Metal release/accumulation during the decomposition of Potamogeton crispus in a shallow macrophytic lake

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ABSTRACT

Changes in metal concentrations in the litter of Potamogeton crispus were monitored during a consecutive 40-day in situ decomposition experiment using the litterbag method. The accumulation index was calculated and used to indicate the changes in the metals in litter. The results showed that the concentrations of Al, Cd, Cr, Mn, and Pb in litter increased significantly during the decomposition, while Cu and Zn concentrations decreased dramatically. Significant positive correlations were found between the concentrations of Al, Cr, Fe, and Mn and between Cu and Zn. Moreover, Cu and Zn both negatively correlated with Al and Fe. The remaining dry mass was negatively correlated with Al and Fe concentrations but positively correlated with Cu and Zn concentrations. Generally the accumulation index values of metals other than Al were less than one, indicating that the litter of P. crispus acted as a source of metals to the surrounding water body. Al was the only metal that showed continuous net accumulation in litter. The net accumulation of Fe and Mn in litter during the last 10 days of the experiment may indicate the precipitation of Fe- and Mn-oxides. It was estimated that 160 g/m² (dry weight) P. crispus was decomposed in 40 days. This was equivalent to releasing the following amounts of metals: 0.01 mg Cd, 0.03 mg Cr, 0.71 mg Cu, 0.55 mg Mn, 0.02 mg Pb and 13.8 mg Zn into surrounding water, and accumulating 149 mg Al and 11 mg Fe, in a 1 m² area.

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Introduction

Aquatic plants play an important role in the structure and function of aquatic ecosystems. Their status is a direct indicator of environmental conditions, because they grow in close contact both with the water that surrounds them and the sediments in which they are rooted (Brönmark and Hansson, 1998). Several studies have proposed that submerged plants in the growing period are natural sinks for metals, as they can take up and adsorb large quantities of metals from sediments and water (Cardwell et al., 2002; Weis and Weis, 2004). However, when aquatic plants die, the decomposing plant tissues may be a source of elements released through leaching and mineralization, or sink through litter adsorption or microbial immobilization (Schaller et al., 2011; Eid et al., 2012). Meanwhile, the decomposition of aquatic plants is critical for eutrophic shallow lakes, because it can influence the sediment layer and thus the terrestrial forming processes (Chen et al., 2013).

A number of studies have focused on the metal accumulation in decaying litter of wetland plants over time by using...
Dongping relies mainly on surface runoff with an average of 0.58 m (Chen et al., 2013). Recharge to Lake green, and the water transparency ranges from 0.20 to 1.46 m water all year round, and its multi-annual average of water with an annual precipitation of 640 mm and an annual average continental monsoon climate and has four distinct seasons, of the Yellow River and the Dawen River. In addition, Lake the Yellow River, its main role is to regulate and store floods center for the Dawen River drainage and a retention reservoir of

Dongping County, southwest of Shandong Province, China. With a total area of 627 km² including the old and the new lake basins, Dongping experiences a warm and semi-humid continental monsoon climate and has four distinct seasons, with an annual precipitation of 640 mm and an annual average temperature of 13.3°C, and the average monthly water temperature varies between 4°C (January) and 30°C (August) (Chorography Compilation Committee of Dongping County in Shandong Province, 2006). Metal contents in surface sediment were 53,498 ± 3889 mg/kg for Al, 1.00 ± 0.13 mg/kg for Cd, 79 ± 10 mg/kg for Cr, 43 ± 13 mg/kg for Cu, 30,045 ± 2579 mg/kg for Fe, 666 ± 110 mg/kg for Mn, 22 ± 4 mg/kg for Pb, and 100 ± 13 mg/kg for Zn, respectively (mean ± standard deviation, n = 47, expressed on dry mass basis). Organic matter content in surface sediment varied from 5.2 to 52.1 g/kg with an average of 22.3 g/kg. The mean particle size of surface sediment was 25.8 μm. Lake Dongping has an abundance of vegetation types, such as Trapa, Nymphaea, Typha, Potamogeton, Lemna, and Cyperus, etc. (Chen et al., 2013). P. crispus is the dominant species of the aquatic vascular plants in Lake Dongping. The growth of P. crispus shows great seasonal variations. In Lake Dongping, P. crispus forms shoots in the autumn (September–November) and grows in the winter and spring (December–May). Generally, biomass reaches a peak in May when more than 80% of the lake area is intensively colonized by P. crispus, and the average biomass is about 3.27 kg/m² (wet weight), with a maximum biomass that can reach 5.33 kg/m² (wet weight) (Zhang et al., 2009). In late May and early July (early summer), P. crispus begins to die and decompose, which causes the deterioration of the aquatic ecological environment and the massive death of fishes. The pH of water in Lake Dongping during the decomposition of P. crispus was between 7.17 and 8.92, and the dissolved oxygen (DO) concentrations ranged from 2.58 to 9.03 mg/L, and the range of chemical oxygen demand (COD) was from 4.97 to 11.5 mg/L (Zhang et al., 2009).

