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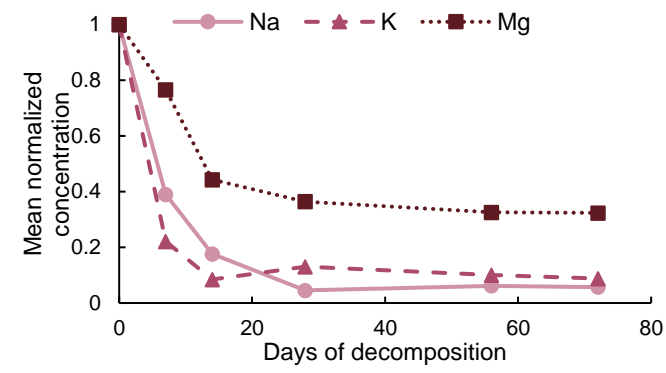
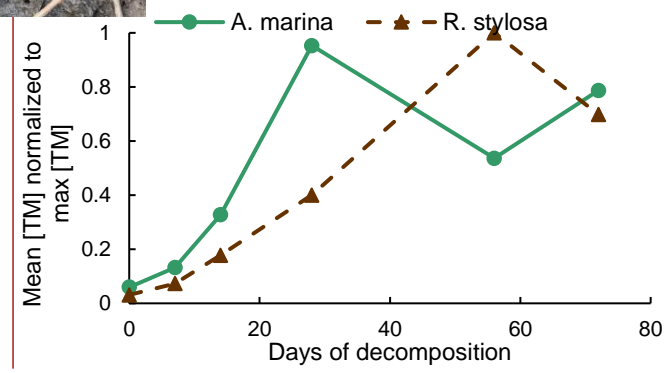
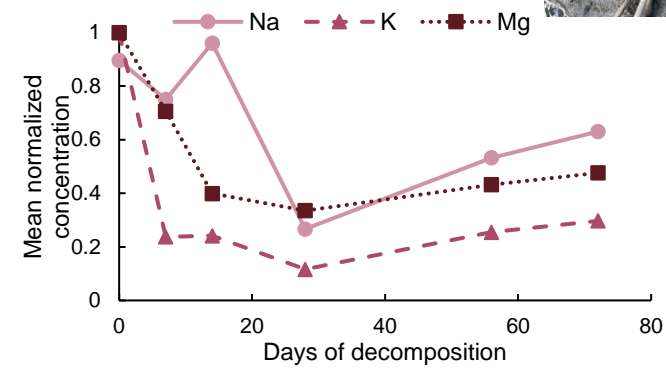
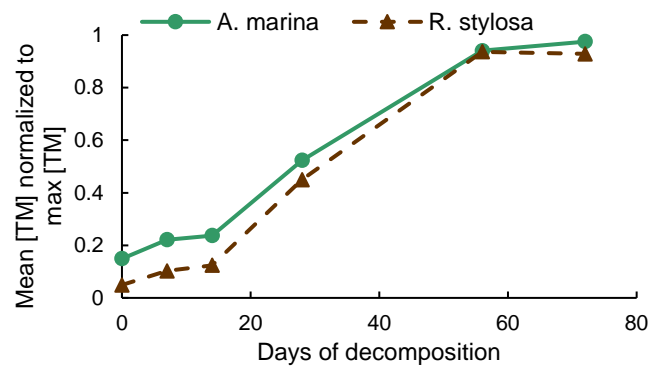
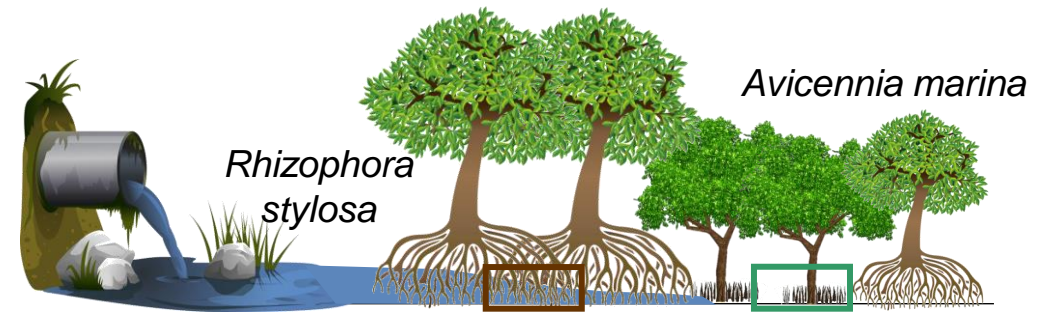
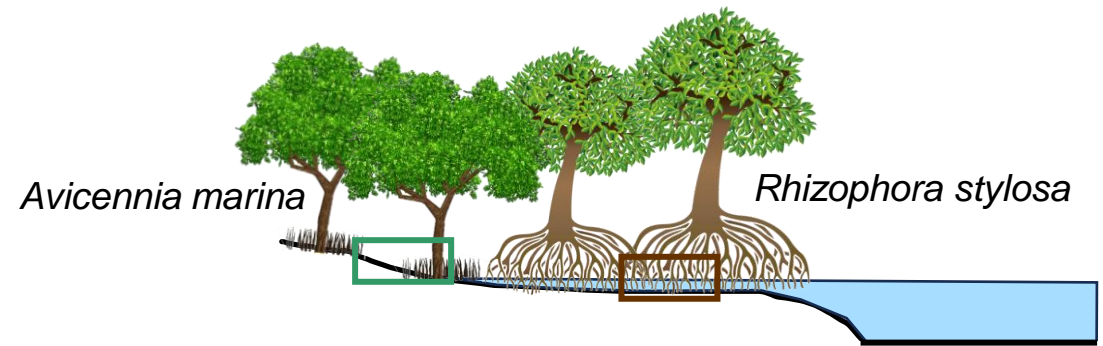


