

Article

Fluxes of Cadmium, Chromium, and Lead Along with Throughfall and Stemflow Vary Among Different Types of Subtropical Forests

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Abstract: The interaction between forests and precipitation plays a crucial role in the material cycling of forest ecosystems. Atmospheric deposition and rainfall leaching promote the transfer of heavy metals to the forest floor, while canopy exchange may potentially slow this process. Therefore, studying heavy metal fluxes and their influencing factors, along with canopy rainfall partitioning, is essential for understanding forest material cycling. We conducted a year-long experiment to examine the dynamics of chromium (Cr), cadmium (Cd), and lead (Pb) concentrations and fluxes in four types of forests (*Cunninghamia lanceolata* plantations, *Castanopsis carlesii* plantations, *Cas. carlesii* natural forests, and *Cas. carlesii* secondary forests) located in the subtropical regions of southeast China. Results showed that (1) the annual mean concentrations of Cr, Cd, and Pb were 167.6, 13.8, and 6180.5 $\mu\text{g L}^{-1}$ in the throughfall and 204.7, 28.4, and 2251.1 $\mu\text{g L}^{-1}$ in the stemflow, respectively, and the annual fluxes of Cr, Cd, and Pb through throughfall were 29.3, 2.4, and 847.7 g ha^{-1} , respectively, and were 1.7, 0.2, and 12.7 g ha^{-1} through stemflow, respectively; (2) the concentrations of these heavy metals associated with throughfall did not vary between forest types, but their fluxes were highest in *Cas. carlesii* natural forests; (3) Cr concentration and flux were higher during the rainy than dry seasons, while Cd and Pb concentrations and fluxes showed an opposite trend. Overall, our results indicate that the fluxes of heavy metals along with rainfall partitioning were highest in natural forests and are primarily controlled by meteorological factors, indicating that the conversion of natural forests to other forest types will substantially change the fluxes of heavy metals along with hydrological processes. These results will contribute to a better understanding of the natural fluxes of heavy metals in forest ecosystems and are valuable for sustainable forest management, particularly in the context of forest type transformation.

Keywords: canopy exchange; heavy metal; hydrological process; stemflow; throughfall



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1. Introduction

Rapid industrialization and urbanization have resulted in higher emissions of pollutants, particularly heavy metals like chromium (Cr). The annual increase in Cr application and usage contributes to its rising concentration in the ecological environment [1,2]. Studies on soil, plants, lake sediments, aerosols, and other media indicate that the heavy metals

most likely linked to human activities are cadmium (Cd) and lead (Pb) [3]. Cr, Cd, and Pb are all harmful metals included in China's heavy metal pollution prevention plan, characterized by high toxicity, solubility, and mobility. The dynamics of heavy metals will significantly affect ecosystem health, functioning, and ultimately human health [4–6]. Heavy metals exhibit bioaccumulation properties, posing toxic effects on organisms when their concentrations exceed certain thresholds in the environment [4,7,8], and the input of heavy metals into forest ecosystems can lead to reduced forest productivity and loss of biodiversity, thus weakening the ecological services provided by forests [9]. Heavy metals adsorbed onto surface dust are highly susceptible to environmental transport and redistribution via runoff generated by rainfall [10,11].

Precipitation and its redistribution processes play a crucial role in material cycling in forest ecosystems [12,13]. When precipitation interacts with the forest canopy, a portion is intercepted, while the rest penetrates through the canopy, forming throughfall, or flows down branches and tree trunks as stemflow [14]. These precipitation-partitioning processes introduce a significant amount of substances into the forest ecosystem, accompanied by changes in element concentrations and fluxes [15,16]. The relationship between precipitation and forests has attracted the attention of experts in forestry, ecology, and other fields, becoming a hot topic in current research [17]. Research has shown that both elevation and geographic location significantly influence heavy metal pollution, with precipitation playing a crucial role. For instance, in high-elevation areas, such as the eastern Tibetan Plateau, the terrain's effect on rainfall results in much higher concentrations of heavy metals in atmospheric precipitation on the windward slopes compared to the leeward slopes [18]. In subtropical regions, abundant rainfall contributes to regional variations in heavy metal pollution across China. Northern cities in north China tend to have higher levels of pollution, while southern and certain southwestern areas experience lower pollution levels [19]. Therefore, rain partitioning emerges as a crucial pathway for material cycling in forest ecosystems. However, available studies mainly focus on the ecological impacts of heavy metals [9,20,21] and hardly notice the dynamics of heavy metals along with ecological processes such as rainfall partitioning, which have limited our understanding on the cycling of heavy metals in forest ecosystems under future environmental change scenarios.