1.2. Sampling methods and elemental analysis

Decomposition of P. crispus was investigated with the litter bag technique, which is widely used in research (e.g. Zawisianski et al., 2001; Longhi et al., 2008; Balasubramaniam et al., 2012). At the end of May, 10 kg of P. crispus (aboveground part) in Lake Dongping was collected just prior to its death. Fresh plant was transported to the laboratory and washed with water and then with deionized water. Cleaned P. crispus was dried at 60°C to constant mass (Longhi et al., 2008), and 5 g of the dried plant detritus was enclosed in each 0.15 mm mesh size nylon bag (20 cm × 20 cm). In total, 45 litter bags were prepared and strung together with a nylon rope for convenience of sampling, and then the bags were put back into Lake Dongping together with the natural P. crispus detritus.

Three bags (triplicate samples) were taken out on the 1, 2, 4, 6, 8, 10, 12, 14, 16, 18, 20, 25, 30, 35, and 40th days after bags were set in the lake, and then transported into the laboratory for analysis. In the laboratory, the residual plant litter in bags was carefully cleaned and repeatedly rinsed with deionized water to remove adhering debris and sediment particles. The cleaned sample in each bag was dried at 60°C again to constant mass (Longhi et al., 2008), then weighed and homogenized. P. crispus and the litter samples were analyzed for metal content. A 0.5 g sample was hot-digested with HNO₃–HClO₄–HF (GR) and then diluted to 50 mL volume with 1:1 (V/V) HNO₃ (Deng et al., 2010). Concentrations of Al, Cd, Cr, Cu, Fe, Mn, Pb, and Zn, were measured by a Varian 710ES ICP-OES spectrometer (Varian Inc., USA). Concentrations were
calculated based on the dry weight of each sample. The analytical accuracy was estimated by the analysis of Certified Reference Materials—Poplar leaves (GSV-3, National Research Center for Certified Reference Materials, China). The recoveries were between 90% and 110%. In order to examine any possible contamination during the analytical procedure, blanks were analyzed synchronously in each analytical batch and always accounted for less than 2% of metal concentrations. The containers used in the analytical procedure were cleaned by immersing in 10% (V/V) HNO₃ for more than 24 h; subsequently, they were washed with fresh water, followed by deionized water to ensure that they were free of contamination.

1.3. Data analysis

The accumulation index (AI) is calculated to indicate the net release or accumulation of metals during the decomposition of *P. crispus* (Romero et al., 2005):

\[
AI = \frac{W_tX_t}{W_0X_0}
\]

where \(W_t\) (g) is the dry mass of litter at time \(t\), \(X_t\) (mg/kg) is the metal \(X\) concentration in litter at time \(t\), \(W_0\) (g) is the initial dry weight of litter, and \(X_0\) (mg/kg) is the initial concentration of metal \(X\) in litter. An AI value of 1 indicates that the decomposed litter at time \(t\) contained the same mass of the metal \(X\) as when the litter of *P. crispus* was placed in the litterbag; AI < 1 indicates a net release of metal \(X\) from the decaying litter of *P. crispus*; while AI > 1 indicates a net accumulation of metal \(X\) by the decaying litter.