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# Control mangrove forest

# Litterbag experiment

# Urban mangrove forest



- 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

# 1 Dynamics of major and trace elements during leaf litter decomposition in semi- 2 arid mangrove subject to urban runoff

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## 11 Abstract

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

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

## 24 1. Introduction

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

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

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432 In mangrove forests, litter production constitutes 31% of the overall net primary production  
5 within the forest (Bouillon et al., 2008). Litterfall decomposition actively participates in the energy  
633 and material flow in the forest and constitutes an energy source for coastal productivity(Lugo and  
7  
834 Snedaker, 1974; Twilley et al., 1986). Mangrove leaves comprise the predominant fraction of litterfall  
9  
1035 (Day et al., 1987; Twilley et al., 1986), ranging between 40% and 90% (Mackey and Smail, 1996).  
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1236 Consequently, most investigations into litterfall decomposition center on leaf litter, employing the  
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1537 litterbag experiment methodology (Nordhaus et al., 2017; Vinh et al., 2020; Yang et al., 2018).Leaf  
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1738 litter degradation occurs in three main stages: 1) the initial leaching of soluble compounds, 2) the  
18  
1939 colonization by microorganisms (i.e., fungi, bacteria, and protozoa) and decomposition via litter  
20  
2140 breakdown (Cundell et al., 1979; Fell et al., 1975; Fell and Master, 1980), and 3) the slow refractory  
22  
23  
2441 phase where the least labile molecules are consumed (Tam et al., 1990; Valiela et al., 1984; Wilson  
25  
2642 et al., 1986). Many factors can influence litterfall decomposition, including the initial composition of  
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2843 the leaves, the leaf consuming fauna and microbial communities on the floor, and frequency and  
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3144 degree of inundation (Fell and Master, 1980; Lugo and Snedaker, 1974; Mfilinge et al., 2002).  
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3646 Litterfall organic matter (OM) can be consumed by autochthonous fauna and microorganisms  
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3847 on the forest floor, dissolved during decomposition, and subsequently be conveyed to adjacent  
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4048 ecosystems during ebb tides, either as debris or dissolved organic/inorganic carbon (Kida et al.,  
41  
4249 2019; Ray et al., 2018; Taillardat et al., 2018). Similarly, elements such as TM can be leached to  
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4550 the surroundings during leaf litter degradation, or absorbed by the litter consuming fauna (Fell and  
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4751 Master, 1980; Fourqurean and Schlau, 2003; Gautam et al., 2016; Nordhaus et al., 2017). Elements  
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4952 found in their soluble form in mangrove leaves, such as most major elements, are easily leached  
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5153 during litter degradation (Bonanomi et al., 2010). On the other hand, the enrichment of major  
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5454 elements and TM in leaf litter during the degradation process is attributed to inputs from the water  
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5655 column (Lacerda et al., 1988; Rice and Windom, 1982; Tam et al., 1990; Vinh et al., 2020). In  
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5856 mangrove soils, TM precipitation/dissolution cycles are directly influenced by floor inundation and  
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6057 redox reactions (Noël et al., 2014), which greatly determine the transfers of TM between the  
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58 mangrove floor and the decomposing litter (Vinh et al., 2020). In literature, enrichment of TM in  
159 decomposing mangrove litterfall has been largely observed and is explained by the complexation of  
2 dissolved TM by organic molecules present in the litter (Scheid et al., 2009; Vinh et al., 2020;  
360 4 Zawislanski et al., 2001). To understand the dynamics of major elements and TM in leaf litter during  
561 6 decomposition is crucial to better understand the export of these elements to the adjacent  
7 8 ecosystems of their intake by litter-consuming fauna.  
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12  
1364 New Caledonia is one of the top four pacific territories with the most mangrove forests  
14 (Hamilton and Casey, 2016), as mangroves cover 80% of the west coast of the main island. The  
1565 16 mangrove forests of this region, characterized by a semi-arid climate and a semidiurnal tidal pattern  
1766 18 (Douillet, 2001), showcase simple biodiversity structures. Species monospecific stands prevail,  
1967 20 shaped by topography and soil salinity (Baltzer, 1981; Marchand et al., 2011). *Rhizophora* spp.,  
21 22 developing seaside, constitute over half of the mangrove flora, while *Avicennia marina*, developing  
2469 25 at higher elevations and saltier soils, represent around 15% of the archipelago's mangrove  
2670 27 vegetation (Deborde et al., 2015). Although the human population density is low in New Caledonia,  
2871 29 the population living in urban areas has significantly increased this last decade, resulting in the  
30 31 development of urban infrastructures along the littoral zone, where mangrove forests develop (Insee,  
3373 34 2020). A recent study looked into the influence of coastal urbanization on these mangroves and  
3574 36 found that part of the mangrove forest, receiving urban rainwater runoff for >50 years, was constantly  
3775 38 submerged by runoff water, leading to the development of *R. stylosa* trees far taller than is typical  
39 40 for such semi-arid mangrove forests (Robin et al., 2022). Robin et al. (2022) showed that the  
41 42 rainwater runoff controls the physico-chemical parameters of the mangrove soil, reducing soil salinity  
43 44 and elevating pH levels. Furthermore, urban TM concentrations in the soil exceed those present in  
4679 47 a "control" mangrove forest, implying greater TM mobility (Robin et al., 2022). A litterbag experiment  
48 49 conducted in the same mangrove forest revealed that the urban runoff favors leaf litter decay on the  
50 51 mangrove floor by promoting microbial development on the soil surface (Robin et al., 2024).  
52 5382

