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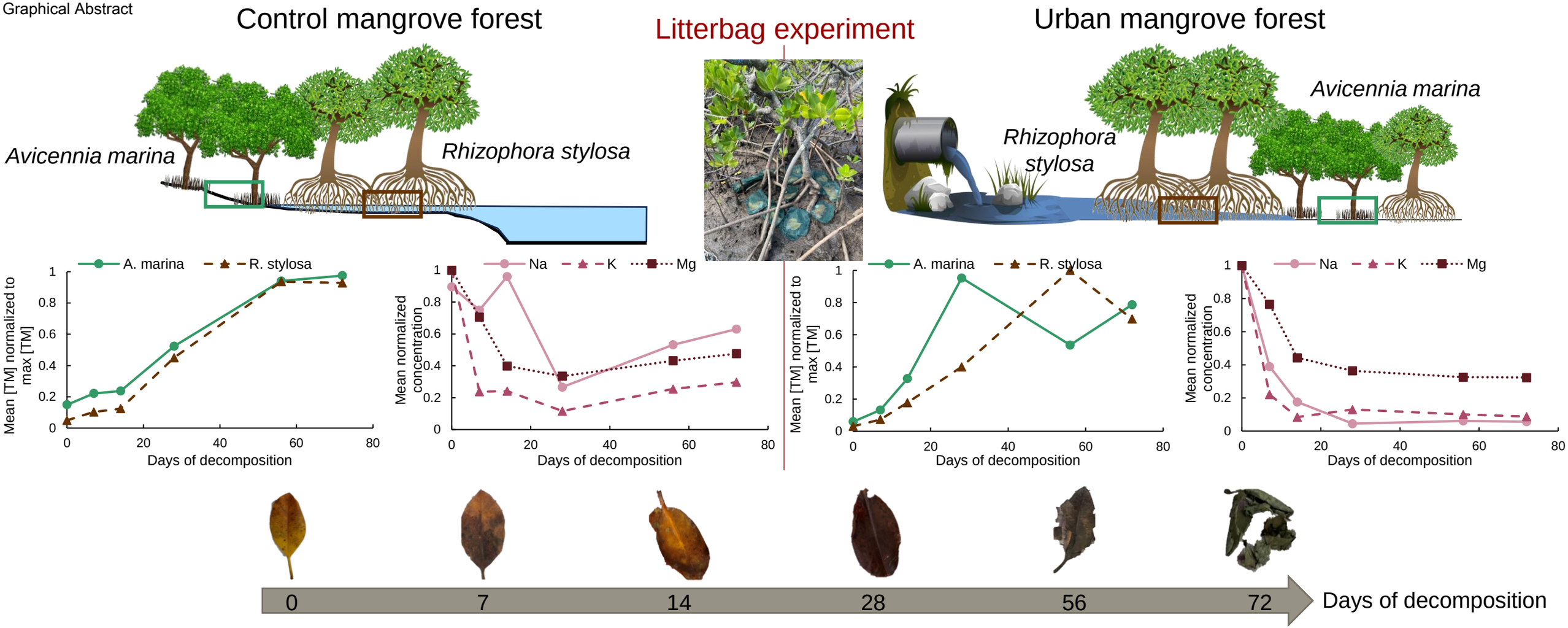
Graphical Abstract



Highlights

- Litterfall decomposition under the influence of urban runoff was investigated
- Stands position and species salt tolerance influence initial leaves composition
- Urban rainwater runoff enhances major elements leaching from litterfall
- Trace metal dynamics are influenced by their bioavailability in mangrove soil
- As and Mn are exceptions due to their complexations and oxidation states

Dynamics of major and trace elements during leaf litter decomposition in semi-arid mangrove subject to urban runoff

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Abstract

This study examined the dynamics of major elements and trace metals (TM) during litterfall decomposition in two mangrove forests—control and urban—along New Caledonia's coast. A litterbag experiment was carried out for 72 days for the two main species (*Rhizophora stylosa*, and *Avicennia marina*) of the island. Results showed that the urban runoff enhances the leaching of some major elements (K, Mg, Na) during litter decomposition. The enrichment of TM in litter is also faster at the urban site. The dynamics of some major elements and TM are rather influenced by species metabolisms or element's characteristics such as Mn which does not increase in concentration in the decomposing litter. This study concludes that urban runoff accelerates the leaching of major elements and the enrichment of TM during leaf litter decomposition in mangrove forests, with species-specific factors and environmental conditions also playing significant roles in influencing these dynamics.

Keywords: mangrove, urbanization, trace metals, litterfall, decomposition, New Caledonia

1. Introduction

The mangrove ecosystem develops within the intertidal zone of tropical and subtropical regions down to temperate regions in the South (Alongi, 2002). Despite constituting less than 0.5% of the global forest surface, mangrove forests provide many ecosystem services including carbon sequestration and contaminants filtration, including trace metals (TM) (Lee et al., 2014). TM are naturally present in limited concentrations in the environment but anthropogenic activities have

increased their concentrations in particular compartments, raising concerns due to their potential negative effects on human health and the environment (Bayen, 2012; Prasad et al., 2006).

In mangrove forests, litter production constitutes 31% of the overall net primary production within the forest (Bouillon et al., 2008). Litterfall decomposition actively participates in the energy and material flow in the forest and constitutes an energy source for coastal productivity(Lugo and Snedaker, 1974; Twilley et al., 1986). Mangrove leaves comprise the predominant fraction of litterfall (Day et al., 1987; Twilley et al., 1986), ranging between 40% and 90% (Mackey and Smail, 1996). Consequently, most investigations into litterfall decomposition center on leaf litter, employing the litterbag experiment methodology (Nordhaus et al., 2017; Vinh et al., 2020; Yang et al., 2018).Leaf litter degradation occurs in three main stages: 1) the initial leaching of soluble compounds, 2) the colonization by microorganisms (i.e., fungi, bacteria, and protozoa) and decomposition via litter breakdown (Cundell et al., 1979; Fell et al., 1975; Fell and Master, 1980), and 3) the slow refractory phase where the least labile molecules are consumed (Tam et al., 1990; Valiela et al., 1984; Wilson et al., 1986). Many factors can influence litterfall decomposition, including the initial composition of the leaves, the leaf consuming fauna and microbial communities on the floor, and frequency and degree of inundation (Fell and Master, 1980; Lugo and Snedaker, 1974; Mfilinge et al., 2002).

