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Transport and fate of microplastics from riverine sediment dredge piles: Implications for disposal

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ABSTRACT

Microplastics (MPs) are an environmental problem of growing concern. Aquatic sediments are considered as a final sink for MPs, but dredging can remobilize sedimentary MPs into both aquatic and terrestrial ecosystems. Although dredging is globally used for waterway deepening and ecological restoration, the environmental impacts of dredging on MP pollutants has not been previously assessed. In this study, Nile Red staining combined with micro-FTIR methods showed sediments containing high MP concentrations (6060–37610 n/kg·DW) from urban/suburban segments of a plain river network in Southeast China. The dredged sediments were stored in piles on farmlands, whereby MPs were subsequently dispersed to surrounding soils and surface waters while awaiting a permanent disposal option. MP concentrations in the soils surrounding the pile were higher in the dry season (wind dispersion), while MP concentrations in waters downstream of the piles were higher in the wet season (rainfall/runoff erosion). Whether dredge sediments are finally used to fertilize farmland, as fill material for coastal land reclamation or dumped into the ocean, MPs have the potential for remobilization into the environment causing concerns with aquatic food webs, agricultural production and human health. Therefore, disposal of dredge sediments containing MPs requires careful assessment to minimize potential environmental impacts.

1. Introduction

Plastic pollution is regarded as a global environmental challenge given its increasing production/use coupled with poor waste/recycling management (UNEP, 2018). Plastic particles smaller than 5 mm, termed microplastics (MPs), are of special concern due to their potential harm to ecosystems. MPs are often categorized as primary (produced to be of microscopic dimensions) or secondary (resulting from degradation and fragmentation processes in the environment) (Andrady, 2017). After release to the environment, MPs may continue to break down into smaller particles over time (Weinstein et al., 2016), making them ingestible at all levels of the aquatic food web (Cole et al., 2013; Woods et al., 2018; Windsor et al., 2019).

The pervasiveness of MPs in marine systems is well documented, while an increasing number of studies show a prevalence of MPs in freshwater systems as well. Sediment is generally considered as final sink for MPs (Nizzetto et al., 2016). MPs with a density higher than water are more likely to sink and incorporated into the sediment, while MPs lighter than water can also be deposited after biofouling (Fazey and Ryan, 2016), ingestion by aquatic animals and egestion in feces (Cole et al., 2013), and aggregation with biogenic materials (Long et al., 2015; Michels et al., 2018) or suspended sediments (Li et al., 2019). As a result, very high concentrations of MPs (>10,000 per kg) can be found in marine (Bergmann et al., 2017; Kane et al., 2020) and freshwater sediments (Wang et al., 2018; Mani et al., 2019; González-Pleiter et al., 2020).

After deposition in sediments, the dark and cold benthic environment prohibits the degradation of plastics (Tansel, 2019). Plastics are degraded through abiotic (photo-degradation, thermal-degradation, oxidative-degradation, hydrolytic-degradation, etc.) and biotic factors

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once in the environment. Among these processes, UV photo-degradation is considered as the most effective mechanism for rapid environmental degradation of plastic polymers (Andrady, 2017). Solar radiation initiates an autocatalytic thermal oxidation reaction that is primarily responsible for the degradation process. While biodegradation (and even hydrolysis) does occur in marine systems (Zettler et al., 2013), the reactions proceed too slowly to result in significant levels of plastic degradation in deep water and sediments (Andrady, 2011; Muthukumar et al., 2011). Further, the anaerobic environment of sediments contributes to slow degradation kinetics involving coupling of thermodynamically favorable and unfavorable reactions and biochemical transformations by microbes. Hence, the resulting degradation environment makes the deposited plastics "quiescent" compare to plastics exposed on land or surface waters.

Notably, deposited plastics may be "re-activated" by disturbance of the sediment-water interface caused by hydrological or biological processes. For example, MPs in the sediments can be resuspended and flushed from river catchments during high-flow flood events (Hurley et al., 2018). Further, MPs in the sediments can be transported upward to the sediment-water interface by bioturbation of benthic macrofauna (Näkki et al., 2019). In addition to natural processes, anthropogenic activities, such as dredging, can drastically alter the sediment environment and entrain particulate pollutants into the water column for subsequent fluvial transport (Berenjkar et al., 2019). A recent study revealed that the distribution of MPs in the bottom sediment was strongly impacted by the dumping of dredged sediments (Baptista Neto et al., 2019). Given the prevalence of dredging in many coastal areas (Bianchini et al., 2019), research is required to assess the transport and fate of MPs in dredged sediments during their collection, storage and ultimate disposal.

Dredging is widely used for improving and sustaining the navigational depth of waterways, harbors and estuaries, as well as for the ecological restoration and remediation of rivers and lakes. Notably, the excavation, transport and disposal of soft-bottom sediments lead to various adverse impacts on the aquatic environment (Erftemeijer and Robin Lewis, 2006). The amount of sediment dredged in China is more than 5 billion m^3 per year. Dredging is a key method to reduce benthic pollutants and eliminate black, odorous surface waters in China. For example, about 136 and 116 million m^3 of riverine sediment was removed for restoration purposes in Zhejiang Province in 2016 and 2017, respectively (Zhejiang Provincial Bureau of Statistics, 2016, 2017). A major dilemma for dredging programs is how to handle the dredged sediments, which are often highly enriched with pollutants (e.g., MPs, heavy metals, pesticides, PAHs). Common disposal practices include landfill for coastal reclamation and ocean dumping, especially in coastal cities. Due to the high cost of transportation and environmental policies, it is common for a large portion of dredged sediments to be placed in storage piles near the dredging site while awaiting a long-term disposal option. For example, in Yueqing City-Wenzhou (our study site), ~3 out of the total 10 million m^3 of river sediments dredged from 2014 to 2016 was transported to Ou River Estuary for use as landfill in a coastal reclamation project, while the remaining 7 million m^3 was piled for storage on more than 400 sites covering nearly 600 ha of farmland near river dredging sites. Migration and diffusion of pollutants from these sediment storage piles are of great environmental concern, including the transport and fate of plastic pollutants.