The distribution of heavy metals in forest precipitation is the result of several interacting processes. These include atmospheric deposition, the leaching of canopy dry deposition by rainfall, and the absorption or ion exchange of specific elements by plants [22,23]. The dynamics of heavy metal fluxes along with rainfall partitioning would vary among different types of forests, because the rainfall partition pattern and the filter capacity of the canopies on rainfall pH and elements have been found to be different among forest types [24–26]. Differences in canopy structure among different forest types would be the main reason driving forest type effects on heavy metal dynamics [27]. In addition, seasonal rainfall patterns can lead to seasonal variations in heavy metal flux [28], rainfall amount and intensity can influence atmospheric substance leaching, with insufficient rainfall potentially leading to incomplete leaching, while excessive rainfall can dilute ion concentrations [29–31]. Both rainfall characteristics and the interception of deposited substances by the canopy exhibit seasonal fluctuations, which can cause variations in heavy metal concentrations and fluxes along with rainfall partitioning [32]. Furthermore, due to the intercepting effects of the forest canopy, the concentrations of elements and pH between stemflow and throughfall generally showed opposite patterns [24,26]. However, how forest type and associated variables may affect the dynamics of heavy metal concentrations and fluxes along with rainfall partitioning have rarely been assessed.

Castanopsis carlesii and *Cunninghamia lanceolata* are dominant species in subtropical regions of east China, playing crucial roles in carbon sequestration, oxygen release, water

conservation, and maintaining biodiversity [33,34]. They also have significant economic value. While studies have focused on their biomass and seed banks, research on forest hydrology has mainly concentrated on rainfall partitioning patterns and the water-holding characteristics of litter [35–38]. Here, we conducted a one-year field experiment to assess the dynamics of Cr, Cd, and Pb among different forest types in a subtropical region of southeast China. We chose four common forest types, namely *Cun. lanceolata* plantations (CLPs), *Cas. carlesii* plantations (CCPs), *Cas. carlesii* natural forests (NFs), and *Cas. Carlesii* secondary forests (SFs), of the research region that have obvious variations in canopy structures and evaluated both the concentrations and fluxes of Cr, Cd, and Pb in stemflow and throughfall. We hypothesized that the concentrations and fluxes of Cr, Cd, and Pb will vary across these forest types, with contrasting patterns between throughfall and stemflow.

2. Materials and Methods

2.1. Study Area

This study was carried out at the Fujian Sanming Forest Ecosystem National Observation and Research Station (26°19' N, 117°36' E). The study area has a subtropical monsoon climate with warm and humid weather, different seasons, and abundant rainfall (average 1610 mm year⁻¹). The terrain consists mainly of low mountains and hills, with an average elevation of 300 m. The dominant forest in this area is subtropical evergreen broad-leaved forest. In 1976, the primary evergreen broad-leaved forests in the region were cut down. Subsequently, natural succession led to their regeneration into natural forests, which were then selectively harvested and artificially promoted, resulting in the formation of secondary forests. At the end of 2011, these secondary forests were transformed into plantations, following established management practices for plantation forests [39,40]. The dominant species of natural forests, secondary forests, and plantations are the native tree species *Cas. carlesii*. More detailed information on the different types of forests was recorded in previous studies [26,41] (Figures 1 and 2).