Litterfall organic matter (OM) can be consumed by autochthonous fauna and microorganisms on the forest floor, dissolved during decomposition, and subsequently be conveyed to adjacent ecosystems during ebb tides, either as debris or dissolved organic/inorganic carbon (Kida et al., 2019; Ray et al., 2018; Taillardat et al., 2018). Similarly, elements such as TM can be leached to the surroundings during leaf litter degradation, or absorbed by the litter consuming fauna (Fell and Master, 1980; Fourqurean and Schrlau, 2003; Gautam et al., 2016; Nordhaus et al., 2017). Elements found in their soluble form in mangrove leaves, such as most major elements, are easily leached during litter degradation (Bonanomi et al., 2010). On the other hand, the enrichment of major elements and TM in leaf litter during the degradation process is attributed to inputs from the water column (Lacerda et al., 1988; Rice and Windom, 1982; Tam et al., 1990; Vinh et al., 2020). In mangrove soils, TM precipitation/dissolution cycles are directly influenced by floor inundation and redox reactions (Noël et al., 2014), which greatly determine the transfers of TM between the

mangrove floor and the decomposing litter (Vinh et al., 2020). In literature, enrichment of TM in decomposing mangrove litterfall has been largely observed and is explained by the complexation of dissolved TM by organic molecules present in the litter (Scheid et al., 2009; Vinh et al., 2020; Zawislanski et al., 2001). To understand the dynamics of major elements and TM in leaf litter during decomposition is crucial to better understand the export of these elements to the adjacent ecosystems of their intake by litter-consuming fauna.

New Caledonia is one of the top four pacific territories with the most mangrove forests (Hamilton and Casey, 2016), as mangroves cover 80% of the west coast of the main island. The mangrove forests of this region, characterized by a semi-arid climate and a semidiurnal tidal pattern (Douillet, 2001), showcase simple biodiversity structures. Species monospecific stands prevail, shaped by topography and soil salinity (Baltzer, 1981; Marchand et al., 2011). *Rhizophora* spp., developing seaside, constitute over half of the mangrove flora, while *Avicennia marina*, developing at higher elevations and saltier soils, represent around 15% of the archipelago's mangrove vegetation (Deborde et al., 2015). Although the human population density is low in New Caledonia, the population living in urban areas has significantly increased this last decade, resulting in the development of urban infrastructures along the littoral zone, where mangrove forests develop (Insee, 2020). A recent study looked into the influence of coastal urbanization on these mangroves and found that part of the mangrove forest, receiving urban rainwater runoff for >50 years, was constantly submerged by runoff water, leading to the development of *R. stylosa* trees far taller than is typical for such semi-arid mangrove forests (Robin et al., 2022). Robin et al. (2022) showed that the rainwater runoff controls the physico-chemical parameters of the mangrove soil, reducing soil salinity and elevating pH levels. Furthermore, urban TM concentrations in the soil exceed those present in a “control” mangrove forest, implying greater TM mobility (Robin et al., 2022). A litterbag experiment conducted in the same mangrove forest revealed that the urban runoff favors leaf litter decay on the mangrove floor by promoting microbial development on the soil surface (Robin et al., 2024).

This study aimed to assess major element (K, Mg, Na, Ca, P) and TM (Al, As, Co, Cr, Cu, Fe, Mn, Ni, Pb, Ti, Zn) dynamics in leaf litter decomposition, examining the influence of urbanization. While major element and TM dynamics during litterfall decay have been explored in various studies

(Chale, 1993; Hossain et al., 2014; Ramos e Silva et al., 2006; Rice and Windom, 1982; Silva et al., 1998; Steinke and Ward, 1987; Tam et al., 1990; Van Der Valk and Attiwill, 1984; Vinh et al., 2020; Zawislanski et al., 2001), no investigation has looked into the effect of urbanization on such processes nor in semi-arid mangrove settings. In this study, two semi-arid mangrove forests—control and urban—along New Caledonia's west coast, were selected. Both forests share a common geological unit as the source of their watersheds, yet the urban forest has received urban rainwater runoff for over five decades. Employing a 72-day litterbag experiment, conducted on the main mangrove species (*Rhizophora stylosa* and *Avicennia marina*), we hypothesize that the half-lives of major elements usually leached early in the decomposition process such as K and Mg will be shorter at the urban site than the control site due to the constantly flooding runoff. We also hypothesize that greater TM enrichment in leaf litter will occur at the urban site due to their higher bioavailability in the soil at this site.

2. Materials and methods

2.1. Study sites

The city of Dumbea in New Caledonia has had a population increase of 33% this last decade, resulting in the development of urban infrastructures on the littoral zone (Insee, 2020). Two mangrove forests in Dumbea were selected as study sites (Fig. 1). These sites had the same volcano-sedimentary watershed and are dominated by the same two mangrove species (*R. stylosa and A. marina*). One site was located in the Apogoti Bay (22°12’08”S, 166°26’20”E). This site has no direct anthropogenic inputs (limited housing density) into the mangrove forest, and it was selected as a control mangrove forest. Another site is the urban mangrove forest (22°12’39”S, 166°27’19”E) exposed to rainwater runoff from the upper allotment (mainly housing lots for 10 000 inhabitants), which has been flowing into the mangrove forest for more than 50 years. At the control site, the two species develop in monospecific stands similar to most mangrove forests of the west coast, that is, *A. marina* landward and *R. stylosa* seaward. At the urban site, another *R. stylosa* stand develops more landward than the *A. marina* stand, at the entrance of the runoff, and its soil is always flooded with the urban rainwater.