Our previous study documented large amounts of MPs deposited in sediments of the plain river network in Wenzhou (~1800 km of waterways) (Wang et al., 2018). Based on this knowledge, we were compelled to examine the occurrence, transport and fate of MPs in dredged sediment piles to evaluate the environmental impact on the surrounding area. In the present study, the MPs in and around two typical sediment piles were investigated at two time points (winter dry season & spring wet season) to elucidate the potential environmental impacts of the MPs associated with these piles.

2. Materials and methods

2.1. Study area

We investigated storage piles of dredged sediments collected from two plain rivers in Yuechen (urban population 152,000) and Huangtian (suburban population 28,000) of Wenzhou, Zhejiang Province, southeast China (Fig. S1). These rivers have minimal flow velocity during the winter dry season when the outlets to the Ou River Estuary are closed to prevent salt-water intrusion, and much larger flows when the outlets are open to the estuary during the spring and summer (monsoon) seasons. Flow velocity at both sites was < 0.02 m s^{-1} during the winter survey and 0.2–0.3 m s^{-1} during the spring survey. The low flow during much of the year results in high sediment deposition within the river channel.

To remove sediments and associated pollutants, the rivers were drained in sections of 500–800 m for subsequent removal of the silt-dominated sediment layer (50–120 cm) on the riverbed using a bucket excavator. Dredged sediment was stored in piles adjacent to the rivers at a distance of ~90 m at Yuechen (Pile 1, P1, 120°58'58.73"E, 28°5'58.00"N) and ~160 m at Huangtian (Pile 2, P2, 120°41'35.81"E, 28°4'36.51"N). The dimensions of the storage piles (length × wide × height) were P1 = 200 × 60 × 2 m and P2 = 80 × 35 × 1.8 m. The area containing the sediment piles was separated from the surrounding farmland by ditches that collected runoff from the piles. The ditches were nearly dry during the winter survey, but had visible flow during the spring survey.

2.2. Sampling

Field surveys were carried out in December 2016 (dry season) and May 2017 (wet season) at both sites (Fig. 1). Surface (0–3 cm) and subsurface (30 cm depth) samples were collected from 3 randomly selected locations at 3 positions (top, slope and foot) on the sediment pile. To examine potential MP contamination of the surrounding area by the dredge storage pile, the surface sediments (0–3 cm) were collected from 3 sites in the adjacent ditches and from 3 sites in ditches of the surrounding farmland (200–300 m from piles). Additionally, surface (0–3 cm) and subsurface (3–6 cm depth) soil samples from 6 farmland sites near the piles (~30 m) were compared to 3 reference farmland sites 200–300 m from the piles. For sediment and soil sampling, triplicate samples (~0.8 L) were collected using a stainless steel spade that was rinsed free of sediments by distilled water between each sample. This resulted in a total of 54 dredged sediments, 18 ditch sediments and 54 soil samples at each site during each survey. Samples were placed in 1 L glass jars for transport to the laboratory. All sampling equipment and glassware were washed thoroughly with distilled water before sampling to avoid contamination.

Surface water samples were collected from the nearby river (the sediment source river) at 5 longitudinally-distributed sites: 500 m upstream, 200 m upstream, just downstream of ditch input, 200 m downstream and 500 m downstream. Triplicate 100 L samples were collected using a stainless steel bucket resulting in a total of 15 water samples per site. Water samples were immediately filtered through a 25 μm stainless steel sieve, and the residue on the sieve was carefully transferred with distilled water into 250 ml glass jars for further processing.

2.3. Sample preparation

For sediment/soil samples, 500 g field-moist sediment was dried at 60 °C for 48 h. The triplicate samples from each site/location were then pooled to give a total of 24 surface and 18 subsurface composite samples from each sediment pile. A 100 g subsample from each well-mixed composite sample was sieved sequentially through 5 mm and 25 μm stainless-steel sieves to isolate plastic particles in the 25 μm to 5 mm size range (defined as the MP fraction in this study). Water samples,

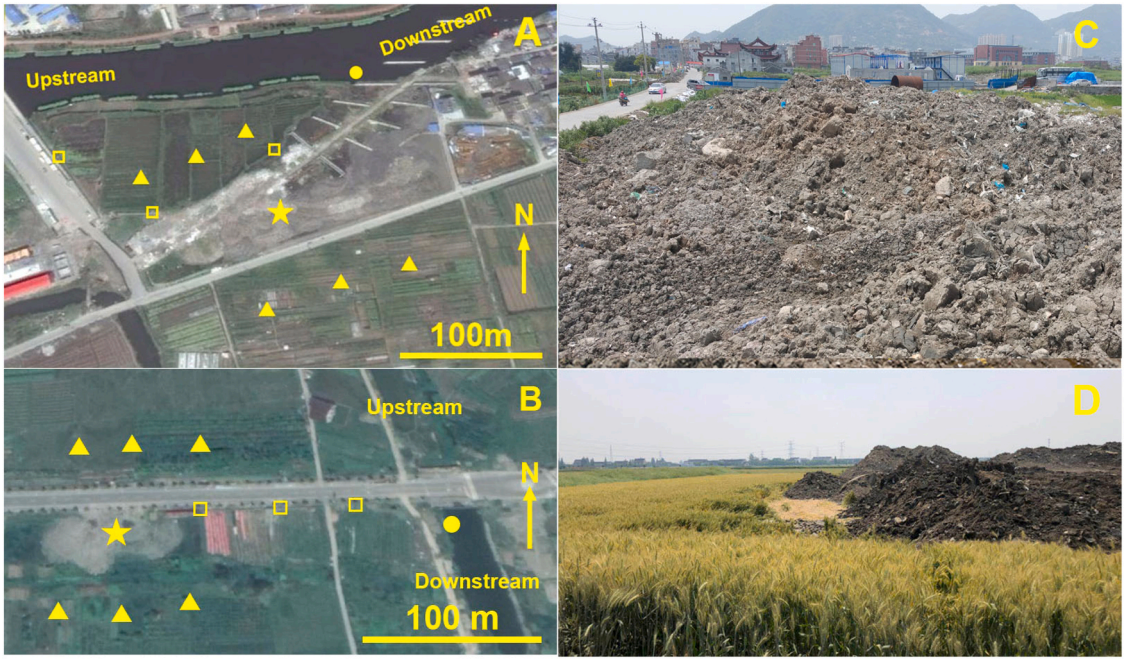

Fig. 1. Sampling locations around dredged sediment piles. Distribution of sampling points at (A) P1 (urban) and (B) P2 (suburban) sites; Landscape view at (C) P1 and (D) P2. Star: sediment pile; hollow block: adjacent ditch; solid triangle: adjacent soil; solid circle: downstream of ditch input.