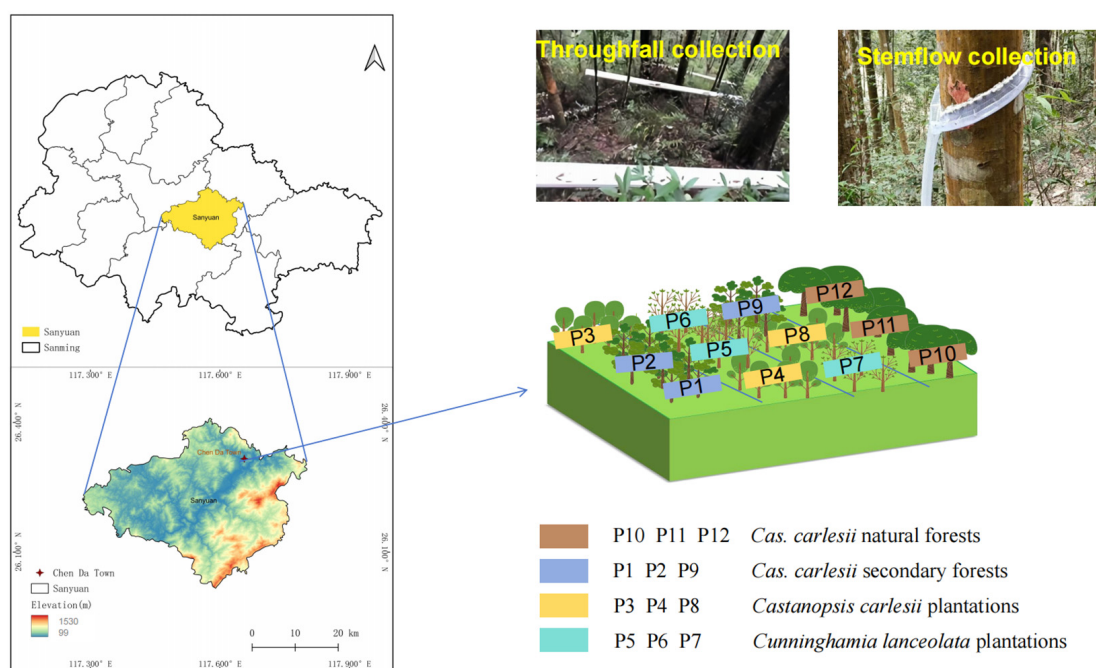


Figure 1. Experimental site location map and sample plot setup.



Figure 2. Comparison of bark and leaf morphology between *Castanopsis carlesii* (left) and *Cunninghamia lanceolata* (right). Photo credit: Wenfeng Jiang.

2.2. Field Experimental Design

Through field measurements, we established three plots (20×20 m) in each of the four forest types. These plots were located in areas with similar positions, slopes, and characteristics to set up the experimental devices. Tipping-bucket rainfall sensors (with a rainwater collection area diameter of 20 cm and a bucket accuracy of 0.20 mm tip^{-1}) were installed in the open space adjacent to the study plots to measure atmospheric precipitation, and 400×14 cm PVC troughs were placed along contour lines, elevated 1 m above the ground. The troughs were securely attached at a 5° incline angle, facilitating the collection and documentation of the fall using 20 L polyethylene containers. Also, we recorded information on trees within the sample plots and observed significant variation in diameter at breast height (DBH) within *Cas. carlesii* natural forests. Therefore, we categorized the trees into five diameter classes (8–12 cm, 12–16 cm, 16–20 cm, 20–24 cm, and >24 cm) and randomly chose one tree of each diameter class within each plot for stemflow measurement, resulting in a total of five trees for each plot. For the other three forest types, we randomly chose five standard trees; we then cut the PVC tubes and wrapped them spirally around the tree trunk at DBH, with the end of the tube connected to a tipping bucket rain gauge to collect stemflow. During each sampling, manually remove debris and impurities, such as animals, from the equipment. Then, use a 500 mL plastic bottle that has been rinsed with deionized water and subsequently rinsed 2 to 3 times with the water sample for collection. The collected water samples were first filtered with a $0.45 \mu\text{m}$ filter membrane, and then the water samples were stored in a refrigerator at 4°C . The collected water samples were filtered through a $0.45 \mu\text{m}$ polypropylene membrane, and the concentrations of Cr, Cd, and Pb were quantified using inductively coupled plasma optical emission spectrometry (ICP-OES, Horiba JY, Paris, France). The pH of the water samples was measured using a pH meter (FE28-Standard, Mettler Toledo, Zurich, Switzerland).