2. Results and discussion

2.1. Litter mass loss

The mass loss of *P. crispus* during the 40-day decomposition is presented in Table 1. Results indicated that *P. crispus* decomposed rapidly in the first 14 days, especially in the first 4 days when more than half of the dry mass was lost. The classic three-phase model of organic matter decomposition was discerned from our results (Zawislanski et al., 2001; Pereira et al., 2007; Zhang et al., 2013). As shown in Table 1, in the first 2 days, the leaching of soluble materials led to the rapid loss (an average of 41.5%) of biomass of *P. crispus*. Then from the 2nd to the 14th day, the biomass continued to decrease due to the microbial degradation of labile substrates (e.g., sugars, starches and proteins), and the average of weight loss was 43.7%. From the 14th to the 40th day, as the refractory materials (e.g., cellulose, waxes, tannins and lignin) accumulated in litter, the decomposition rate decreased gradually. The litter weight even increased a little in this stage (Table 1), which might be caused by the adsorption by litter and by microbial immobilization (Pereira et al., 2007; Longhi et al., 2008). After 40 days, only 21.4% of the dry mass remained.

2.2. Metal concentrations in decomposing litter

The initial concentrations of metals were 84.3 ± 7.5 mg/kg for Al, 0.05 ± 0.01 mg/kg for Cd, 0.30 ± 0.08 mg/kg for Cr, 4.05 ± 0.75 mg/kg for Cu, 64.1 ± 6.8 mg/kg for Fe, 20.5 ± 4.6 mg/kg for Mn, 0.19 ± 0.04 mg/kg for Pb, and 73.7 ± 9.8 mg/kg for Zn (mean ± standard deviation, \(n = 3\)). The changes in metal concentrations in *P. crispus* litter are illustrated in Fig. 1. During the decomposition, metal concentrations in litter exhibited different temporal variations. Concentrations of Al, Cd, Cr, Fe, Mn, and Pb showed significant increases (\(p < 0.05\)), while concentrations of Zn and Cu decreased significantly (\(p < 0.05\)). During decomposition, the proportions of metal concentrations in litter bags to the mean initial concentrations are 2.7–50.0 for Al, 1.0–2.7 for Cd, 0.5–2.4 for Cr, 0.4–15 for Cu, 1.7–9.3 for Fe, 0.5–6.5 for Mn, 0.8–3.9 for Pb, and 0.3–1.3 for Zn. The proportions of Al and Fe were all greater than one, which indicated that concentrations of Al and Fe in litter always increased during the decomposition. The maximum proportions, i.e., the highest concentrations in the litter during the decomposition, occurred at the 1st day for Cd, Cu, and Zn, and at the 10th day for Pb, while for Al, Cr, Fe, and Mn, they were found in the last 10 days of the experiment. At the end of the experiment, the proportions varied in the order Al (49.1) > Fe (9.3) > Mn (4.3) > Pb (2.5) > Cr (2.4) > Cd (1.5) > Cu (0.6) > Zn (0.3).

As shown in Fig. 1, concentrations of some metals showed similar changes, indicating that they may undergo similar biogeochemical cycling during the decomposition (Pereira et al., 2007). The correlations between metal concentrations and remaining dry mass are listed in Table 2. There were significant positive correlations between Al, Cr, Fe, and Mn, and between Cu and Zn. Nevertheless, Cu and Zn were both negatively correlated with Al (\(p < 0.01\)) and Fe (\(p < 0.05\)). No significant correlations were found for Pb and Cd with the other six metals.

Changes in metal concentrations in plant litter were also observed by other researchers. Zawislanski et al. (2001) found decaying litter of *Spartina foliosa* underwent a very rapid increase in all metal species during the first few weeks of decomposition followed by a subsequent slower increase. Windham et al. (2004) found large increases (10–100 fold) in metal concentrations in the litter of *P. australis* and *S. alterniflora* in the field. Du Laing et al. (2006) found increasing Cd, Cr, Cu, Ni, Pb, and Zn concentrations in stems, leaf sheaths and leaf blades.
of *P. australis* during the decomposition in a brackish tidal marsh. In this study, concentrations of Al, Fe, Mn, and Cr increased significantly as the decomposition proceeded (Fig. 1). During the decomposition, microbes as well as their exudates, especially the exopolysaccharides (EPS), form a heterotrophic biofilm that can accumulate high amounts of metals (Schaller et al., 2010, 2011). Furthermore, Al, Fe, Mn, and Cr oxides might be bound by humic substances or precipitate on the surface and interstices of the litter during the decomposition (Pereira et al., 2007). This process may be further confirmed by the significant negative correlation between Al and Fe with remaining dry mass (Table 2). Therefore, the increase of Al, Fe, Mn, and Cr concentrations may be attributed to the combination of biofilms that grow on the decomposing litter and adsorption or precipitation of metals on the decomposing litter. The variation of Mn concentrations in the first 14 days is small compared with Al, Fe, and Cr. Organic matter oxidation may have led to the use of Fe- and Mn-oxides as electron acceptors,
and the reduced Fe and Mn forms may have leached from the litter (Pereira et al., 2007). As the oxidation of organic matter slowed down after 14 days, Mn concentrations in litter increased gradually, which could be interpreted as the precipitation of Mn-oxides. The microbes in the biofilm also facilitate the precipitation of manganese (Ohnuki et al., 2008; Schaller et al., 2010). The behavior of Al and Cr are linked to both Fe and Mn redox cycling and may be bound to the oxides and humic substances (Tipping et al., 2002; Hamilton-Taylor et al., 2005).