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55  
5683 This study aimed to assess major element (K, Mg, Na, Ca, P) and TM (Al, As, Co, Cr, Cu,  
57 Fe, Mn, Ni, Pb, Ti, Zn) dynamics in leaf litter decomposition, examining the influence of urbanization.  
5884 59 While major element and TM dynamics during litterfall decay have been explored in various studies  
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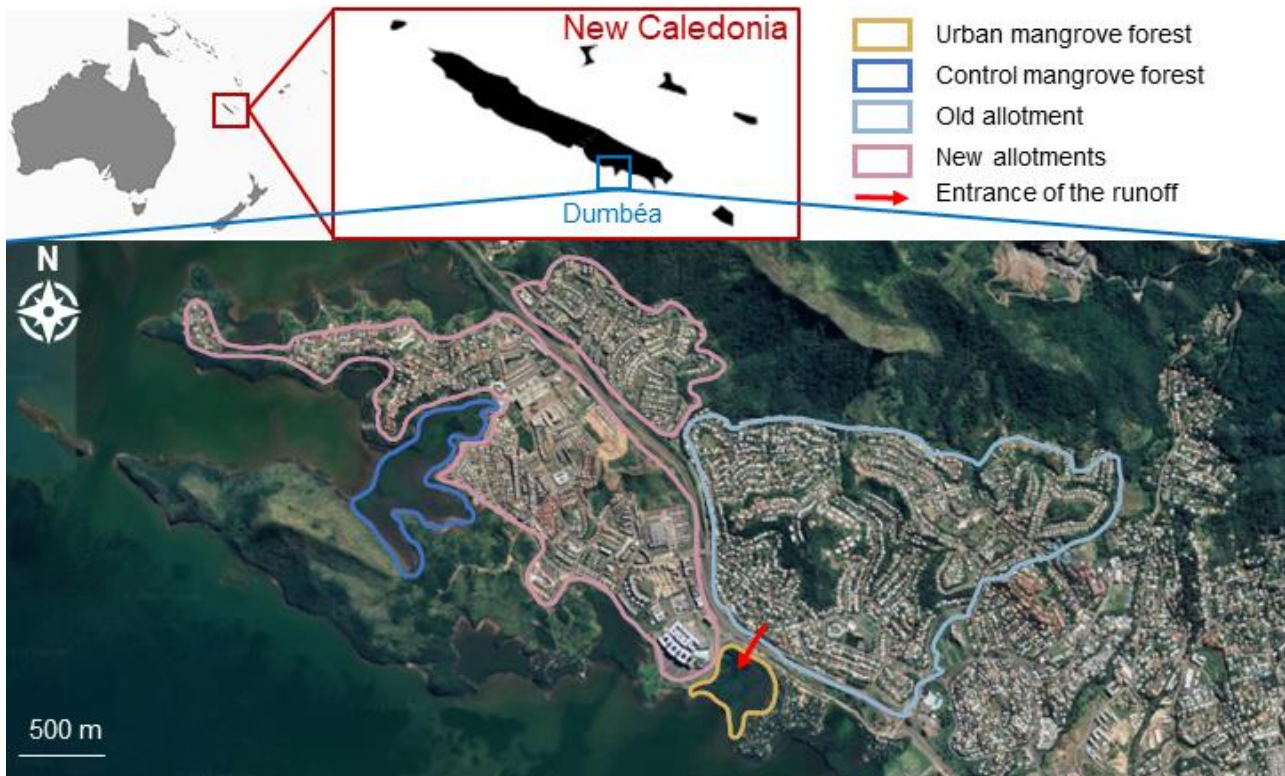
86 (Chale, 1993; Hossain et al., 2014; Ramos e Silva et al., 2006; Rice and Windom, 1982; Silva et al.,  
187 1998; Steinke and Ward, 1987; Tam et al., 1990; Van Der Valk and Attiwill, 1984; Vinh et al., 2020;  
2  
388 Zawislanski et al., 2001), no investigation has looked into the effect of urbanization on such  
4  
589 processes nor in semi-arid mangrove settings. In this study, two semi-arid mangrove forests—  
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790 control and urban—along New Caledonia's west coast, were selected. Both forests share a common  
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991 geological unit as the source of their watersheds, yet the urban forest has received urban rainwater  
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11 runoff for over five decades. Employing a 72-day litterbag experiment, conducted on the main  
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13 mangrove species (*Rhizophora stylosa* and *Avicennia marina*), we hypothesize that the half-lives of  
1493  
15 major elements usually leached early in the decomposition process such as K and Mg will be shorter  
1694  
17 at the urban site than the control site due to the constantly flooding runoff. We also hypothesize that  
1895  
19 greater TM enrichment in leaf litter will occur at the urban site due to their higher bioavailability in  
20  
2196 the soil at this site.  
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## 2698 2. Materials and methods

