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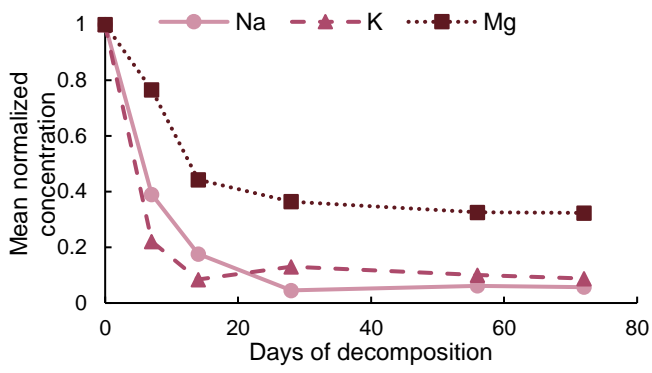
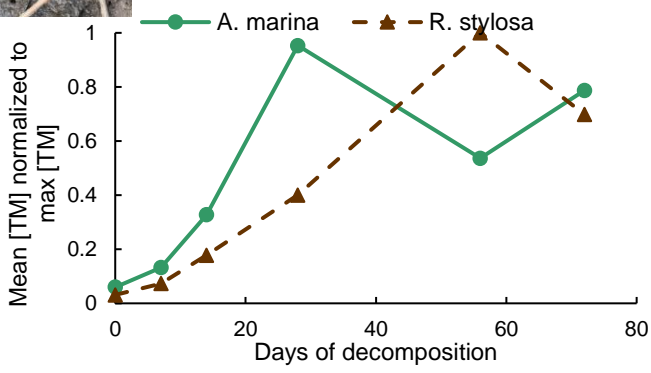
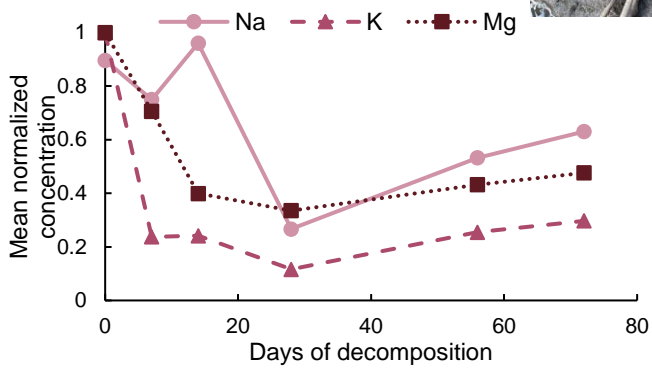
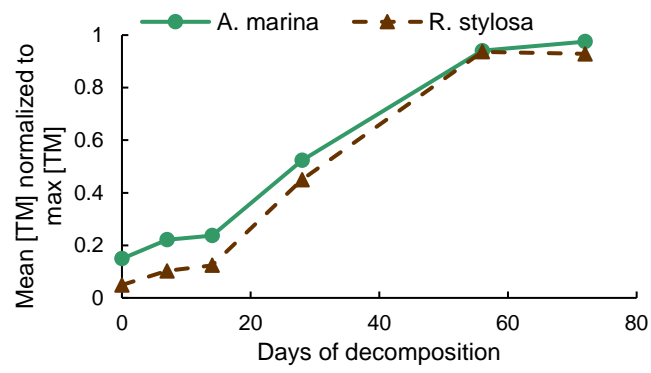
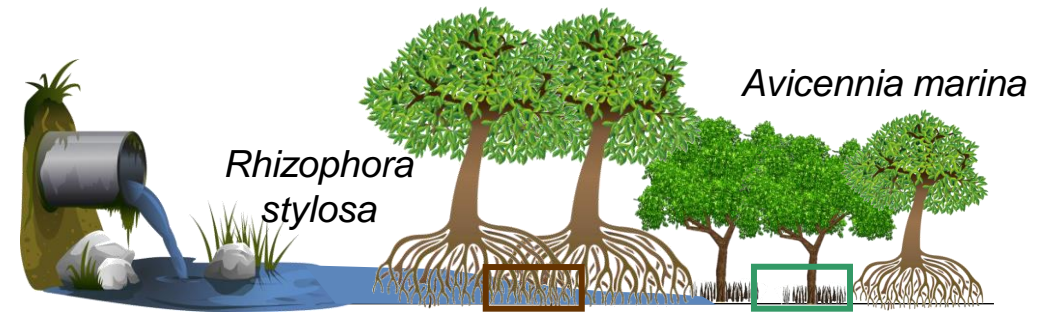
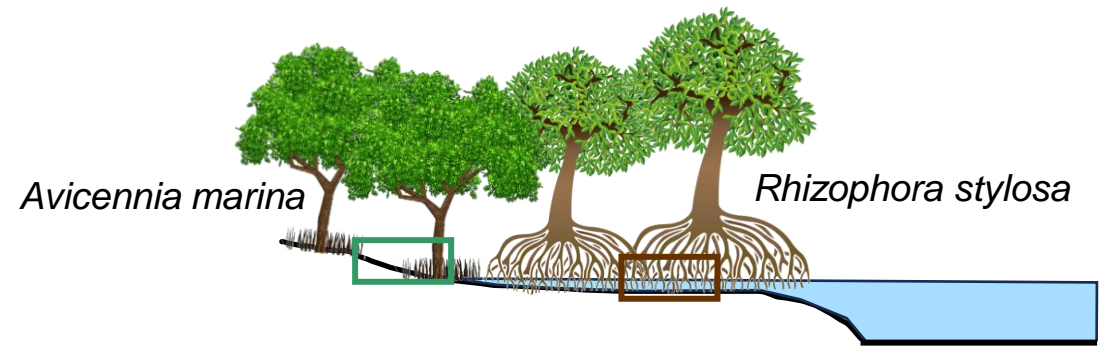