It is well known that Cu and Zn have similar geochemical behaviors, and significant correlations are generally found between Cu and Zn concentrations in soil, sediment, and street dust (Shi et al., 2008). During the decomposition of P. crispus, they were also well correlated with each other (p < 0.01) and showed a similar temporal variation pattern opposite to those of Al, Fe, Mn, and Cr. The highest concentrations of Cu and Zn in litter were found on the 1st day. This might be related to the rapid leaching of soluble materials in P. crispus, which resulted in an increase in the concentrations of Cu and Zn in litter. Along with the litter decomposition, concentrations of Cu and Zn in litter decreased from the 2nd to the 14th day. Cu and Zn as micro nutrients for microbial growth are utilized by microorganisms during the litter decomposition; on the other hand, Cu and Zn were linked to their leaching mechanism and the surrounding water body (Dong et al., 2000, 2003; Hamilton-Taylor et al., 2005), and they could be easily released following the microbial degradation of labile substrates in P. crispus. The significant positive correlations between Cu and Zn with the residual ratio of dry mass also suggest that Cu and Zn were released with the litter decomposition. After 14 days, the decrease of Cu and Zn concentrations slowed down, especially for Cu, for which concentrations were almost constant from the 16th day to the 40th day. This might be correlated to the slow decomposition of the refractory materials in P. crispus, adsorption by litter, and microbial immobilization (Du Laing et al., 2006; Pereira et al., 2007).

Cd concentrations in litter fluctuated dramatically in the first 14 days, and then increased gradually till the end of the experiment, while Pb concentrations showed a great fluctuation in the first 20 days, followed by an increase from the 20th day to the 30th day, and then remained relatively constant in the last 10 days of the experiment. On the one hand, the fluctuation of Cd and Pb concentrations might be influenced by the first two stages of litter decomposition. For example, the fact that the highest concentration of Cd was on the 1st day might be due to the fast leaching of the soluble materials in P. crispus. On the other hand, Cd and Pb in the litter were more likely affected by the surrounding water body, such as from suspended particles entering the litterbags, due to the low initial concentrations of Cd (0.05 ± 0.01 mg/kg) and Pb (0.19 ± 0.04 mg/kg) in P. crispus. This may also be the reason why Cd and Pb did not have significant positive correlations with Fe and Mn (Table 2) although they could be adsorbed by Fe- and Mn-oxides (Dong et al., 2000, 2003; Hamilton-Taylor et al., 2005). In general, Cd and Pb concentrations in litter tended to maintain a relatively constant level in the later stage of the experiment, indicating that an equilibrium between adsorption and desorption was attained in the litterbag.

Increasing metal concentrations do not mean that the litter bags functioned as a sink for metals, because the biomass of litter was lost at the same time (Pereira et al., 2007). Therefore, Al values were calculated according to Eq. (1), and the results are illustrated in Fig. 2. It is found that the Al values of Cr, Cu, and Zn were all less than one, suggesting the export of metals from litter to the surrounding water body during the 40-day decomposition. The same results were found with Cd, Pb, and Mn, except for the Al values of Cd on the 1st day, Pb on the 2nd day, and Mn on the 35th day. The Al values of Al were all more than one, with a range between 1.55 and 10.88, indicating the great accumulation of Al in litter. As for Fe, its Al values fluctuated around 1 till the 25th day, then increased by 137% to 1.97 at the 30th day, and remained relatively constant in the last 10 days.