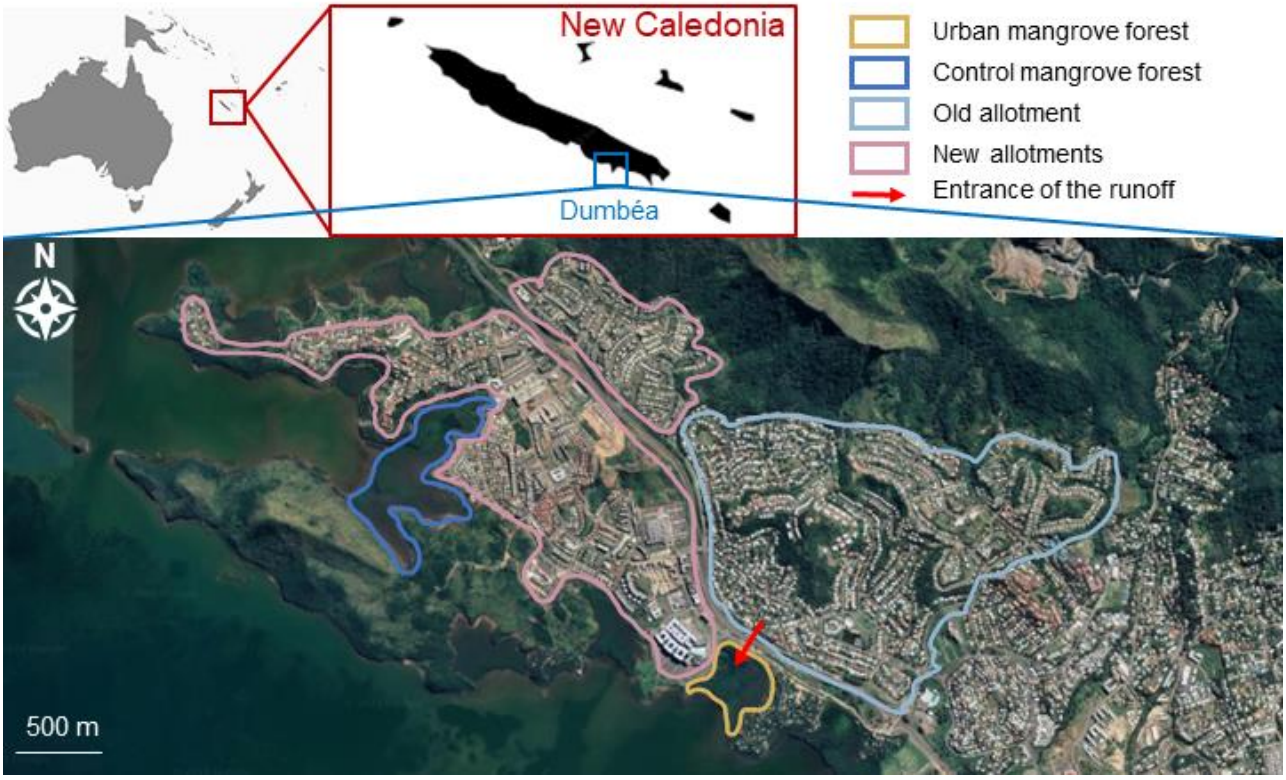

Figure 1. Control mangrove forest and urban mangrove forest in Dumbea city in New Caledonia with delimitation of the new and old allotments, and a red arrow to show the entrance and flow direction of the urban rainwater runoff (satellite image from Google Earth in 2022).

2.2. Litterbag experiment

Yellow senescent leaves about to fall from *A. marina* and *R. stylosa* trees were collected at the control site. The leaves were carefully washed with Milli-Q water and air-dried at the laboratory until constant mass. Three random leaf samples of 200 g per species were frozen, freeze-dried, and ground with a cutting mill, corresponding to the samples at t0 “senescent leaves”. In order to investigate the influence of urbanization on the dynamics of major elements and TM, we decided to use the leaves of the same trees (for each species) to place in the litterbags, that were afterward shared between the control and the urban site. This is to limit the effect of initial composition of the leaves on our results. We were able to do so as previous analyses showed no significant differences in TM concentrations in the leaves between the control and urban sites (Robin et al., 2022). A total of 90 samples of about 6 g of dried *A. marina* leaves and 90 samples of 15 g of dried *R. stylosa* leaves were weighed to the nearest 0.1 g and placed into nylon litterbags (2 mm mesh size). In each mangrove forest and for each species, three trees about 30 m apart were selected. A total of 15 bags of the corresponding species were attached to the roots of each of these three trees, at the

soil surface. Triplicate litterbags were collected at each of these three trees, for each species, and for each site after 7, 14, 28, 56, and 72 days (36 litterbags per day of collection). The experiment was stopped after 72 days because the decomposition was already very advanced, and samples further decomposed would be too hard to analyze properly. The litterbags were immediately opened at the laboratory; the leaves were carefully washed with Milli-Q water and air-dried until constant mass prior to weighing. Samples were then frozen, freeze-dried, and ground with a cutting mill. For initial comparison, fresh green leaves were also collected at both sites and for both species in triplicate, washed, freeze-dried, and ground.

2.3. Major element and trace metal quantification

The elements were measured in the dried ground leaves by ICP-OES after multiwave extraction. Briefly, 200 mg of each fresh, senescent, and degraded sample was weighed in a Teflon vessel. A volume of 5 ml of 70% HNO_3 (Ajax Finechem) and 1 ml of 30% H_2O_2 (Carlo Erba) were added to the vessel. The digestion program started with a 10 min heating step from room temperature to 160 °C, then a 15 min step to 210 °C, followed by a 10 min plateau and a 20 min cooldown step. The result of the extraction was transferred to a polypropylene tube. The volume was adjusted to 13 ml with Milli-Q water and placed at 4 °C until ICP-OES (Varian 730-ES) analysis at the chemistry laboratory LAMA of the French Research Institute for Sustainable Development in New Caledonia. For quality control, certified material (IPE sample ID 949) was also extracted and analyzed (Supplementary Table 1). The concentrations of major elements and TM were obtained using a calibration curve prepared from a stock solution (CPAChem) of the measured elements at 100 mg L^{-1}.