previously passed through a 25 μm sieve in the field, were passed through a 5 mm stainless-steel sieve to remove large particles and isolate the 25 μm to 5 mm fraction.

The isolated fractions from sediment, soil and water samples were treated with 30% hydrogen peroxide at 70 °C for 2 h to oxidize natural organic matter (Masura et al., 2015). Density separation using a $ZnCl_2$ solution (density = 1.7 g ml^{-1}) was utilized to separate MPs through floatation (Imhof et al., 2012). The density separation step was repeated and supernatants combined; the combined separation/extract was > 90% efficient in isolating the MP fraction (see Supplementary materials). The supernatant with isolated MPs was filtered through a 0.4 μm polycarbonate membrane filter (HTTP04700, Millipore) to collect particles for further identification.

2.4. Analysis

A combined method of Micro Fourier Transform Infrared Spectrometer analysis (μ-FT-IR) and Nile Red (NR) staining was applied to minimize false-positive misidentifications (i.e., non-plastics recorded as MPs). A total of 20–30 suspected plastic items on each filter were randomly selected to verify polymer composition using μ-FT-IR (VERTEX 70 plus HYPERION 2000; Bruker, USA) under transmittance mode. Resulting spectra were compared to a known polymer spectra library to identify the chemical composition using a criterion of at least 70% similarity for confirmation. Based on the μ-FT-IR results, we established a general rule for excluding non-plastic items in the visual examination using a microscope. Subsequently, MPs on the filter were stained with 3 drops of 5 mg ml^{-1} Nile Red (NR) for 30 min at room temperature (Shim et al., 2016), and then photographed using a fluorescence stereo microscope (M165FC, Leica) at up to 120 × magnification (Maes et al., 2017) (Fig. S2). NR-stainable items in the images were enumerated and measured using Image J (https://imagej.nih.gov/ij/). MPs were further classified according to their morphology as fragment (fragment of large plastic waste), fiber (fibrous plastic), pellet (industrial plastic pellet), or foam (sponge-like plastic), and also by size class (based on longest particle dimension): 20–100, 100–300, 300–1000 and 1000–5000 μm.

To avoid contamination from airborne MPs, extraction processes were performed in a laminar-flow hood, and all glassware was thoroughly cleaned before use. All samples and equipment were covered with glass petri dishes or aluminum foil after cleaning. Laboratory blanks of distilled water were repeated every day as a "negative control" during extraction and identification processes. Background contamination was deemed negligible (1.3 ± 1.4 MPs/filter, n = 46) compared to field samples (>100 MPs/filter).

One-way ANOVA followed by the Holm−Sidak all-pairwise multiple comparison test ($P < 0.05$) was used to assess spatial and seasonal differences in MP abundance in the dredged sediment, soil and river water using SPSS 20.0 (IBM, Armonk, NY, USA). All data are reported as mean±SD, unless otherwise stated. All "differences" referred to in presentation of the results denote a statistical significant of $P < 0.05$.

3. Results

3.1. Microplastics in dredge sediment piles

High MP contents were found in all samples of the two dredge sediments piles. MP concentrations in P1 (urban) subsurface samples were 13,710–37,610 n/kg·DW (24,784 ± 6953 n/kg·DW) (mean±std dev) in the dry season (winter) and 17,780–37,780 n/kg·DW (29,031 ± 5869 n/kg·DW) in the wet season (spring). The P2 (suburban) subsurface dredge sediments had a significantly lower MP content with 7020–14,670 n/ kg·DW (10,580 ± 2616 n/kg·DW) in winter and 6060–18,450 n/kg·DW (11,247 ± 4131 n/kg·DW) in spring. Average MP concentration in P1 surface samples was similar to subsurface samples in winter, but lower in the spring. No difference was found between surface and subsurface samples in P2 (Fig. 2).

Particles <300 μm constituted the majority of MPs in both piles: P1 = 75.4–91.1% and P2 = 71.3–89.7% (Fig. S3). The 1–5 mm fraction only accounted for 2.3–5.2% of total MPs in P1 samples and 2.4–5.0% in P2 samples (Fig. S3). The percentage of MPs in the 20–100 μm fraction for surface sediments of P1 in spring was lower than other samples in both surface and subsurface dredge sediments. Accordingly, the percentage of MPs in the 100–300 μm size fraction of the P1 surface sediment in spring was higher than most other samples except for P1 subsurface sediment in spring. The percentage of MPs in the 300–1000 μm size fraction of the P1 surface sediment in spring was

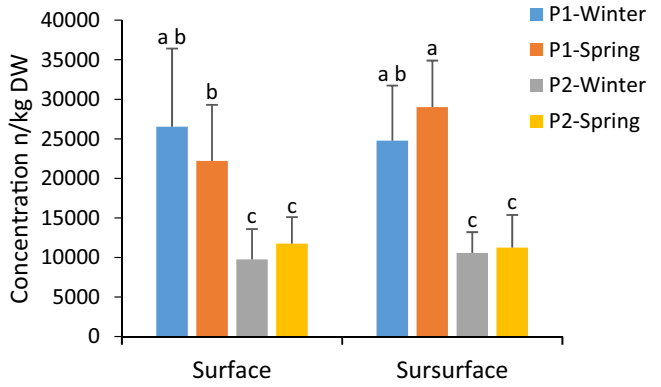

Fig. 2. Microplastic concentration in surface and subsurface dredge sediments of P1 and P2 during winter (dry season) and spring (wet season) surveys (mean ± std dev). Samples with different lower case letters are significantly different at $P < 0.05$.

higher than most other samples except P1 surface sediment in winter. Finally, the percentage of MPs in the 1000–5000 μm size fraction of the P1 surface sediment in spring was higher than most other samples except P2 surface sediment in winter (Fig. S3).