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Control mangrove forest

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).

2
3
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).
11
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
16
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
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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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35
3646 Litterfall organic matter (OM) can be consumed by autochthonous fauna and microorganisms
37
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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44
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
50
5153 during litter degradation (Bonanomi et al., 2010). On the other hand, the enrichment of major
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53
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.
9 1063

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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).
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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
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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
1292 runoff for over five decades. Employing a 72-day litterbag experiment, conducted on the main
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1493 mangrove species (*Rhizophora stylosa* and *Avicennia marina*), we hypothesize that the half-lives of
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1694 major elements usually leached early in the decomposition process such as K and Mg will be shorter
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1895 at the urban site than the control site due to the constantly flooding runoff. We also hypothesize that
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20
2196 greater TM enrichment in leaf litter will occur at the urban site due to their higher bioavailability in
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2397 the soil at this site.

2698 2. Materials and methods

2999 2.1. Study sites

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3200 The city of Dumbea in New Caledonia has had a population increase of 33% this last decade,
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3401 resulting in the development of urban infrastructures on the littoral zone (Insee, 2020). Two
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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*
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40
4104 and *A. marina*). One site was located in the Apogoti Bay (22°12'08"S, 166°26'20"E). This site has
42
4305 no direct anthropogenic inputs (limited housing density) into the mangrove forest, and it was selected
44
4506 as a control mangrove forest. Another site is the urban mangrove forest (22°12'39"S, 166°27'19"E)
46
4707 exposed to rainwater runoff from the upper allotment (mainly housing lots for 10 000 inhabitants),
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49
5008 which has been flowing into the mangrove forest for more than 50 years. At the control site, the two
51
5209 species develop in monospecific stands similar to most mangrove forests of the west coast, that is,
53
5410 *A. marina* landward and *R. stylosa* seaward. At the urban site, another *R. stylosa* stand develops
55
56
5711 more landward than the *A. marina* stand, at the entrance of the runoff, and its soil is always flooded
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5912 with the urban rainwater.