2.4. Data analyses

2.4.1. Major elements

To plot the changes in major element concentrations in the litterfall with degradation, the measured element concentrations in µg g^{-1} of dry sample were converted to percentage relative to t0. The values were calculated as follows:

$$\text{Element concentration at } t_n\ (\%) = ([\text{element}]_n * 100) / [\text{element}]_0 \quad (1)$$

with $[element]_n$ the concentration of the element at t_n in µg g^{-1} and $[element]_0$ the concentration of the element at t_0 in µg g^{-1}. For comparison with literature, single exponential trends were plotted when possible:

$$W_t = W_0 e^{-kt} \quad (2)$$

with W_t the element percentage at t, W_0 the initial element percentage (100 %), k the exponential factor, and t the time in days. The half-lives ($t_{1/2}$) in days were obtained with the following equation:

$$t_{1/2} = \ln(2) / k \quad (3)$$

2.4.2. Trace metals

To plot the changes in TM concentrations in the litterfall with degradation the measured TM maximum concentration in µg g^{-1} was set at 1.00 and the other values were calculated as follows:

$$\text{TM concentration at } t_n = [TM]_n / [TM]_{max} \quad (4)$$

with $[TM]_n$ the concentration of the TM at t_n in µg g^{-1} and $[TM]_{max}$ the maximum concentration of the TM between t0 and t72 in µg g^{-1}.

3. Results

3.1. Characterization of the fresh and senescent leaves

Prior to comparing major elements and TM in the litterfall during decomposition, it is necessary to look at the differences in the chemical composition of leaves between species and between fresh and senescent leaves. The concentrations of the measured elements in fresh and senescent leaves of *A. marina* and *R. stylosa* are displayed in table 1. In all samples and of all TM, Co had the lowest concentration varying from 0.02 to 0.18 µg g^{-1}, while Mn and Fe had the highest concentrations with values up to 238 µg g^{-1} for Fe and 321 µg g^{-1} for Mn.

At the control site, Al, Cr, Mn, and Pb had concentrations between 2 and 10 times higher in the fresh leaves of *R. stylosa* than *A. marina*, while it was the opposite for the other TM. For the senescent leaves, Mn and Ni were the only TM with higher concentrations in the *R. stylosa* trees. Regarding the major elements, P and K had concentrations 5 and 3 times higher in the senescent

leaves of *A. marina*, respectively, while Na and Ca had concentrations 2 and 14 times higher in the senescent leaves of *R. stylosa*, respectively.

For both mangrove species, at the control site where the senescent leaves were collected, Co, Cu, Ni, Pb, Ti, and Zn were measured between 1.5 and 31 times in higher concentrations in the fresh leaves than the senescent leaves. Al, As, and Fe were measured in higher concentrations in the fresh leaves than the senescence leaves of *R. stylosa* but it was the opposite for *A. marina*.

Between sites, only Mn was measured in significantly higher concentrations in the leaves of the urban mangrove forest for both species compared to the control mangrove forest. For *A. marina*, it was also the case for Cr, and for *R. stylosa* it was the case for Co, Cu, Fe, and Zn. Conversely, Ni and Pb had higher concentrations in the leaves collected at the control site for both species.

Table 1. Mean element concentrations (±SD) in the fresh and senescent leaves of *A. marina* and *R. stylosa* at the urban and control sites.

Species	*A. marina*			*R. stylosa*		
Site	**Control**		**Urban**	**Control**		**Urban**
Leaf	**Fresh**	**Senescent**	**Fresh**	**Fresh**	**Senescent**	**Fresh**
Major element concentrations (mg g^{-1})						
Na	NA	17±1	NA	NA	33±1	NA
Mg	NA	12±1	NA	NA	13±1	NA
P	0.93±0.05	1.0±0.1	NA	0.66±0.01	0.21±0.02	NA
K	NA	14±1	NA	NA	5.6±0.5	NA
Ca	NA	1.0±0.2	NA	NA	14±1	NA
Trace metal concentrations (µg g^{-1})						
Al	13±1	39±10	6.2±1.2	31±44	9.0±0.01	29±10
As	3.3±1.5	4.1±0.1	0.98±0.44	0.49±0.12	0.30±0.17	0.49±0.14
Co	0.15±0.02	0.09±0.04	0.18±0.03	0.09±0.05	0.02±0.01	0.15±0.04
Cr	0.05±0.01	0.43±0.16	0.38±0.04	0.50±0.31	0.51±0.19	0.34±0.13
Cu	14±6	4.4±0.7	8.6±0.7	2.6±0.7	0.60±0.12	4.6±0.6
Fe	122±24	238±25	42±2	47±51	32±1	67±16
Mn	33±9	34±1	66±7	99±10	157±10	321±130
Ni	4.3±0.3	0.14±0.24	1.4±0.1	3.7±0.6	1.9±0.7	2.1±1.1
Pb	0.15±0.25	0.10±0.01	0	0.57±0.20	0.08±0.01	0.24±0.21
Ti	2.6±0.7	1.7±0.1	1.6±0.5	1.6±2.1	0.46±0.02	2.0±0.50
Zn	18±3	9.2±0.8	14±1	3.5±2.1	1.2±0.1	6.5±1.4

3.2. Changes in major element concentrations during leaf litter decomposition

For all stands, K was the fastest released major element from the litterfall during decomposition with a loss between 65% and 87% of initial K concentration in the first 7 days of decomposition (Fig. 2A). K concentration was rather stable for the rest of the decomposing

experiment. Similarly, the loss of Mg was also high in the first 14 days of decomposition, between 50% and 60% (Fig. 2B).

At the urban site, Na showed single exponential decay, with calculated half-lives of 53 days for *A. marina* and 24 days for *R. stylosa*. After 72 days of decomposition, only 2% of the initial Na concentration was left in the litterfall of *R. stylosa* and 10% in that of *A. marina*. However, at the control site, Na fluctuated in the litterfall of both species with maximum enrichment after 14 days of decomposition and maximum loss after 28 days of decomposition (Fig. 2C).

For *R. stylosa*, compared to the initial concentration in the senescent leaves, P increased by 72% at the control site and by 408% at the urban site. The litterfall of *A. marina* at the urban site had a stable P concentration, while that of the control site slowly decreased with increasing decomposition (Fig. 2D). Ca also showed different trends between species. For *R. stylosa*, about 65% of initial Ca concentration was lost in the litterfall after 72 days of decomposition at both sites. In the litterfall of *A. marina*, Ca concentration compared to t0 increased by 130% in the first 14 days of decomposition (Fig. 2E).