The proportion of MP morphological types in both piles followed fragments > fibers > pellets ≈ foams (Fig S4). The proportion of fragments in P1 (50.2% ± 5.0%) was lower than P2 samples (57.7% ± 4.7%). In contrast, P1 had slightly higher concentrations of fibers, pellets and foams (Fig. S4). The proportion of foams in the surface layer was lower than for subsurface sediments in both P1 and P2 (Fig. S5g, h). In winter, the proportion of fragments in the surface layer was higher than for subsurface sediments in P1 (Fig. S5a), while fibers in the surface layer were higher than subsurface sediments in P2 (Fig. S5d).

A total of 13 and 12 polymer types were identified by μ-FT-IR among randomly selected particles from P1 and P2 samples, respectively. The randomly selected MPs were dominated by PE (polyethylene, 25.5%), PP (polypropylene, 15.7%), PS (polystyrene, including EPS, 11.8%) and PA (nylon, 9.8%) in P1 samples, and by PE (20.8%), PA (16.7%), PS (14.6%) and PVC (polyvinyl chloride, 12.5%) in P2 samples (Figs. S6 and S7). The other identified polymers each accounted for less than 10% of the total particles analyzed.

3.2. Microplastics in the area surrounding the dredge sediment piles

3.2.1. Ditch sediments

The surface sediment of ditches adjacent to the dredge sediment piles (D1, D2) and ditches in reference farmlands (D1R, D2R) located 200–300 m from the piles all contained MPs. The average concentration of MPs was 12,730 ± 3625 n/kg·DW in D1 sediments in the dry (winter) season, which was lower than that in P1 surface samples. MP concentrations increased to 23,717 ± 5788 n/kg·DW in the wet (spring) season with concentrations similar to P1 surface samples (Fig. 3a). Average MP concentration (3383 ± 555 n/kg·DW) in D2 sediments during the wet season was also higher than that (2117 ± 527 n/kg·DW) in the dry season (Fig. 3b), but both concentrations were significantly lower than that in P2 surface samples. MP concentration in D1 sediments was significantly higher than that in D1R sediments during both dry and wet seasons (Fig. 3a). While the MP concentration in D2 sediments was similar to that in D2R sediments in the dry season, the MP concentration was higher in the wet season (Fig. 3b).

Similar to the size distribution of MPs in dredge sediment piles, MPs smaller than 300 μm constituted the majority of MPs (78.3–85.8% in D1 and 79.6–86.2% in D2) (Fig. S8). Large (1–5 mm size) MPs comprised <5% of total MPs in D1 and D2 sediments. There was no significant difference in MP size distributions of D1/D2 sediments between the two seasonal surveys. The proportion of small (<300 μm) MPs in D1R sediment was lower than that in D1 sediments, while the proportion of larger MPs was higher in D1R sediment (Fig. S8e, f). In contrast, the proportion of MP size groups was similar in D2 and D2R sediments (Fig. S8g, h).

The proportion of MP fragments in both ditch sediments was lower in the dry season (53.7 ± 2.8% of D1 and 55.0 ± 3.5% of D2) than that in wet season (65.9 ± 5.0% of D1 and 63.7 ± 4.2% of D2) (Fig. S9a). The proportion of fibers, pellets and foams were not different between the two sampling seasons. The only significant difference in morphological types between adjacent and reference ditches was a lower proportion of foams in D1 sediments than that in D1R sediments (Fig. S9b).

Polymer composition in D1 sediments was dominated by PE (23.5%), PS (13.7%), PP (11.8%) and PA (11.8%), while PP (23.1%), PS (21.2%), PE (15.4%) and PVC (13.5%) dominated in D2 sediments (Fig. S10). PE, PS, PP and PA were also common in reference ditch sediments of both sites (Fig. S10).

3.2.2. Farmland soil

MP concentrations in the farmland soil near the dredge sediment piles were significantly lower than that in the sediment piles at both sites. The average MP concentration in surface soil (0–3 cm) near P1 (SS1, 3135 ± 684 n/kg·DW) was higher than that near P2 (SS2, 1548 ± 529 n/kg·DW) in the dry season, but no difference was found in the wet season (Fig. 4a). MP concentrations in the subsoil (3–6 cm) (BS1, 1750 ± 432 n/kg·DW) were lower than that in SS1 in the dry season, while the MP concentrations in the two layers were similar in the wet season (Fig. 4a). There was no significant difference between MP concentrations in the surface (SS2) and subsoil (BS2) layers of soil near P2. The MP concentration in SS1 was higher in the dry season than that in wet season (2245 ± 619 n/kg·DW), and also higher than the MP concentrations in the reference surface soil (RSS1, 1853 ± 495 n/kg·DW) in dry season (Fig. 4b). There was no difference in MP concentration

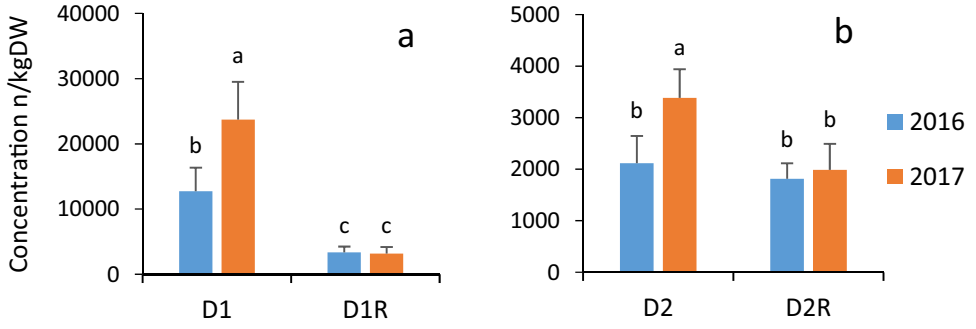

Fig. 3. Microplastic concentration in sediments of (a) adjacent (D1 & D2) and (b) reference (D1R & D2R) ditches (mean±std dev). Samples with different lower case letters are significantly different at $P < 0.05$.