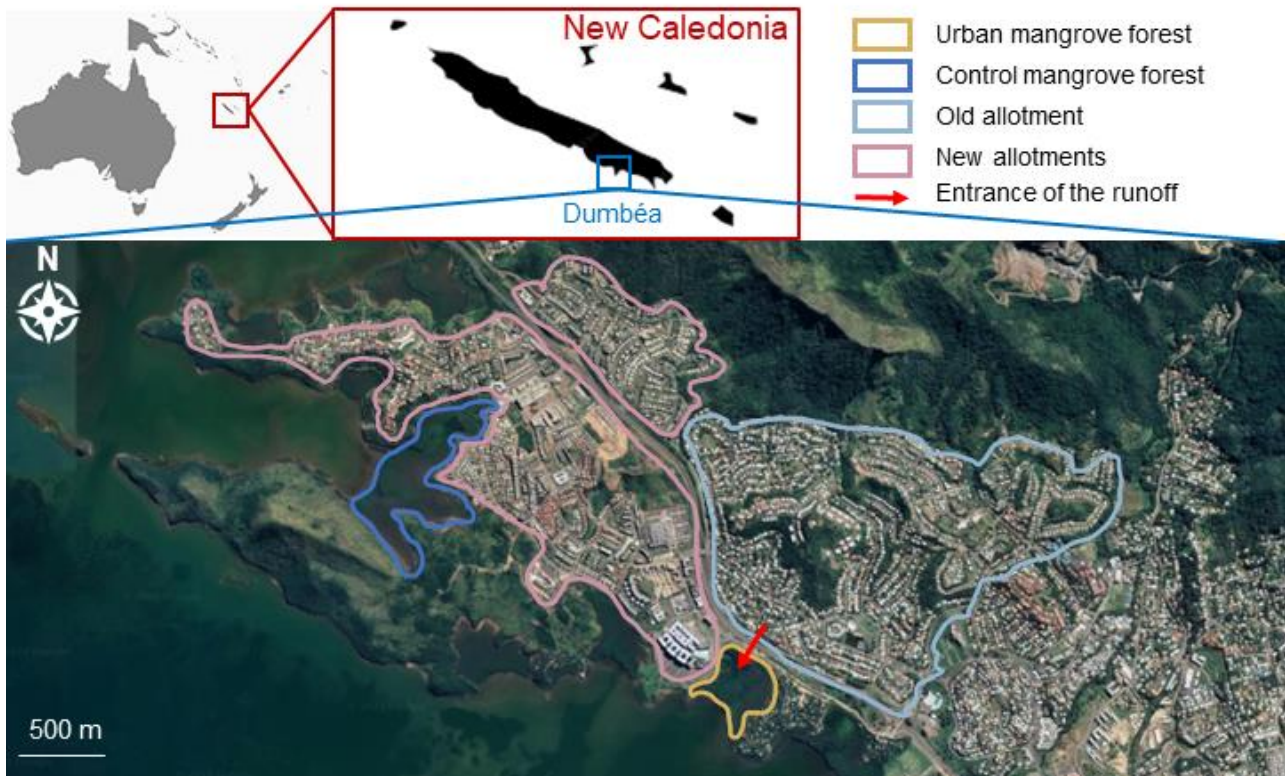


Figure 1. Control mangrove forest and urban mangrove forest in Dumbea city in New Caledonia with delimitation of the new and old allotments, and a red arrow to show the entrance and flow direction of the urban rainwater runoff (satellite image from Google Earth in 2022).

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

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% HNO₃ (Ajax Finechem) and 1 ml of 30% H₂O₂ (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⁻¹.