### 2999 2.1. Study sites

31  
3200 The city of Dumbea in New Caledonia has had a population increase of 33% this last decade,  
33  
3401 resulting in the development of urban infrastructures on the littoral zone (Insee, 2020). Two  
35  
3602 mangrove forests in Dumbea were selected as study sites (Fig. 1). These sites had the same  
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3803 volcano-sedimentary watershed and are dominated by the same two mangrove species (*R. stylosa*  
39  
40 and *A. marina*). One site was located in the Apogoti Bay (22°12'08"S, 166°26'20"E). This site has  
4104  
42 no direct anthropogenic inputs (limited housing density) into the mangrove forest, and it was selected  
4305  
44 as a control mangrove forest. Another site is the urban mangrove forest (22°12'39"S, 166°27'19"E)  
4506  
46 exposed to rainwater runoff from the upper allotment (mainly housing lots for 10 000 inhabitants),  
47  
4807 which has been flowing into the mangrove forest for more than 50 years. At the control site, the two  
49  
5008 species develop in monospecific stands similar to most mangrove forests of the west coast, that is,  
51  
5209 *A. marina* landward and *R. stylosa* seaward. At the urban site, another *R. stylosa* stand develops  
53  
5410 more landward than the *A. marina* stand, at the entrance of the runoff, and its soil is always flooded  
55  
5611 with the urban rainwater.  
57  
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5912





**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



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

### 139 *2.3. Major element and trace metal quantification*

140 The elements were measured in the dried ground leaves by ICP-OES after multiwave  
141 extraction. Briefly, 200 mg of each fresh, senescent, and degraded sample was weighed in a Teflon  
142 vessel. A volume of 5 ml of 70% HNO<sub>3</sub> (Ajax Finechem) and 1 ml of 30% H<sub>2</sub>O<sub>2</sub> (Carlo Erba) were  
143 added to the vessel. The digestion program started with a 10 min heating step from room  
144 temperature to 160 °C, then a 15 min step to 210 °C, followed by a 10 min plateau and a 20 min  
145 cooldown step. The result of the extraction was transferred to a polypropylene tube. The volume  
146 was adjusted to 13 ml with Milli-Q water and placed at 4 °C until ICP-OES (Varian 730-ES) analysis  
147 at the chemistry laboratory LAMA of the French Research Institute for Sustainable Development in  
148 New Caledonia. For quality control, certified material (IPE sample ID 949) was also extracted and  
149 analyzed (Supplementary Table 1). The concentrations of major elements and TM were obtained  
150 using a calibration curve prepared from a stock solution (CPAChem) of the measured elements at  
151 100 mg L<sup>-1</sup>.

### 152 *2.4. Data analyses*

#### 153 *2.4.1. Major elements*

154 To plot the changes in major element concentrations in the litterfall with degradation, the  
155 measured element concentrations in µg g<sup>-1</sup> of dry sample were converted to percentage relative to  
156 t<sub>0</sub>. The values were calculated as follows:

$$157 \text{Element concentration at } t_n (\%) = ([\text{element}]_n * 100) / [\text{element}]_0 \quad (1)$$

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

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

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

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

#### 165 2.4.2. Trace metals

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

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

169 with  $[\text{TM}]_n$  the concentration of the TM at  $t_n$  in  $\mu\text{g g}^{-1}$  and  $[\text{TM}]_{\text{max}}$  the maximum concentration of the  
170 TM between  $t_0$  and  $t_{72}$  in  $\mu\text{g g}^{-1}$ .