The half-lives of K were in order: control *R. stylosa* (13 d) > urban *A. marina* (12 d) > urban *R. stylosa* (9 d) > control *A. marina* (7 d). The half-lives of Mg were in order: urban *A. marina* (24 d) > control *A. marina* (19 d) > control *R. stylosa* (16 d) > urban *R. stylosa* (14 d). For Na, only at the urban site did the element follow an exponential trend and was lost at a higher rate for *R. stylosa* ($t_{1/2}$ of 24 d) than *A. marina* ($t_{1/2}$ of 53 d). In addition, only did the litterfall of *R. stylosa* exponentially lost Ca with decomposition, with half-lives of 43 days at the urban site and 30 days at the control site. Finally, P had a half-life of 99 days in the litterfall of *A. marina* at the control site.

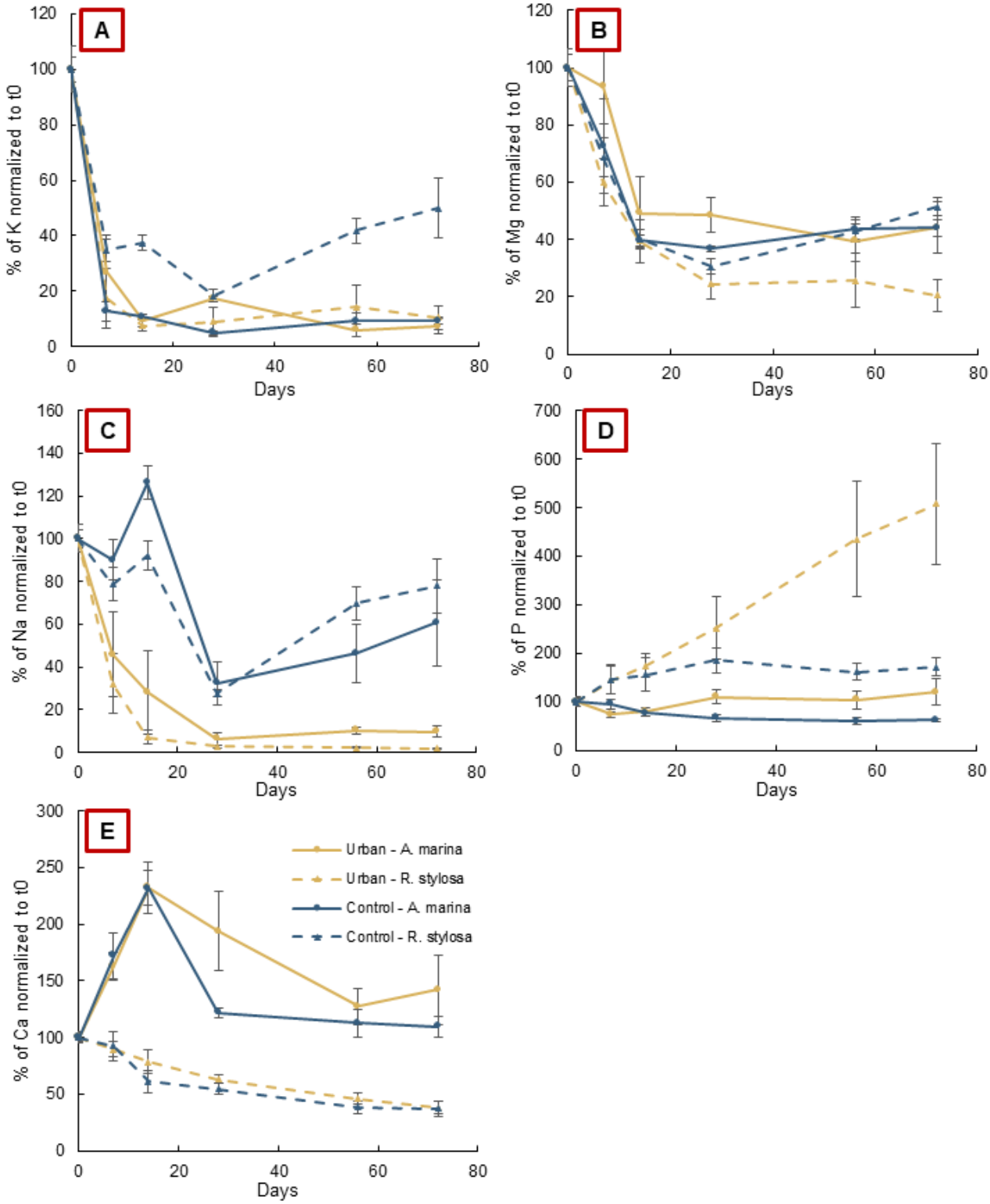

Figure 2. Mean percentage (SD as error bars) of (A) potassium, (B) magnesium, (C) sodium, (D) phosphorous, and (E) calcium in the leaf litter with decomposition relative to t0 at the urban and controls sites for *A. marina* and *R. stylosa*.

3.3. Changes in trace metal concentrations during leaf litter decomposition

The sum of concentrations of TM in the leaf litter continuously increased with decomposition at the control site with concentrations 32 times and 48 times higher for *A. marina* and *R. stylosa,*

respectively, after 72 days (Fig. 3A & 3B). At the urban site, the sum of TM increased prior to a decreasing phase after 28 days for *A. marina* and 56 days for *R. stylosa*. After 72 days of decomposition the sum of TM was 43 times higher than in the senescent leaves for *A. marina* and 27 times higher for *R. stylosa* (Fig. 3C & 3D).

Regardless of site, most TM had significantly higher concentrations in the leaf litter after 72 days of decomposition, relative to the initial concentrations in the senescent leaves. The exceptions were As for *A. marina* and Mn for *R. stylosa* at both sites. Distinct trends were noticed for most TM with litterfall decomposition: 1) increase in concentrations up to 72 days, observed at the control site, 2) increase in concentrations up to 56 days, observed for *A. marina* at the control site and *R. stylosa* at the urban site, and 3) increase in concentrations up to 28 days, for the litterfall of *A. marina* at the urban site (Fig. 3). Mn for both species and in both mangrove forests, and As for the litterfall of *A. marina,* differed from the other TM with a decreasing phase in the first days of the experiment. For *A. marina* at the urban site, Cu and Zn also showed a decreasing phase in the first days of the experiment (Fig. 3C).