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⁻¹ of dry sample were converted to percentage relative to
156 t₀. 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
2
160 when possible:

$$161 \quad W_t = W_0 e^{-kt} \quad (2)$$

162 with W_t the element percentage at t , W_0 the initial element percentage (100 %), k the exponential
10
163 factor, and t the time in days. The half-lives ($t_{1/2}$) in days were obtained with the following equation:

$$164 \quad t_{1/2} = \ln(2) / k \quad (3)$$

165 2.4.2. Trace metals

166 To plot the changes in TM concentrations in the litterfall with degradation the measured TM
21
167 maximum concentration in $\mu\text{g g}^{-1}$ was set at 1.00 and the other values were calculated as follows:

$$168 \quad \text{TM concentration at } t_n = [\text{TM}]_n / [\text{TM}]_{\text{max}} \quad (4)$$

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
29
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
40
174 necessary to look at the differences in the chemical composition of leaves between species and
42
175 between fresh and senescent leaves. The concentrations of the measured elements in fresh and
44
176 senescent leaves of *A. marina* and *R. stylosa* are displayed in table 1. In all samples and of all TM,
47
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
49
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
54
180 the fresh leaves of *R. stylosa* than *A. marina*, while it was the opposite for the other TM. For the
56
181 senescent leaves, Mn and Ni were the only TM with higher concentrations in the *R. stylosa* trees.
58
182 Regarding the major elements, P and K had concentrations 5 and 3 times higher in the senescent
61

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
Major element concentrations (mg g⁻¹)						
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
Trace metal concentrations (μg g⁻¹)						
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 t₀ 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.