### 171 3. Results

#### 172 3.1. Characterization of the fresh and senescent leaves

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

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

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

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

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

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

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

### 3.2. Changes in major element concentrations during leaf litter decomposition

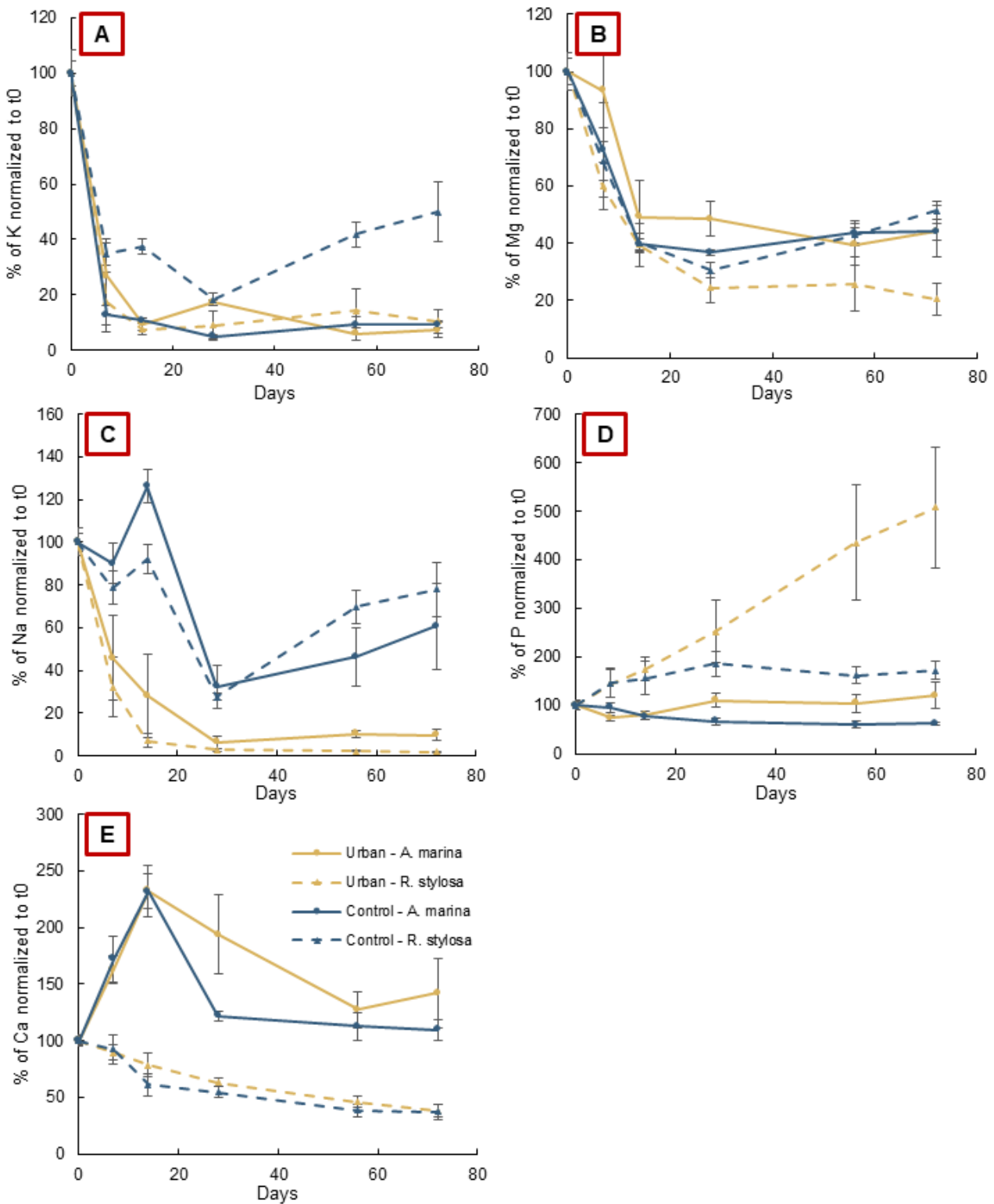
196 For all stands, K was the fastest released major element from the litterfall during  
 197 decomposition with a loss between 65% and 87% of initial K concentration in the first 7 days of  
 198 decomposition (Fig. 2A). K concentration was rather stable for the rest of the decomposing

199 experiment. Similarly, the loss of Mg was also high in the first 14 days of decomposition, between  
200 50% and 60% (Fig. 2B).

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

206 For *R. stylosa*, compared to the initial concentration in the senescent leaves, P increased by  
207 72% at the control site and by 408% at the urban site. The litterfall of *A. marina* at the urban site had  
208 a stable P concentration, while that of the control site slowly decreased with increasing  
209 decomposition (Fig. 2D). Ca also showed different trends between species. For *R. stylosa*, about  
210 65% of initial Ca concentration was lost in the litterfall after 72 days of decomposition at both sites.  
211 In the litterfall of *A. marina*, Ca concentration compared to t<sub>0</sub> increased by 130% in the first 14 days  
212 of decomposition (Fig. 2E).