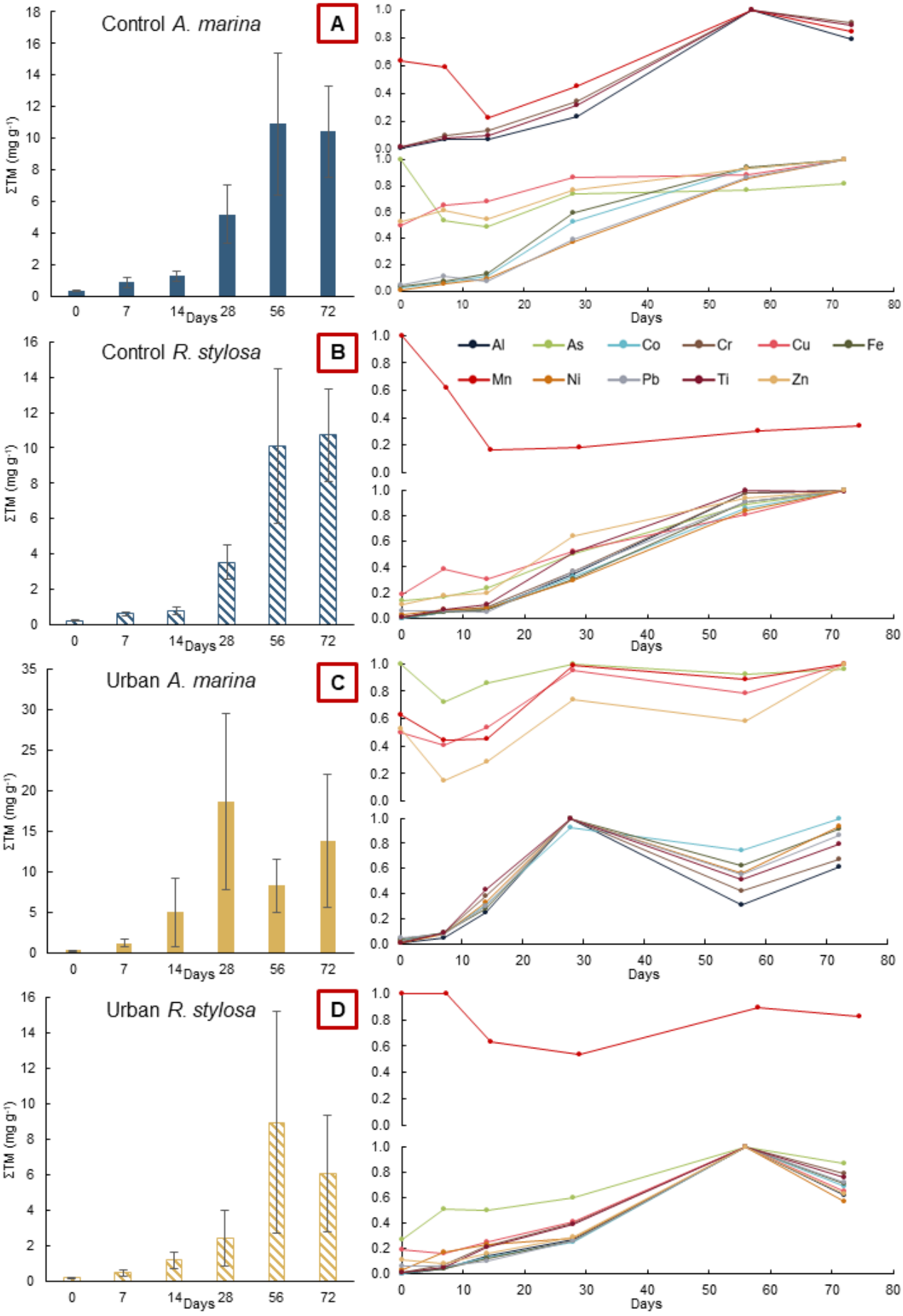
Control A. marina
A
ΣTM (mg g-1)
Days
Control R. stylosa
B
Al
As
Co
Cr
Cu
Fe
Mn
Ni
Pb
Ti
Zn
Urban A. marina
C
Urban R. stylosa
D

Figure 3. Mean sum of the measured trace metals in mg g^{-1} (SD as error bars) in the litterfall during decomposition and concentrations of the individuals trace metals in the litterfall during decomposition normalized to the highest concentration for each trace metal at the control site for (A) *A. marina* and (B) *R. stylosa* and at the urban site for (C) *A. marina* and (D) *R. stylosa*.

4. Discussion

4.1. Initial composition of mangrove leaves differed between species and sites

In the current study, leaf TM concentrations were compared between two sites. Higher concentrations of As, Cr, Fe, and Ni were found in leaves at the control site, likely due to elevated TM levels in the soil. A previous study reported greater soil concentrations for these TM at the control site due to conditions enabling TM storage in soils (Robin et al., 2022). Conversely, Cu and Mn were more concentrated in fresh leaves from the urban site in the present study. Earlier research on the same mangrove area showed significantly higher Cu and Mn in the urban site's soil fractions as they are brought to the forest via the urban runoff (Robin et al., 2022). Furthermore, Co, Cr, and Fe were measured in higher levels in fresh leaves of one or both species at the urban site in the present study. Previous findings demonstrated that leaf bioconcentration factors for these TM were notably higher in the urban site, indicating greater TM transfer from soil to leaves, likely due to salt-TM complexes and pyrite-TM interactions limiting mobility in the control site's soil (Robin et al., 2022). The results suggest that the urban runoff is a factor controlling mangroves leaves' TM concentrations, but other environmental factors may also largely contribute to TM dynamics.