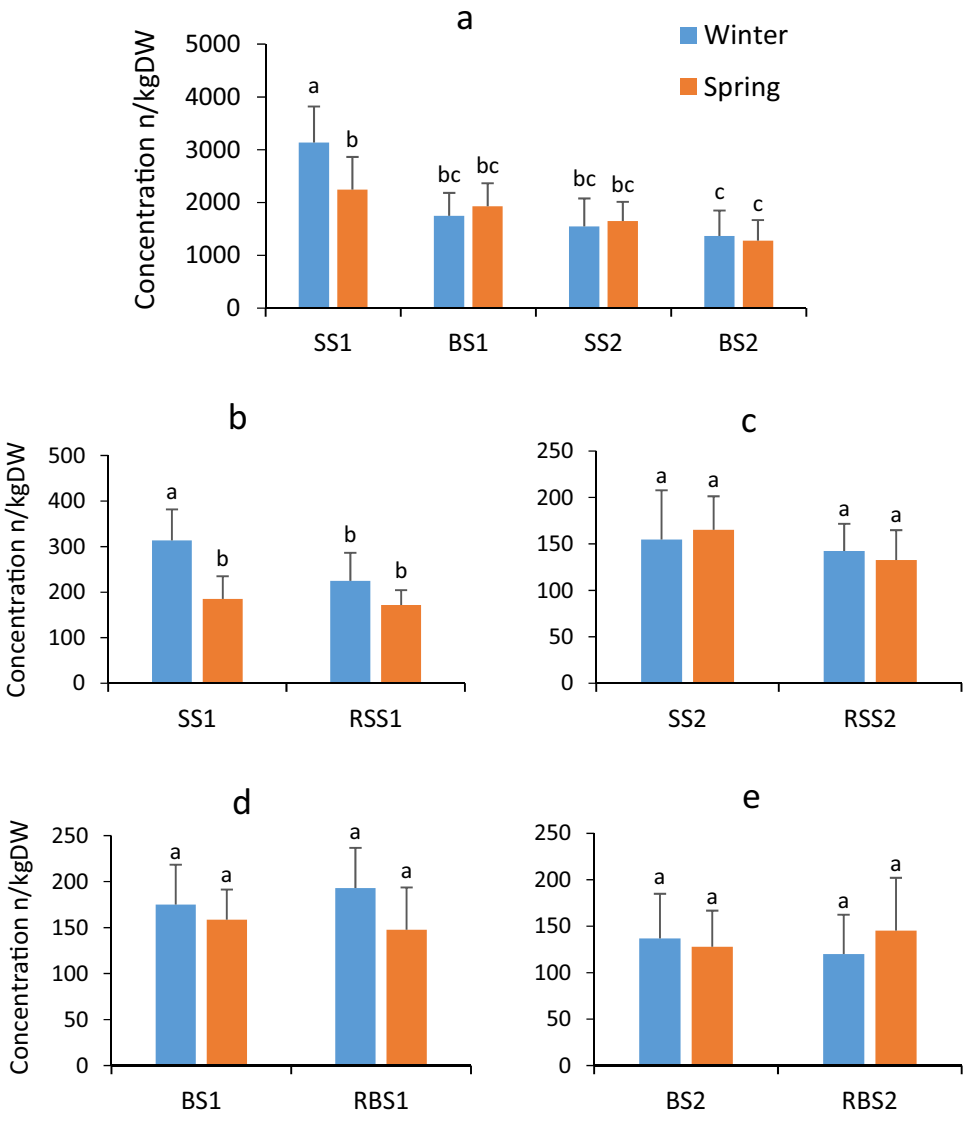

Fig. 4. Microplastic concentrations in surface and subsoil layers of soils adjacent to storage piles and in reference farmland located away from the storage piles (mean±std dev). a: surface and subsoil of adjacent farmland; b: surface soil of adjacent and reference farmland of P1; c: surface soil of adjacent and reference farmland of P2; d: subsoil of adjacent and reference farmland of P1; e: subsoil of adjacent and reference farmland of P2. Samples with different lower case letters are significantly different at $P < 0.05$. SS1: surface soil near P1; BS1: subsoil near P1; RSS1: reference surface soil of P1; RBS1: reference subsoil of P1; SS2: surface soil near P2; BS2: subsoil near P2; RSS2: reference surface soil of P2; RBS2: reference subsoil of P2.

between SS1 and RSS1 in the wet season (Fig. 4b). The concentration of MPs in both SS2 and BS2 were similar between seasons, as were the reference soil samples at both sites (Fig. 4c,e).

The proportion of small MPs (<300 μm) in SS1 (76.7–85.6%) was lower than that in BS1 (83.0–89.6%) during the dry season, but they were similar in the wet season (Fig. S11). No difference was found in the small MP proportion among P2 soil samples in either season. The small MP proportion in most surface and subsoil samples from reference farmland was similar to that in the farmland adjacent to the sediment storage piles, except BS1 was higher than RBS1 in the wet season (Fig. S11).

Fragments dominated the MP composition among all soil samples, followed by fibers, foams and pellets. The proportion of fibers was lower (23.2 ± 4.1% versus 31.2 ± 4.1%) and foams higher (20.1 ± 2.6% versus 11.5%±1.1%) when comparing SS1 with RSS1 in winter (Fig. S12a,b). A higher proportion of foams was also found in SS2 than that in RSS2 during both seasons (Fig. S12c).