213 The half-lives of K were in order: control *R. stylosa* (13 d) > urban *A. marina* (12 d) > urban  
214 *R. stylosa* (9 d) > control *A. marina* (7 d). The half-lives of Mg were in order: urban *A. marina* (24 d)  
215 > control *A. marina* (19 d) > control *R. stylosa* (16 d) > urban *R. stylosa* (14 d). For Na, only at the  
216 urban site did the element follow an exponential trend and was lost at a higher rate for *R. stylosa*  
217 ( $t_{1/2}$  of 24 d) than *A. marina* ( $t_{1/2}$  of 53 d). In addition, only did the litterfall of *R. stylosa* exponentially  
218 lost Ca with decomposition, with half-lives of 43 days at the urban site and 30 days at the control  
219 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*.

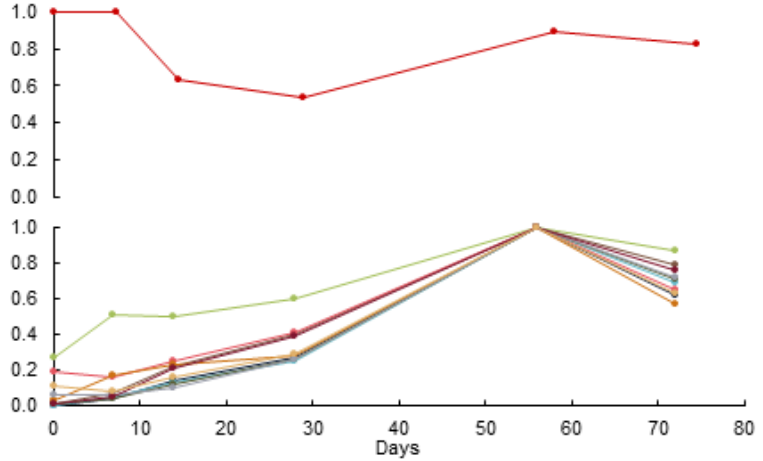
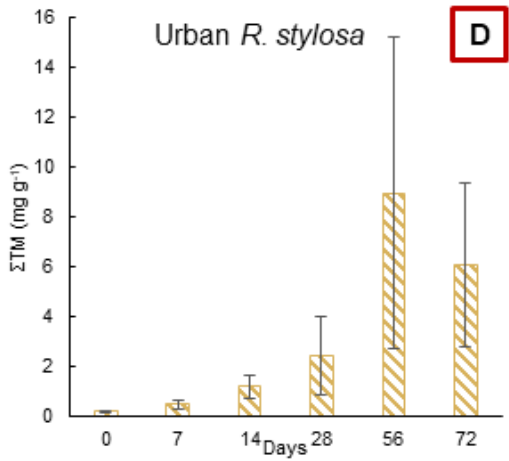
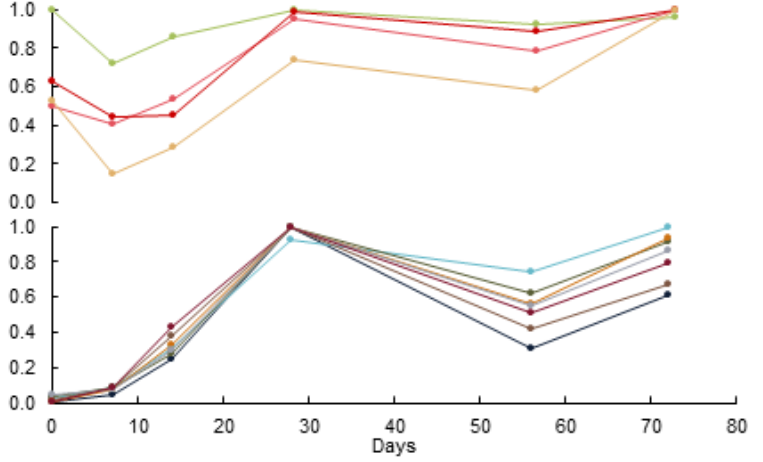
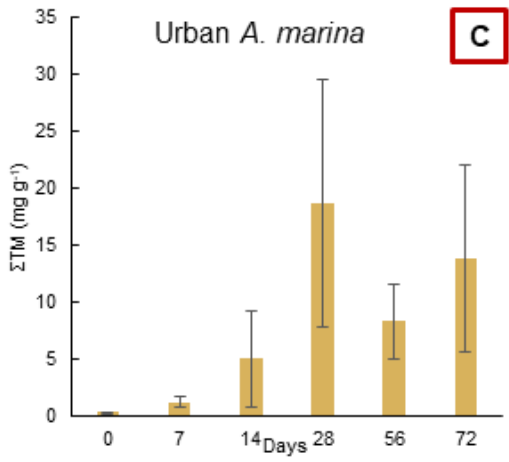
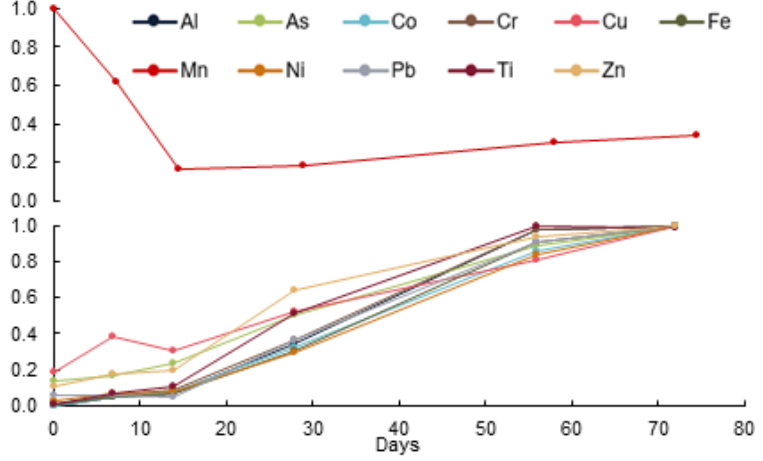
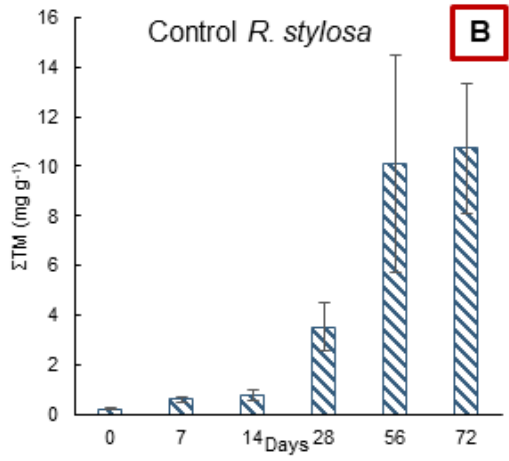
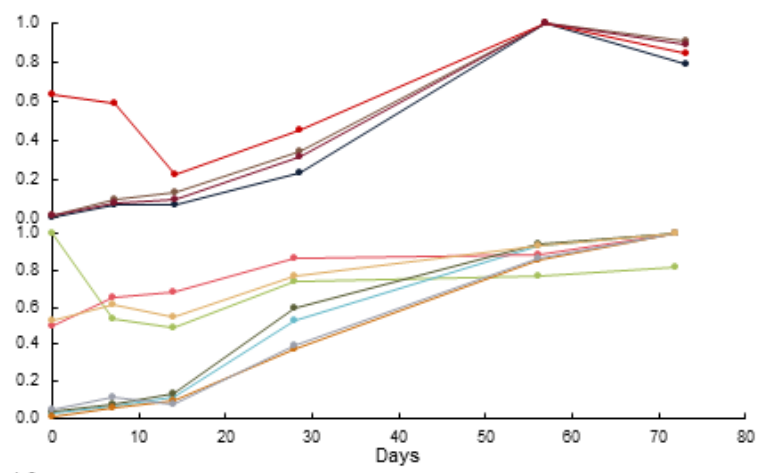
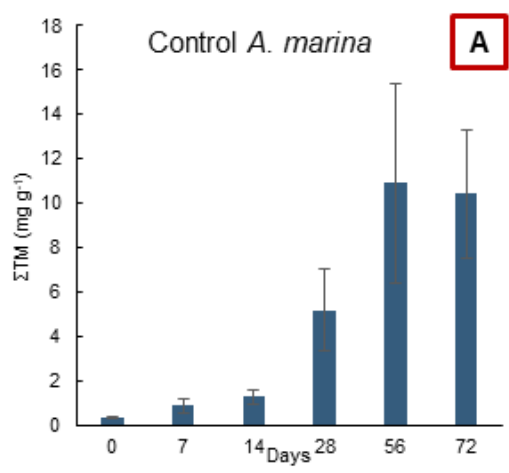
### 3.3. Changes in trace metal concentrations during leaf litter decomposition