Al values reflect whether the litter was a net source or sink of metals during the decomposition of P. crispus. Al values showed the export of Cd, Cr, Cu, Pb, Mn, and Zn from litter to the surrounding water body during the 40-day experiment. However, the variation of Al values between sampling times meant that the export was not uniform, especially for Cr, Pb, and Mn. These results are in accord with the findings of Pereira et al. (2007), who also reported that metal release exceeded sorption as the decomposition proceeded. Moreover, Windham et al. (2004) found no net metal accumulation due to the rapid mass loss, although concentrations of metals in litter samples of P. australis and S. alterniflora. These results indicated that the metal stocks might not increase with the enrichment of metals in decomposing litter compared with the initial metal content, and they might even decrease due to the more rapid mass loss.

In order to investigate the influence of the biomass loss on the metal pools of the litter, the residual ratios of litter are also depicted in Fig. 2. It could be inferred from Fig. 2 that the loss of metals in litter was not synchronous with the dry mass loss. The AI values of Cu and Zn, except for the 1st day, were generally smaller than the residual ratio, which might cause the decrease of Cu and Zn concentrations in the decomposing litter due to the more rapid loss of dry biomass (Fig. 1). The AI values of Cu and Zn on the 1st day are much higher than

<table>
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<th>Metal</th>
<th>AI Cr</th>
<th>AI Cu</th>
<th>AI Fe</th>
<th>AI Mn</th>
<th>AI Pb</th>
<th>AI Zn</th>
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<td>0.37a</td>
<td>0.52a</td>
<td>0.54b</td>
<td>0.54b</td>
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<tr>
<td>Cd</td>
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<td>0.78a</td>
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<tr>
<td>Cr</td>
<td>0.68a</td>
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</tr>
<tr>
<td>Cu</td>
<td>0.52b</td>
<td>0.52b</td>
<td>0.72a</td>
<td>0.72a</td>
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<tr>
<td>Fe</td>
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<td>0.62a</td>
<td>0.86a</td>
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<tr>
<td>Mn</td>
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<td>0.18a</td>
<td>0.19a</td>
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<tr>
<td>Pb</td>
<td>0.43a</td>
<td>0.43a</td>
<td>0.72a</td>
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<tr>
<td>Zn</td>
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<td>0.89a</td>
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Table 2 - Spearman correlations between metal concentrations in litter and remaining dry mass during the consecutive 40-day decomposition experiment of P. crispus.

* RM: remaining dry mass.

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**2.3. AI change of metals in decomposing litter**

Increasing metal concentrations do not mean that the litter bags functioned as a sink for metals, because the biomass of litter was lost at the same time (Pereira et al., 2007). Therefore, AI values were calculated according to Eq. (1), and the results are illustrated in Fig. 2. It is found that the AI values of Cr, Cu, and Zn were all less than one, suggesting the export of metals from litter to the surrounding water body during the 40-day decomposition. The same results were found with Cd, Pb, and Mn, except for the AI values of Cd on the 1st day, Pb on the 2nd day, and Mn on the 35th day. The AI values of Al were all more than one, with a range between 1.55 and 10.88, indicating the great accumulation of Al in litter. As for Fe, its AI values fluctuated around 1 till the 25th day, then increased by 137% to 1.97 at the 30th day, and remained relatively constant in the last 10 days.
the residual ratio, indicating that the fact that the highest concentrations of Cu and Zn occurred on the 1st day might be related to the rapid leaching of soluble materials, which caused Cu and Zn to be concentrated in litter. The AI values of Al, Cd, Cr, Pb, Fe, and Mn were generally higher than the residual ratio of dry mass, suggesting the more rapid loss of plant materials. Therefore, an increase of metal concentrations was observed during the decomposition (Fig. 1).