The most common polymer compositions found in MPs of soil samples adjacent to P1 were PE (21.5%), PS (16.0%), PP (15.9%) and PVC (12.2%), while samples adjacent to P2 were comprised of PS (21.3%), PP (20.3%), PE (19.2%) and PA (9.6%) (Fig. S13). The PE, PP, PS and PA types were also common in the reference soils at both sites and followed a relatively similar distribution to the soils adjacent to the sediment piles (Fig. S13).

3.2.3. *Riverine water*

MPs were ubiquitous in surface waters in the vicinity of the sediment piles. MP concentrations for the 5 river sites were higher in wet season and sites near P1 were higher than P2 sites (Fig. 5). While there was no difference between MP concentrations of upstream and downstream sites in the dry season (Fig. 5a, c), MPs concentrations in downstream waters (all 9 samples) were higher than upstream waters (all 6 samples) in the wet season (Fig. 5b, d).

The proportion of MPs <300 μm in P1 river water was 77.7 ± 2.0% in the dry season, which was significantly lower than that in surface samples from the P1 sediment pile. In contrast, the wet season river samples had a higher proportion of small MPs at P1 (81.2 ± 2.5%) than that in P1 surface samples (Fig. S14a). The small MP proportion in the P2 river was 78.2 ± 3.3% and 80.6 ± 2.6% in the two seasons, both lower than that in P2 surface samples (Fig. S14b). The proportion of small MPs in upstream waters was lower than that in downstream waters in both P1 and P2 rivers in the wet season, but there was no difference between upstream and downstream waters in the dry season (Fig. S14c, d).

Similar to the morphological composition of MPs in ditch sediments, the proportion of fragments in dry season was lower than that in wet season for both rivers (Fig. S15a). The proportion of pellets was lower

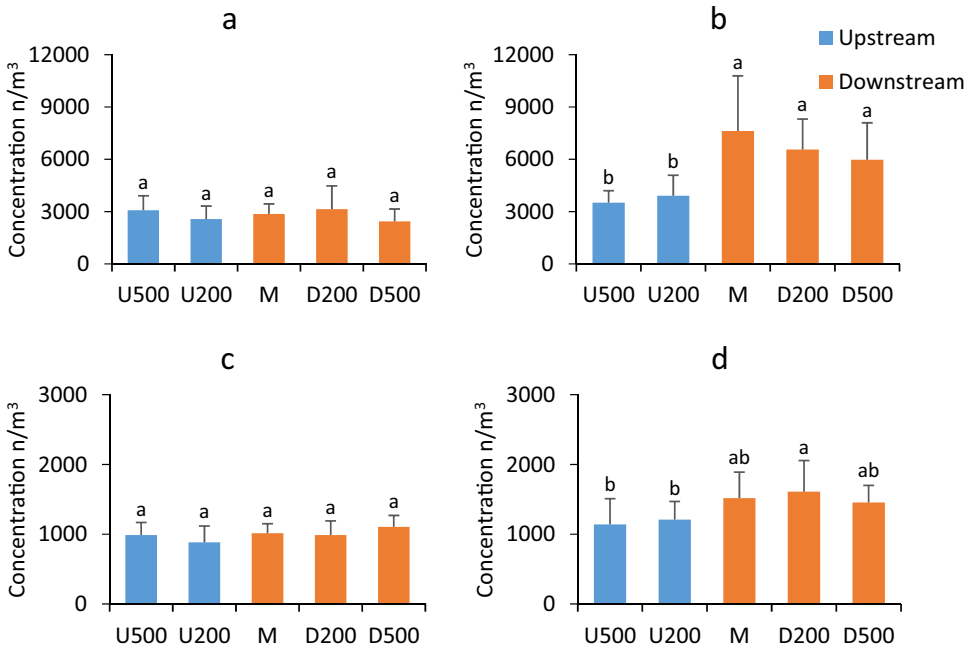

Fig. 5. Microplastic concentration in river waters near the dredge storage piles (mean ± std dev). U500: 500 m upstream; U200: 200 m upstream; M: just downstream of ditch input; D200: 200 m downstream D500: 500 m downstream. a: P1 in winter; b: P1 in spring; c: P2 in winter; d: P2 in spring. Samples with different lower case letters are significantly different at $P < 0.05$.

and foams higher during the dry season in P1 (Fig. S15c,d). The fiber proportion was higher in dry season for both rivers (Fig. S15b). No differences were found between morphological composition of upstream and downstream waters in the dry season. However, the proportion of fragments was higher, and the proportion of fibers and foams was lower in downstream waters of the P1 river in wet season (Fig. S16). The morphological composition was similar for upstream and downstream waters of the P2 river, except the fiber proportion was lower in downstream waters in the wet season (Fig. S16).

Dominant polymer types in the P1 nearby river were PS (23.5%) and PE (21.6%) during the dry season and PE (27.7%) and PA (21.3%) in the wet season (Fig. S17). The most common polymers found in the P2 river were PE (28.6%) and PP (20.4%) in dry season and PE (29.8%) and PS (27.7%) in wet season (Fig. S17).

4. Discussion

4.1. Microplastics in dredge sediments

Microplastic pollution is commonly found in river sediments worldwide. In this study, sediments dredged from the urban/suburban river system contained high concentrations of MPs (6060−37,610 items kg^{-1}), which are among the highest MP concentrations reported in freshwater sediments worldwide (Table S2). The total load of MPs contained in dredge sediments was about 6.46×10^{11} and 5.50×10^{10} in P1 and P2, respectively. Assuming the dredge sediments in P1 and P2 are representative of urban/suburban river sediments in Zhejiang Province, there was ~4.77×10^{15} MPs in the 2.52×10^{8} m^3 of dredged sediments removed from Zhejiang Province rivers and transferred to farmland storage piles from 2016 to 2017. MP concentrations in the dredge sediments of the urban river (P1) were significantly higher (>2-fold) than those of the suburban river (P2). This is consistent with other studies examining MP pollution in river sediment that report large differences in abundance and types of MPs across study areas (Peng et al., 2018; Mani et al., 2019). The high variability may result, in part, from localized inputs of sewage treatment effluents that are often the main source of MPs released to rivers in developed countries and discharge a predominance of fibers (Mahon et al., 2017; Lares et al., 2018). However, inefficiencies in solid waste disposal and sewage treatment in developing countries may result in large quantities of plastic debris released directly into rivers, and contributing a wider range of morphological types, such as fragments (Wong et al., 2020) or pellets (Peng et al., 2018). Fragments contributed more than 50% of total MPs in the dredge sediments in this study, which is very different from the typical MP composition (mainly fibers) released from wastewater treatment plants in China (Li et al., 2018). This implies that there are other appreciable sources of MPs besides sewage in riverine sediments from our study area.