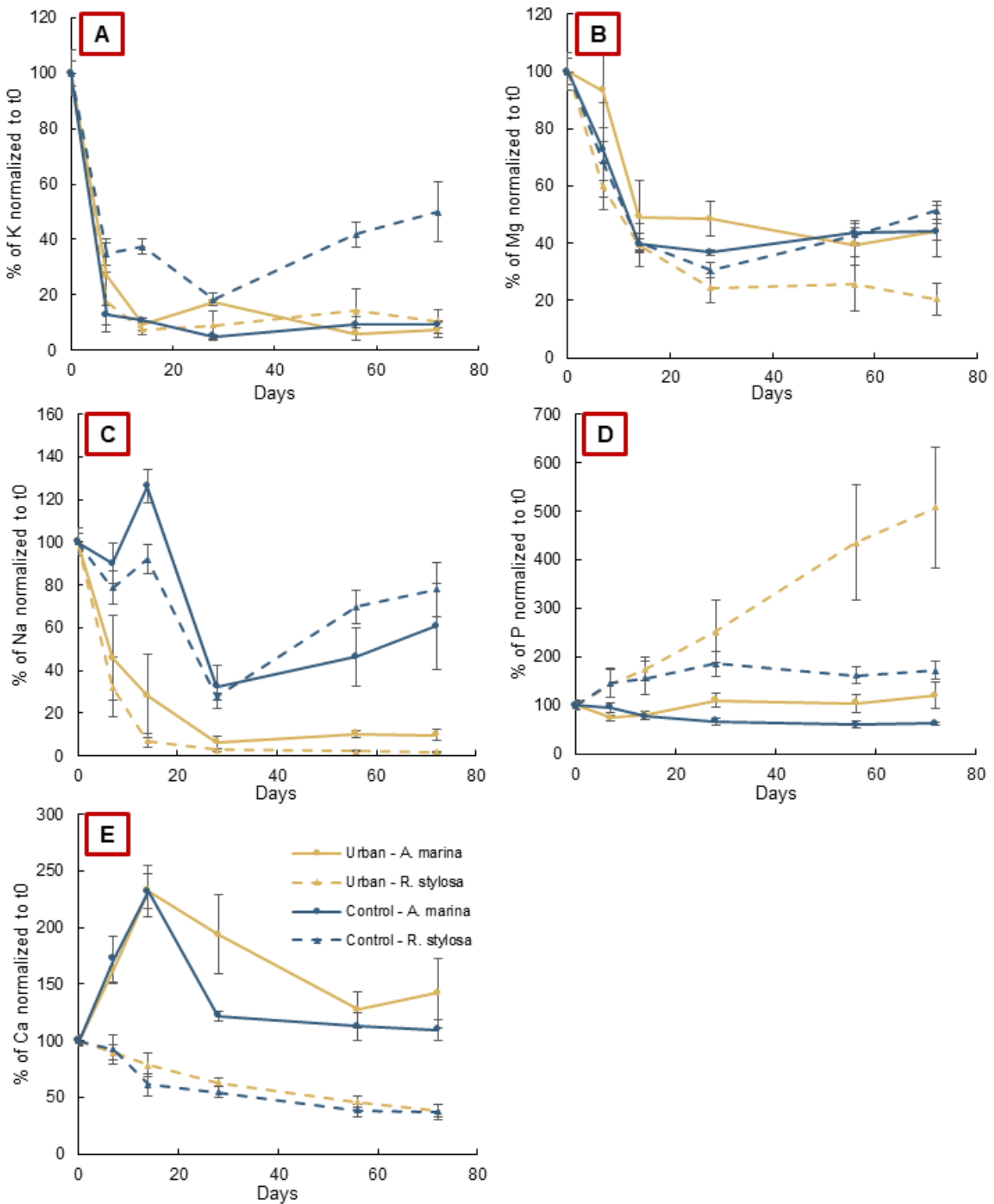


Figure 2. Mean percentage (SD as error bars) of (A) potassium, (B) magnesium, (C) sodium, (D) phosphorous, and (E) calcium in the leaf litter with decomposition relative to t0 at the urban and controls sites for *A. marina* and *R. stylosa*.