Whether metal is lost from litter or accumulated during the decomposition is determined by two major processes:

**Fig. 2** – Changes in the accumulation index (AI) of metals in litter during the consecutive 40-day decomposition experiment of *P. crispus*. (Error bars represent one standard deviation, n = 3).
(1) the release of metals from dead plant tissues as they oxidize; and (2) the passive sorption on organic surfaces of the litter and active accumulation by microbial colonizers (Windham et al., 2004; Du Laing et al., 2006; Schaller et al., 2010). The AI values of Cd and Pb are less than one, except on the 1st day for Cd and the 2nd day for Pb, indicating that Cd and Pb may be absorbed from the surrounding water body into litter at the beginning of the decomposition. Cu and Zn are micro-nutrients for microbial growth; therefore, the steady fall of AI values of Cu and Zn during the first 14 days might indicate the utilization of Cu and Zn by the microbes as they degraded the litter. As the refractory materials accumulated in litter (Table 1), Cu and Zn maintained nearly constant AI values till the end of the experiment. Fe, as a macro nutrient for microbial growth, appears to be more active than Mn, as the AI values of Fe in the first 25 days fluctuated around one, indicating that intense adsorption and desorption occurred in litter, while Mn was mainly released from litter. The increase of the AI values of Fe and Mn during the last 10 days of the experiment was probably due to microbial immobilization and the precipitation of Fe- and Mn-oxides (Sundby et al., 2003; Schaller et al., 2010). The minimum AI values of Cd, Cr, Cu, Fe, Mn, and Zn occurred on the 14th day, which is in accordance with the residual ratio change of the dry mass (Fig. 2). This indicated that metals were released to the maximum extent during the first 14 days. The AI values of Al above one indicated the continuous incorporation of Al into litter. Al is not a nutrient element for plants, and aluminum ions have a toxic impact on plant growth. The ratio of Al concentration in P. crispus to that in sediment is the smallest compared with those of the other metals. Furthermore, Al has significant interactions with natural organic matter (mainly humic substances) (Tipping et al., 2002). Therefore, during the decomposition of the litter, most humic substances may adsorb a large amount of Al from the surrounding water body, causing the increase of aluminum Al values.

According to our decomposition experiment, about 80% of the initial dry mass of P. crispus was lost after 40 days (Table 1). Lake Dongping could produce more than 250 × 10^4 kg/km^2 fresh P. crispus annually (Zhang et al., 2009), and the dry matter of P. crispus accounts for about 8% of the fresh weight. Thus, about 160 g/m^2 (dry weight) P. crispus was estimated to be decomposed in 40 days. Based on the metal concentrations in the initial litter and those at the 40th day, the decomposition of P. crispus in 40 days was equivalent to releasing the following amounts of metals into the surrounding water (in mg/m^2): 0.01 Cd, 0.03 Cr, 0.71 Cu, 0.55 Mn, 0.02 Pb, and 13.8 Zn; but accumulating 149 Al and 11 Fe in litter from the water body.

### 3. Conclusions

During the decomposition of P. crispus, the concentrations of Al, Cd, Cr, Fe, Mn, and Pb significantly increased in litter, while Cu and Zn showed the opposite trend. Significant positive correlations were found among Al, Cr, Fe, and Mn, and also between Cu and Zn; furthermore, Al and Fe were both negatively correlated with Cu and Zn. This indicates that Al, Cr, Fe, and Mn had similar biogeochemical cycling, which was different from that of Cu and Zn in litter during the decomposition. In addition, Al and Fe were negatively correlated with the remaining dry mass, and Cu and Zn were positively correlated with the remaining dry mass, which suggests the accumulation of Al and Fe and the release of Cu and Zn in litter. In terms of metal stocks in litter bags, the AI values of Cd, Cr, Cu, Pb, Zn, and Mn were less than one, indicating that the release of these metals exceeded the sorption during the decomposition.

The AI values of Al were all higher than one and increased with time in litter, suggesting that litter might adsorb the abundant Al in the surrounding water body. No significant net release or accumulation were found for Fe in the first 25 days, followed by a net accumulation of Fe in the last 10 days, which might indicate the precipitation of Fe- and Mn-oxides. In general, litter of P. crispus acted as a source of Cd, Cr, Cu, Mn, Pb, and Zn, but a sink of Al and Fe, as revealed by our decomposition experiment. Our results not only reveal the strong influence of P. crispus decomposition on metal cycling in lakes, but also give warning that the application of submerged macrophytes for phytoremediation of the metal contaminated water body should consider the secondary pollution caused by the decomposition of aquatic plants.

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