The typical primary MPs, pellets, constituted only 13.6 ± 2.0% and 11.2 ± 3.2% of total MPs in dredge sediments from urban and suburban rivers, respectively, while the other MPs recovered from the dredge sediments were mainly composed of secondary MPs (e.g., fragments formed from degradation of macroplastics). Solar UV radiation initiates autocatalytic thermal oxidation, which is the primary mechanism for environmental degradation of plastic polymers (Andrady, 2017). However, for the dark and anaerobic environment characterizing riverine sediments, the photo-, chemical- and biodegradation pathways for plastic breakdown are greatly diminished (Tansel, 2019). Therefore, the abundance of secondary MPs in the dredge sediments is unlikely the result of in situ degradation of large plastic materials, but rather transport into the river system by runoff and wind after weathering on land. The secondary MPs in riverine sediments may also formed within the river channel by abrasion of plastic trashes during strong hydrologic events.

An unfavorable environment for plastic degradation also occurs within the dredge sediment storage piles, similar to plastic disposal in solid-waste landfills. Due to extremely slow rates for transformation processes of waste materials (including plastics), plastics with high persistence can remain intact for several centuries in landfills (He et al., 2019). Although plastics in riverine sediments degrade slowly, they may be released back into the waterbody upon dredging. The weathering velocity of plastics will accelerate substantially when they encounter an aerobic and irradiant environment during resuspension in surface waters, or transfer to the land surface. The plastics within dredge sediments

remain 'quiescent', but the plastics on the surface of the sediment piles are exposed to more active weathering conditions (e.g., physical, chemical and biological) that may generate an abundance of smaller MP breakdown products. However, MP concentrations in surface samples were not higher than interior samples in the dry season and even lower in the wet season at P1. Thus, it is necessary to distinguish MP production (formation) and transport processes (removal) to fully understand MP dynamics at the atmosphere-sediment pile interface.

4.2. Microplastics in the environment surrounding dredge sediment piles

As MPs are common in urban runoff and landfill leachates (Praagh et al., 2018; He et al., 2019), washing by rain or wind may be responsible for the loss of MPs from the surface sediment of the piles in this study. The MP concentration in ditch sediments near the piles was higher than the reference ditches in most cases, suggesting that MPs were released from the field containing the sediment piles. However, the MP concentrations in the farmland soil surrounding the sediment piles were not higher than the reference soils in most cases. This may result from interception of the MPs washed/eroded from the piles by ditches surrounding the piles. Most of the MPs washed from the sediment piles by rainfall will be effectively collected by the ditches where they are subsequently transported to the river. In contrast, MP concentration in the soil surface around P1 (>3000 n/kg·DW) was higher than that in reference surface soil (1000–2000 n/kg·DW) in the dry season. The dry season enrichment of MPs in the soils surrounding the pile maybe attributed to wind erosion of MPs from the sediment piles and deposition within the surrounding farmland soils.

Both the soil surrounding the sediment piles (780−4180 n/kg·DW) and the reference farmland soils (810−2340 n/kg·DW) located 200–300 m from that sediment piles had higher MP concentrations than suburban farmland soils of Shanghai (<100 n/kg·DW) (Liu et al., 2018), but lower than that in southwestern China (18,760 n/kg·DW) (Zhang and Liu, 2018). The MPs in farmland soils may originate from soil amendments (e.g., compost, sewage sludge, or residues of plastic mulching), input with irrigation water, flooding, and atmospheric deposition (Bläsing and Amelung, 2018). Previous studies reported only minimal degradation of synthetic plastic polymers in the soil environment, allowing PE fragments from plastic mulching to persist for several years to decades in soil, while physically breaking down to form smaller plastic residues (Krueger et al., 2015). Recent studies revealed significant changes in plant biomass, tissue elemental composition, root traits and soil microbial activities in soils polluted by MPs, implying that MP contamination in soil may affect plant and agroecosystem performance (Boots et al., 2019; Machado et al., 2019). MPs also have negative impacts on the reproduction, growth and mortality of animals like soil dwelling earthworms and nematodes (Huerta Lwanga et al., 2016). Furthermore, MPs can act as a carrier for other pollutants, thereby introducing toxic compounds into the food web (Bläsing and Amelung, 2018).

Although the existence of ditches lead to a moderate diffusion of MPs from the dredge sediment piles to the surrounding farmland, they were a main pathway for the transport of MPs to the river system. MP concentrations in the ditch sediments around P1 were much higher than that in the reference ditches, and MP concentrations increased to the same level as in the dredge sediment pile in the wet season experiencing strong rainfall/erosion. The proportion of small MPs in the P1 ditch increased in the wet season, and was higher than that in the reference ditches. We interpret these results to indicate greater MP transport from the sediment piles to ditches during the wet season. The proportion of fragments in ditch sediment MPs was higher than found in the sediment piles, implying that fragments are selectively deposited in the ditch compared to the other morphological MP forms. These findings are consistent with a study of agricultural soils near Hangzhou Bay where MP types in soils were dominated by fragments, while fibers were the dominant type in waters and fragments the least abundant (Zhou et al., 2020). This suggests that different MP morphologies may influence their fate in ditches, such as fibers preferentially transported by water flow, while fragments tend to deposit in the sediments. As the core of farmland irrigation systems, ditches are the primary pathway for material transport in and out of farmland systems. Given the height of sediment piles relative to the ditches, MPs from the sediment piles will be continuously released to ditches as rain-induced erosion removes the surface sediments. While, ditches are thought to mediate the flow of pollutants from agroecosystems to downstream water bodies (Needeman et al., 2007), heavy rainfall/runoff events can readily remobilize the MPs deposited in the ditches and transport them to downstream rivers (Hurley et al., 2018).

