher Na content of the remediated station suggests that it could have been added from the soil-washing agent used in the remediation. This is plausible because the stations are located within same geological formation and therefore, without any addition from an external source, the concentration of the cation should have no significant difference between stations. The contention that the higher content of the mineral at the remediated is from the soil-washing agent is justified by the equal concentration of the metal at the polluted and the unpolluted stations. The chemistry responsible for this observation can be explained by Diao *et al.* (2017) in which it was stated that desorption and complexation of cations through surfactant-associated chemical reactions can lead to the increase. Potassium (K) content showed variation between the sampling stations. High potassium content of 1779.02 mg/kg was recorded in the unpolluted station. Both polluted and remediated stations were observed to have 1500.72 mg/kg and 1370.18 mg/kg respectively. The reduction in metal concentration at the polluted stations could likely be due to the chemical processes of dissolution and subsequent out-washing of the nutrient by tidal water or, by leaching of the cation down soil profile. Soil-washing agent could also form chemical complex which resulted in the further reduction of the remediated station. Calcium (Ca) also showed significant statistical variation between the sampled stations. The trend of concentration of the metal showed that the polluted station had the highest while the unpolluted station had the least concentration of the metal. This result suggests that the pollutant (crude oil) spill increased concentration of the metal at the station. Further in the concentration of the metal at the remediated station means the soil-washing agent used may have introduce more calcium to sediment leading to this observation. Magnesium result also showed variation between the stations. The polluted station was observed to record 1280.45 mg/kg, the remediated station had the highest amount of magnesium concentration (368.03 mg/kg) which was closely followed by the unpolluted station which recorded 1352.63 mg/kg of the cation. Mg result does not show any trend and therefore cannot be attributed to the effects of pollution and remediation. Cadmium (Cd) and copper (Cu) were observed to be below detectable limits across all sites. The concentration of iron (Fe) varied significantly between the stations. The unpolluted station had the highest concentration (15368.10 mg/kg) of the mineral across all the stations sampled. Polluted and remediated stations recorded 10068.65 mg/kg and 8310.92 mg/kg respectively. The trend of this result showed that the concentration of the cation decrease from the unpolluted station to the polluted station and further reduction at the remediated station. The implication of this is that the pollution and sediment flushing with the surfactant may have caused complexation of the cation molecules by some organic compounds in the pollutant and surfactant leading to the recorded reduction in the concentration of the cation at both stations compared with the unpolluted station. However, zinc (Zn) result showed that pollution had effect on its concentration but, the soil flushing remediation caused significant reduction in the concentration of the cation. Lead, Pb was not detected at the polluted and the remediated stations but was detected at the unpolluted station. The low concentration of the mineral at the unpolluted station and below detection limit of Pb at the polluted and remediated stations is an indication of the low concentration of the cation in the parent rock from the soil of the area is derived.

Table 1: Metal contents of sample station sediments

Metal	Concentration (mg/kg)		
	Polluted	Remediated	Unpolluted
Exchangeable Sodium (Na)	134.33±0.34[b]	137.53±0.32[a]	134.2±0.22[b]
Exchangeable Potassium (K)	1500.72±0.31[b]	1370.18±0.56[c]	1779.02±0.30[a]
Exchangeable Calcium (Ca)	284.25±0.54[b]	296.48±0.29[a]	224.52±0.32[c]
Exchangeable Magnesium (Mg)	1280.45±0.33[c]	1368.03±0.55[a]	1352.63±0.27[b]
Cadmium (Cd)	<0.001[a]	<0.001[a]	<0.001[a]
Iron (Fe)	10068.65±0.33[b]	8310.92±0.25[c]	15368.10±0.15[a]
Zinc (Zn)	61.68±0.35[a]	48.27±0.66[b]	61.31±0.34[a]
Lead (Pb)	< 0.001[b]	< 0.001[b]	3.37±0.31[a]
Copper (Cu)	< 0.001[a]	< 0.001[a]	< 0.001[a]

Conclusion: This study assessed effects of soil flushing pollution remediation technique on some physico-chemical properties and metals contents of hydrocarbon-polluted Bodo Creek mudflat. Was found that it did not alter sediment TOC, AN, C/N, TOM and Na and Mg contents. However, it reduced sediment AP concentration content. It was observed to increase Ca content and reduced K concentration. It can be concluded that soil flushing remediation had no detrimental impact on Bodo Creek mudflat. Therefore, the remediated mudflat is safe for ecosystem re-establishment.

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