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*,

227 respectively, after 72 days (Fig. 3A & 3B). At the urban site, the sum of TM increased prior to a  
228 decreasing phase after 28 days for *A. marina* and 56 days for *R. stylosa*. After 72 days of  
2 decomposition the sum of TM was 43 times higher than in the senescent leaves for *A. marina* and  
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230 27 times higher for *R. stylosa* (Fig. 3C & 3D).  
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231           Regardless of site, most TM had significantly higher concentrations in the leaf litter after 72  
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10 232 days of decomposition, relative to the initial concentrations in the senescent leaves. The exceptions  
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12 were As for *A. marina* and Mn for *R. stylosa* at both sites. Distinct trends were noticed for most TM  
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14 with litterfall decomposition: 1) increase in concentrations up to 72 days, observed at the control  
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16 site, 2) increase in concentrations up to 56 days, observed for *A. marina* at the control site and *R.*  
17 235  
18 *stylosa* at the urban site, and 3) increase in concentrations up to 28 days, for the litterfall of *A. marina*  
19 236  
20 at the urban site (Fig. 3). Mn for both species and in both mangrove forests, and As for the litterfall  
21 237  
22 of *A. marina*, differed from the other TM with a decreasing phase in the first days of the experiment.  
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24 For *A. marina* at the urban site, Cu and Zn also showed a decreasing phase in the first days of the  
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27 experiment (Fig. 3C).  
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242 **Figure 3.** Mean sum of the measured trace metals in mg g<sup>-1</sup> (SD as error bars) in the litterfall during  
243 decomposition and concentrations of the individuals trace metals in the litterfall during  
244 decomposition normalized to the highest concentration for each trace metal at the control site for  
245 (A) *A. marina* and (B) *R. stylosa* and at the urban site for (C) *A. marina* and (D) *R. stylosa*.