Differences in major element concentrations were observed in senescent leaves between species. In this study, *A. marina* had higher P and K concentrations compared to *R. stylosa*, while *R. stylosa* had elevated Na and Ca concentrations, as previously observed in other mangrove forests (Alongi, 2021; Clough, 1984; Steinke and Ward, 1987; Tam et al., 1990). High Na and Ca concentrations in *R. stylosa* may be due to the proximity of the stand to the sea as suggested in another study on mangrove leaves chemical composition (Ahmed et al., 2010). The K^+ and Na^+ balance differences between the salt excluder (*R. stylosa*) and salt secretor (*A. marina*) species likely explain the K concentration variation (Parida and Jha, 2010; Ray et al., 2021). Lower P levels in *R. stylosa*'s senescent leaves may be attributed to P redistribution within fresh tissues to limit losses during litterfall, which also accounts for higher P values in fresh leaves (Kathiresan and

Bingham, 2001). The higher P concentrations in the leaves of *A. marina* may result from enhanced P transfer facilitated by its microbial community on the forest floor rich in phosphatase-producing bacteria, as indicated in prior research (Abhijith et al., 2018).

Three factors play a role in differences between fresh and senescent leaves, including species position in the forest, salt tolerance mechanisms, and metabolism. The subsequent discussion assesses the impact of each of these factors on changes in major element concentrations during leaf litter decomposition.

4.2. Major element dynamics during leaf litter decomposition

4.2.1. An expected order of lability for the major elements

For both species and sites, K showed the fastest leaching among major elements. Literature shows K concentration drops rapidly initially due to leaching, then stabilizes during mangrove litter decomposition (Chale, 1993; Hossain et al., 2014; Steinke and Ward, 1987; Tam et al., 1990; Vinh et al., 2020). K^+ is highly mobile, lacking structural roles in plants, making it more labile than other cations (Bonanomi et al., 2010; Steinke and Ward, 1987). Similarly, Mg concentration quickly decreases during initial leaching before stabilizing, consistent with literature findings in multiple environments (Bonanomi et al., 2010; Gautam et al., 2016; Vinh et al., 2020). Partial Mg leaching in early decomposition may be due to the absence of complex organic binding, as suggested in the literature (Van Der Valk and Attiwill, 1984). The major element lability sequence in this study follows K > Mg > Ca > P, aligning with previous litterbag experiments (Bonanomi et al., 2010; Gautam et al., 2016). This confirms consistent major element lability in litterfall for both species. Hence, we suggest that the urban runoff has no impact on the major element lability order in litterfall.

4.2.2. Major element dynamics influenced by the urban runoff

In the urban forest, K and Mg decayed faster under *R. stylosa* than *A. marina*. *R. stylosa*'s floor stays submerged by ongoing runoff, potentially enhancing leaching. In fact, previous research showed that at this site, the urban runoff intensified leaf litter degradation, influencing both mass loss and molecular changes in OM (Robin et al., 2024). The change in Na concentrations during the decomposition of the leaf litter was site-specific, potentially associated with the forest structure or

the soil physico-chemical parameters. At the urban site, the rate of Na loss exceeded the rate of mass loss (Robin et al., 2024), with a half-life of 24 days for *R. stylosa*, similar to that obtained in a tropical mangrove forest in Vietnam during the rainy season, suggesting that the runoff affects Na loss comparable to high pluviometry (Vinh et al., 2020). In the present study, the release of Na to the surroundings was greater in the first days of decomposition suggesting that loss is due to leaching. Water-soluble Na in leaves may encourage leaching, and the low salinity of the urban runoff (0 g L^{-1}) (Robin et al., 2022) might promote this process in flooded *R. stylosa* areas. The high salinity at the control site (35 g L^{-1}) and higher Na concentration in the leaves, possibly contributes to increasing Na levels during litter decomposition. Finally, P enrichment was significantly greater at the urban site, reaching 400% after 72 days, implying urban runoff may be a source of P (garden fertilizers and pet waste).

The urban runoff therefore enhances K, Mg, and Na leaching during litter decomposition due to its low salinity and its continuous flow through the mangrove forest, especially for *R. stylosa*. The runoff also seems to influence major element dynamics during leaf litter decomposition as it can be a source of elements such as P.

4.2.3. Major element dynamics may be influenced by the species position

Ca concentration in *R. stylosa* leaf litter linearly declined at both sites, while *A. marina* showed an increase over the 14 first days of decomposition before loss. Initial Ca concentration was 4.5 times higher in *R. stylosa* than *A. marina*, possibly making its Ca more leachable. Stand immersion may have influenced Ca dynamics as well, aligning with results from a river-influenced mangrove in Vietnam (Vinh et al., 2020). In the study in Vietnam, greater loss occurred in more flooded areas like *R. stylosa* stands of the current study, submerged by tides or runoff. Ca complexation with refractory compounds may explain the initial increase in Ca concentration in the leaf litter of *A. marina*.

In the present study, the P concentration in the leaf litter of *A. marina* remained stable, but the concentration in the litter of *R. stylosa* increased relative to senescent leaves at both sites. Literature typically shows mangrove litter P decrease initially due to leaching (Hossain et al., 2014;

Mfilinge et al., 2002; Sánchez-Andrés et al., 2010; Steinke and Ward, 1987; Tam et al., 1990; Vinh et al., 2020), and then microbial use (Chale, 1993). P rise during decomposition can result from complexation of P with refractory molecules (Vinh et al., 2020) or floor-to-litter transfer via bacteria or fungi (Lindahl et al., 2007). We suggest that the increase in P concentration in the litter of *R. stylosa* might be due to water column P input, and therefore be dependent on the stand position in the forest. Initial leaf composition may also contribute to the dynamic of P; *R. stylosa* had 5 times less P in senescent leaves than *A. marina*, potentially offering more P capture sites.

In summary, Ca and P concentrations changes during decomposition are species-specific. Hydroperiod-driven Ca loss and P enrichment depend on species' forest positions. Initial litter composition also influences trends based on species. In this study, major elements showed concentrations variations depending on the element, the mangrove forest, and the mangrove species. Next, we will discuss if those parameters also influence TM dynamics during leaf litter decomposition.