Seasonal difference of MP concentration in river waters near the dredge sediment piles infer that rainfall plays an important role in regulating the release of MPs from the sediment piles to nearby rivers. During the dry season, the lack of rainfall prevents the release of MPs from the sediment piles to the river. Additionally, the low dry season flows of plain river networks increase the hydraulic retention time in the dry season, which allows for longer biological fouling or hetero-aggregation with suspended sediments that in turn contributes to enhanced sedimentation of suspended MPs from surface waters (Chen et al., 2019; Li et al., 2019). While there was no difference in MP concentrations between upstream and downstream waters during the dry season, higher MP concentrations occurred below the ditch inputs in downstream waters during the wet season. Thus, the area surrounding the sediment piles becomes a point source for MP pollution during the wet season. Hence, river sediments act as an overall sink for MPs originating from the watershed; however, some of these MPs are remobilized following river dredging when they are released back into the rivers from the sediment storage piles. As such, the MPs remain a threat to freshwater and marine ecosystems.

4.3. Impact of microplastic pollution from dredge sediments

The present study shows, as a concession to treatment costs, long-term piling of dredge sediments on farmland has an obvious environmental risk for MP release back into terrestrial and aquatic environments. Although ditches reduce the impact of MPs washed from the surface of the dredge sediment piles to the surrounding farmland soil, wind transport during the dry season may still contaminant the adjacent farmland with MPs. Most of the MPs released from sediment piles returned to the rivers through the ditch drainage system, which creates the potential for negative effects on aquatic organisms (Hu et al., 2018; Kokalj et al., 2018). The river water is also used as a source of agricultural irrigation water; thereby the release of MPs from dredged sediment piles to rivers may increase the ecological risk for farmlands along the rivers. Similar MP dispersion could result from other outdoor dumping of wastes, like sludge or trash, making these sites long lasting point sources for MP pollution.

Moreover, piling of dredge sediments on land is not the final destination for these sediments, but only a temporary step in the dredging process. Similar to other organic wastes, like compost and sewage sludge, the sediments of rivers, lakes and ponds are rich in plant nutrients and organic carbon. Hence they are widely used as a soil amendment to improve soil physico-chemical properties, nutrient content and increase yields (Urbaniak et al., 2017). Thus, while compost may provide a beneficial amendment in agriculture, it may also serve as a medium for MP additions to soil (Bläsing and Amelung, 2018). Sewage sludge is known to contain several pollutants evoking regulations that include limits for pathogen and heavy metal contents. However, no current regulations exist for plastic as a potential, unwanted ingredient. Most MPs in sewage are removed by wastewater treatment and remain in the sludge (Carr et al., 2016; Mahon et al., 2017), which makes land application of sewage sludge an important pathway for dispersion of MP pollution (Nizzetto et al., 2016; Corradini et al., 2019; van den Berg et al., 2020). In our study area, if heavy metal concentrations in dredge

sediments are below the regulatory threshold, local governments tend to use the material as an agricultural fertilizer. When heavy metal concentrations in dredge sediments exceed regulatory thresholds, it is often used as an amendment for landscape plantings. Both applications will transfer MPs from dredge sediments to the soil environment where it may cause potential ecological risk.

Another destination for dredge sediments is coastal waters. Following dewatering of dredge sediments during storage in piles, it can be used as fill material for coastal land reclamation. However, the loose structure of dredge sediments is not preferred by reclamation projects, greatly limiting its use. Finally, if no feasible options are identified for dredge sediment disposal, the materials are disposed into the coastal sea. If the 7 million m^3 of dredge sediments from Yueqing City during 2016–2017 are finally dumped into the adjacent Yueqing Bay (469 km^2), the release of MPs will result in an average concentration of more than 280,000 MPs/m^2 in the surface water of this bay. If the dispersion impact is extended to the adjacent continental shelf of China, the disposal of 250 million m^3 of riverine sediments dredged in Zhejiang Province will result in about 18,000 MPs/m^2 in the sea area more than 600 km away from the coastline. Dumping of dredged sediments may further increase turbidity, decrease dissolved oxygen, and release nutrients (contributing to eutrophication/hypoxia/harmful algal blooms) and pollutants in coastal waters (Essink, 1999), hence causing several negative impacts on marine ecosystems (Erftemeijer and Robin Lewis, 2006). MPs, and associated toxic compounds, released from ocean dumping of dredge sediments maybe incorporated into the marine ecosystem through food web dynamics, thereby introducing risks to human health through consumption of seafood (Rochman et al., 2015).

5. Conclusion

Dredging is known to have deleterious impacts on aquatic habitats and ecosystems, but the environmental effects of dredging on MP pollutants has not been previously assessed. This study identified that MPs from river sediments were transported to dredge sediment storage piles, where they were subsequently weathered and dispersed to surrounding soils and surface waters while awaiting a permanent disposal option. Hence, MPs accumulated in dredge sediments can be remobilized causing serious and widespread environmental issues. Whether the dredge sediments are finally used to fertilize farmland, as fill material for coastal land reclamation or dumped into the ocean, MPs have the potential for remobilization into the environment causing potential issue with human health, aquatic foodwebs and agricultural production. Therefore, dredging programs should devote more attention to safe disposal options for all potential pollutants, including microplastics.

CRediT authorship contribution statement

Xiaoliang Ji: Investigation, Writing - original draft. **Yuan Ma:** Methodology. **Ganning Zeng:** Investigation. **Xiaoqun Xu:** Investigation. **Kun Mei:** Visualization. **Zhenfeng Wang:** Formal analysis. **Zheng Chen:** Methodology. **Randy Dahlgren:** Writing - review & editing. **Minghua Zhang:** Writing - review & editing. **Xu Shang:** Conceptualization, Writing - original draft.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2020.124132.

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