#### 246 4. Discussion

##### 247 4.1. Initial composition of mangrove leaves differed between species and sites

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

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

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

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

#### 278 *4.2. Major element dynamics during leaf litter decomposition*

##### 279 *4.2.1. An expected order of lability for the major elements*

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

##### 292 *4.2.2. Major element dynamics influenced by the urban runoff*

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

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

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

#### 313 4.2.3. Major element dynamics may be influenced by the species position

314 Ca concentration in *R. stylosa* leaf litter linearly declined at both sites, while *A. marina*  
315 showed an increase over the 14 first days of decomposition before loss. Initial Ca concentration was  
316 4.5 times higher in *R. stylosa* than *A. marina*, possibly making its Ca more leachable. Stand  
317 immersion may have influenced Ca dynamics as well, aligning with results from a river-influenced  
318 mangrove in Vietnam (Vinh et al., 2020). In the study in Vietnam, greater loss occurred in more  
319 flooded areas like *R. stylosa* stands of the current study, submerged by tides or runoff. Ca  
320 complexation with refractory compounds may explain the initial increase in Ca concentration in the  
321 leaf litter of *A. marina*.

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

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

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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

351 As, Cu, Mn, and Zn displayed initial leaching upon litterfall decomposition. Leaching of As  
352 was observed only for *A. marina* litter, possibly indicating molecular binding. In a prior study using  
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353 the same samples as the present study, it was observed that the leaf litter of *A. marina* initially  
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354 contained more lignin than that of *R. stylosa*. However, within the first seven days of decomposition,  
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355 a significant reduction in lignin concentration was observed for *A. marina* (Robin et al., 2024).  
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356 Therefore, we suggest that As may form complexes with lignin molecules. Urban site Cu and Zn  
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1357 also showed leaching before enrichment in the litterfall of *A. marina*, a pattern observed in previous  
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1358 mangrove litterbag experiments (Ramos e Silva et al., 2006; Vinh et al., 2020). Cu and Zn  
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1359 concentrations in upper soil porewaters (0-5 cm) were notably lower beneath *A. marina* at the urban  
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1360 site (Supplementary Figure 1) (Robin et al., 2022), suggesting favorable exchange from litterfall in  
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1361 early decomposition.  
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2362 Mn alone exhibited relative concentration decrease in the first weeks for both species and  
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2363 sites, which was already observed during a litterbag experiment in a Vietnamese mangrove forest  
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2364 (Vinh et al., 2020). Mn is expected to be more leachable than other TM such as Fe and Zn as it is  
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2365 less able to bound with refractory molecules (Silva et al., 1998). Mn's soluble form in leaves ( $Mn^{2+}$ )  
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2366 (Joardar Mukhopadhyay and Sharma, 1991), easily solubilized during decomposition, may not  
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2367 oxidize to immobilized  $Mn^{4+}$  due to mangrove floor anoxia (Vinh et al., 2020).  
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3368 Loss of TM from litterfall appears element-dependent, influenced by oxidation state,  
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3369 molecular interactions, and porewater concentrations. Results of the present study also show  
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3370 differences in TM enrichment between stands and are discussed next.  
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#### 3371 4.3.3. Differences between stands due to trace metal bioavailability and species absorption capacity 46

3372 While most TM concentrations were generally higher in decomposed litterfall than senescent  
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3373 leaves, diverse trends were observed. At the urban site, peak TM concentrations relative to bulk leaf  
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3374 litter occurred at 28 days for *A. marina* and 56 days for *R. stylosa*. In contrast, at the control site,  
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3375 these peaks were reached at 56 or 72 days for *A. marina* and 72 days for *R. stylosa*. A recent study  
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3376 on the same sites has indicated higher TM bioavailability in the urban soils compared to the control  
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3377 soils (Robin et al., 2022). The lower bioavailability at the control site was linked to a greater  
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3378 proportion of TM in the refractory soil fraction (Robin et al., 2022). Elevated root and leaf  
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379 bioconcentration factors for most TM were significantly higher at the urban site, potentially leading  
380 to enhanced TM transfer from mangrove soil to litterfall (Marchand et al., 2016).

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

#### 394 4.4. Limitations of the experiment and models

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

## 406 5. Conclusion

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