4.3. Trace metal dynamics during leaf litter decomposition

4.3.1. An expected trace metal enrichment during leaf litter decomposition

All TM, except As and Mn, exhibited higher concentrations in the leaf litter after 72 days of decomposition compared to senescent leaves, aligning with literature expectations (Ramos e Silva et al., 2006; Rice and Windom, 1982; Vinh et al., 2020). The enrichment of TM during decomposition relates to their exchange from forest soil or water column to leaf litter on the soil surface (Silva et al., 1998). Microbial immobilization through TM uptake by microbial biomass or TM complex formation with microbial-produced extracellular material can also enhance TM accumulation in leaf litter (Rice and Windom, 1982). Additionally, a previous study on the same samples showed the increase of N concentration in decomposing litterfall (Robin et al., 2024), which likely leads to TM accumulation due to complex formation with N-rich compounds like proteins (Rice and Windom, 1982). Still, initial leaching phases were observed for some TM (As, Cu, Mn, Zn).

4.3.2. Initial leaching of trace metals controlled by their states and concentrations in porewaters

As, Cu, Mn, and Zn displayed initial leaching upon litterfall decomposition. Leaching of As was observed only for *A. marina* litter, possibly indicating molecular binding. In a prior study using the same samples as the present study, it was observed that the leaf litter of *A. marina* initially contained more lignin than that of *R. stylosa*. However, within the first seven days of decomposition, a significant reduction in lignin concentration was observed for *A. marina* (Robin et al., 2024). Therefore, we suggest that As may form complexes with lignin molecules. Urban site Cu and Zn also showed leaching before enrichment in the litterfall of *A. marina*, a pattern observed in previous mangrove litterbag experiments (Ramos e Silva et al., 2006; Vinh et al., 2020). Cu and Zn concentrations in upper soil porewaters (0-5 cm) were notably lower beneath *A. marina* at the urban site (Supplementary Figure 1) (Robin et al., 2022), suggesting favorable exchange from litterfall in early decomposition.

Mn alone exhibited relative concentration decrease in the first weeks for both species and sites, which was already observed during a litterbag experiment in a Vietnamese mangrove forest (Vinh et al., 2020). Mn is expected to be more leachable than other TM such as Fe and Zn as it is less able to bound with refractory molecules (Silva et al., 1998). Mn's soluble form in leaves (Mn^{2+}) (Joardar Mukhopadhyay and Sharma, 1991), easily solubilized during decomposition, may not oxidize to immobilized Mn^{4+} due to mangrove floor anoxia (Vinh et al., 2020).

Loss of TM from litterfall appears element-dependent, influenced by oxidation state, molecular interactions, and porewater concentrations. Results of the present study also show differences in TM enrichment between stands and are discussed next.

4.3.3. Differences between stands due to trace metal bioavailability and species absorption capacity

While most TM concentrations were generally higher in decomposed litterfall than senescent leaves, diverse trends were observed. At the urban site, peak TM concentrations relative to bulk leaf litter occurred at 28 days for *A. marina* and 56 days for *R. stylosa*. In contrast, at the control site, these peaks were reached at 56 or 72 days for *A. marina* and 72 days for *R. stylosa*. A recent study on the same sites has indicated higher TM bioavailability in the urban soils compared to the control soils (Robin et al., 2022). The lower bioavailability at the control site was linked to a greater proportion of TM in the refractory soil fraction (Robin et al., 2022). Elevated root and leaf

bioconcentration factors for most TM were significantly higher at the urban site, potentially leading to enhanced TM transfer from mangrove soil to litterfall (Marchand et al., 2016).

In absolute concentrations (μg g^{-1}), peak TM concentrations in the litterfall of *A. marina* at the urban site surpassed those in the other three areas (Supplementary Table 2). This suggests that maximum TM concentrations were attained earlier during litter decomposition of *A. marina*, possibly due to the leaf litter reaching its absorption limit. Additionally, absolute TM concentrations showed the litterfall of *A. marina* had higher levels than the litter of *R. stylosa* in the initial decomposition days at both sites. However, after 56 days at the urban site and 72 days at the control site, leaf litter of *R. stylosa* exhibited higher total TM concentrations and the majority of individual TM concentrations (Supplementary Table 2). Tidal flooding at the control site and constant urban rainwater flow at the urban site favor redox reactions on the floor of *R. stylosa* stands, increasing biogeochemical reactivity that influences TM precipitation-dissolution cycles (Noël et al., 2014), potentially facilitating TM export or transfer to litterfall on the soil surface. Prior work by Marchand et al., (2016) indicated decreasing TM stocks in mangrove soil closer to the shoreline. We suggest that the stand immersion influences TM accumulation in mangrove litter.

4.4. Limitations of the experiment and models

The litterbag experiment was conducted using senescent leaves from the control site only for both species. This design of experiment aimed to evaluate the changes in leaf litter major element and trace metal concentrations over 72 days of degradation with a focus on the influence of urban runoff on these changes. The choice of having the same senescent leaves at both sites enabled to limit the factors potentially affecting these changes other than runoff such as initial senescent leaves composition. The influence of initial composition was evaluated between species but not between sites. This experiment design was therefore not a limitation regarding the general processes investigated in this study. However, it is not possible from this experiment to quantify the elements released, stored, or gained by the leaf litter at the urban site. Furthermore, the experiment ended after 72 days of degradation as the mass remaining was the lower limit to conduct all the analyses. A longer experiment could have given further information relative to the refractory degradation stage.

5. Conclusion

The comparison between an urban mangrove forest and a control mangrove forest highlighted that continuous urban rainwater runoff enhances leaching of specific major elements (K, Mg, Na) during litterfall decomposition and could serve as a source of P. However, this runoff did not alter major element lability order and is not the sole influencer of element dynamics in litter decomposition. Ca and P concentrations exhibited species-specific patterns tied to stand position, hydroperiod, and potentially initial litter composition. TM primarily increased during litterfall decomposition, implying a transfer from forest floor to litterfall. As and Mn diverged due to complexation and oxidation state differences. Like major elements, urban runoff appeared to impact TM enrichment, boosting TM bioavailability for forest floor-to-litter transfer. Initial litter composition and species physiology shaped TM absorption capacity during decomposition. Future research could assess leaf litter's contribution to exported TM and nutrients in adjacent ecosystems as the results obtained here suggest that the urban site may contribute more than the control site to the export of potentially toxic TM but also nutrients.

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