3.3. Changes in trace metal concentrations during leaf litter decomposition

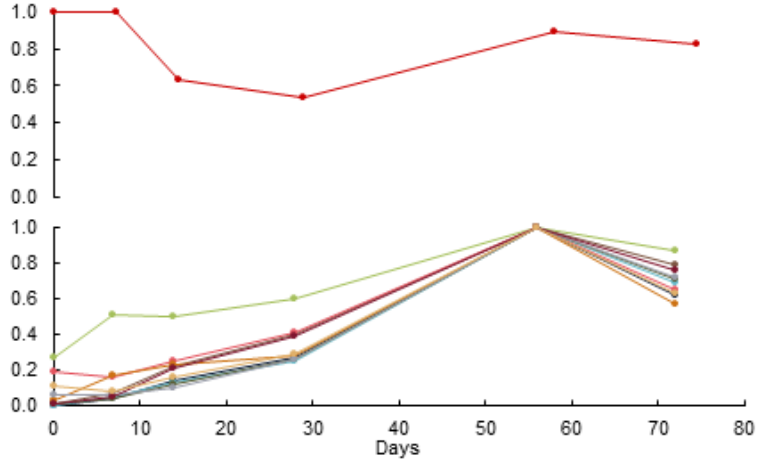
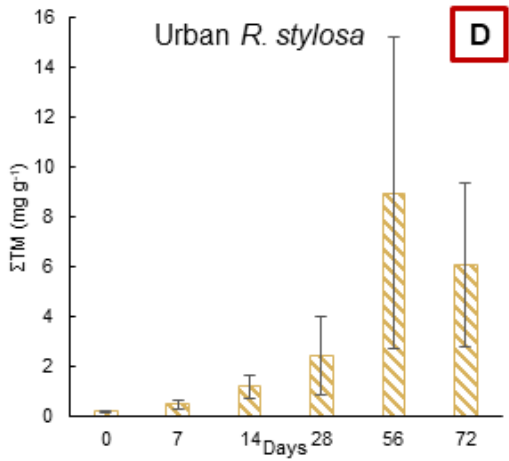
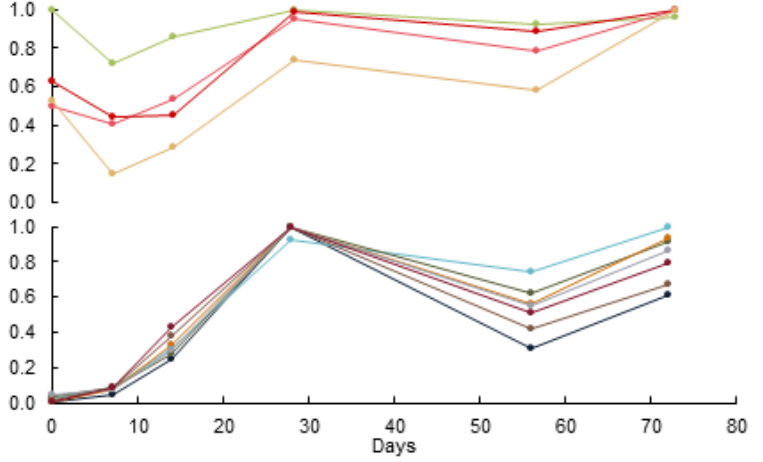
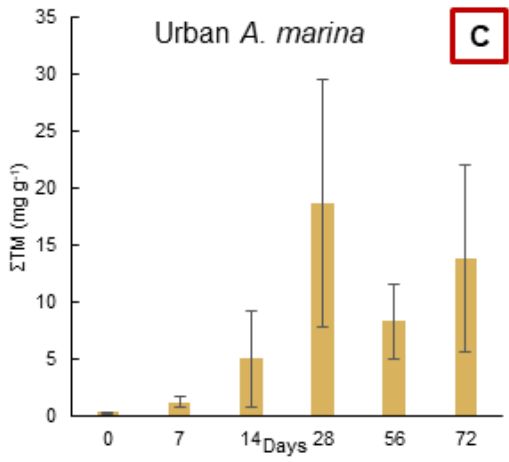
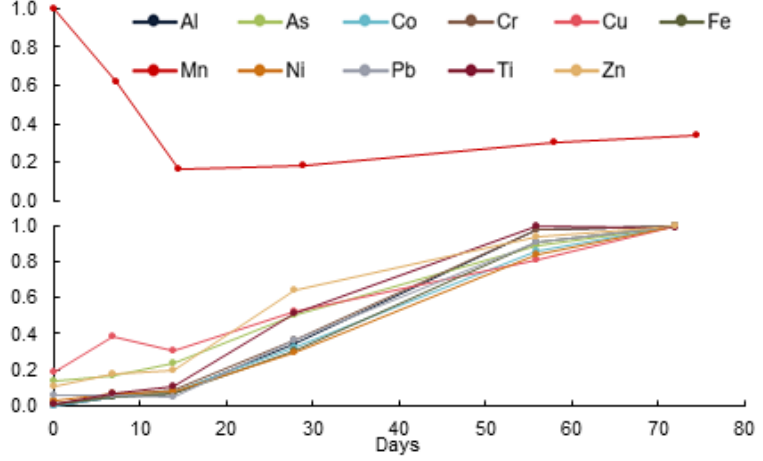
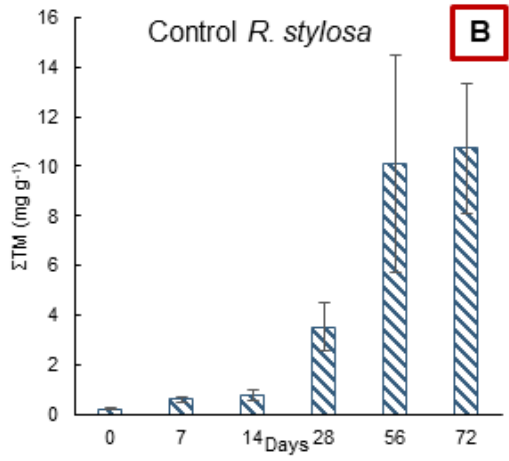
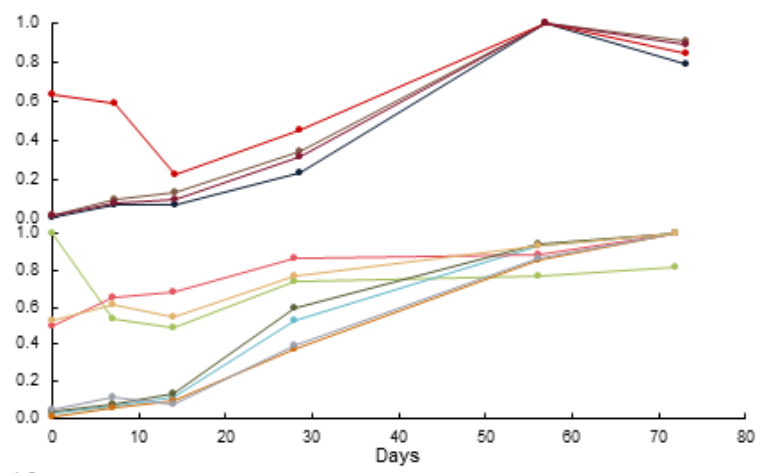
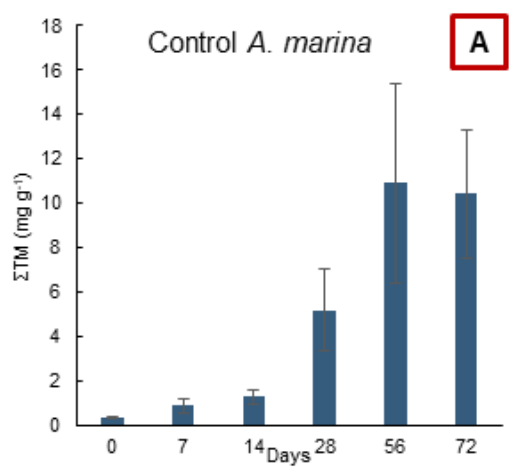
The sum of concentrations of TM in the leaf litter continuously increased with decomposition at the control site with concentrations 32 times and 48 times higher for *A. marina* and *R. stylosa*,

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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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).

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242 **Figure 3.** Mean sum of the measured trace metals in mg g⁻¹ (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⁺ and Na⁺
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⁺ 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⁻¹) (Robin et al., 2022) might promote this process in flooded *R. stylosa* areas. The high
305 salinity at the control site (35 g L⁻¹) 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
327 complexation of P with refractory molecules (Vinh et al., 2020) or floor-to-litter transfer via bacteria
328 or fungi (Lindahl et al., 2007). We suggest that the increase in P concentration in the litter of *R.*
329 *stylosa* might be due to water column P input, and therefore be dependent on the stand position in
330 the forest. Initial leaf composition may also contribute to the dynamic of P; *R. stylosa* had 5 times
331 less P in senescent leaves than *A. marina*, potentially offering more P capture sites.

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

338 *4.3. Trace metal dynamics during leaf litter decomposition*

339 *4.3.1. An expected trace metal enrichment during leaf litter decomposition*

340 All TM, except As and Mn, exhibited higher concentrations in the leaf litter after 72 days of
341 decomposition compared to senescent leaves, aligning with literature expectations (Ramos e Silva
342 et al., 2006; Rice and Windom, 1982; Vinh et al., 2020). The enrichment of TM during decomposition
343 relates to their exchange from forest soil or water column to leaf litter on the soil surface (Silva et
344 al., 1998). Microbial immobilization through TM uptake by microbial biomass or TM complex
345 formation with microbial-produced extracellular material can also enhance TM accumulation in leaf
346 litter (Rice and Windom, 1982). Additionally, a previous study on the same samples showed the
347 increase of N concentration in decomposing litterfall (Robin et al., 2024), which likely leads to TM
348 accumulation due to complex formation with N-rich compounds like proteins (Rice and Windom,
349 1982). Still, initial leaching phases were observed for some TM (As, Cu, Mn, Zn).

350 *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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362 Mn alone exhibited relative concentration decrease in the first weeks for both species and
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368 Loss of TM from litterfall appears element-dependent, influenced by oxidation state,
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371 4.3.3. Differences between stands due to trace metal bioavailability and species absorption capacity

372 While most TM concentrations were generally higher in decomposed litterfall than senescent
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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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