Sampling was conducted from 2 April 2021 to 28 March 2022, with a total of 36 sampling periods. During the dry season (September 2021 to February 2022), samples were collected 10 times, while in the rainy season (April to August 2021 and March 2022), samples were collected 26 times.

2.3. Meteorological Observation

Temperature and humidity sensors (MicroLite5032-RH, Beijing Eastsummit, Beijing, China) and a wind speed transmitter (RS485) were installed at a height of 1.5 m in both open areas outside the forest and within the forest interior (one for each forest type). These instruments continuously monitored temperature, humidity, and wind speed, recording data every 30 min. The temperature sensor had an accuracy of ± 0.04 °C, the humidity sensor had an accuracy of $\pm 0.5\%$, and the wind speed transmitter had an accuracy of ± 0.3 m/s.

2.4. Statistical Analysis

We calculated throughfall (TF, mm) as follows:

$$TF = \frac{V}{A} \quad (1)$$

where V is the amount of throughfall (L), and A is the receiving area of throughfall collector (m^2).

The stemflow (SF, mm) was calculated as follows:

$$SF = \sum_{i=1}^n \frac{C_i \times M_i}{S_p \times 1000} \quad (2)$$

where n is tree trunk diameter series; C_i is the amount of stemflow observed (mL) of the diameter class i ; M_i is the number of trees of the diameter class; and S_p is the sample area (400 m^2).

The calculation formula for heavy metal flux in throughfall and stemflow is as follows [42]:

$$F_j = C_{jk} \times V_{jk} / 100 \quad (3)$$

where F_j is the fluxes of heavy metals migrating along hydrological path j (g/ha), C_{jk} is the concentration of the heavy metal concentration ($\mu\text{g L}^{-1}$) of the hydrological path j in the k th sampling period, and V_{jk} is the equivalent precipitation (mm) of the corresponding hydrological path.

The tipping-bucket rainfall sensor was calibrated using the standard procedure outlined in GB/T 11832-2002 [43]. The temperature and humidity sensors were calibrated according to the procedures described in DB14/T 1730-2018 [44]. The wind speed transmitter was calibrated following the standard procedure detailed in RS485. The ICP-OES instrument used in this study was operated according to the guidelines specified in ISO 11885:2007 [45] for the analysis of multiple elements in water samples. Calibration was performed using certified reference materials, and quality control was maintained by including blank samples and spiked standards.

To investigate the impacts of forest type on heavy metal concentrations, we performed a one-way analysis of variance (ANOVA) to compare and analyze the differences in heavy metal concentrations and fluxes among different forest types, as well as the differences in heavy metal concentrations across different rainfall intensities. To further examine the influence of seasons and forest types, a two-way ANOVA was used to analyze the heavy metal fluxes in throughfall and stemflow. Specifically, Cr, Cd, and Pb concentrations were analyzed in different forest types and during the dry and rainy seasons. In addition, we used a linear mixed model to assess the influence of environmental factors (atmospheric wind speed, atmospheric temperature, humidity, air pressure, etc.) on heavy metals. To evaluate the impacts of rainfall intensity on Cr, Cd, and Pb fluxes, we classified the intensity into six levels according to the China Meteorological Administration, namely light (0–10 mm), moderate (10–25 mm), heavy (25–50 mm), rainstorm (50–100 mm), severe

rainstorm (100–250 mm), and torrential rain (>250 mm), and its effects were evaluated within each forest type. Each sampling period is defined as the duration between the completion of the previous sampling and the end of the subsequent sampling, inclusive of both continuous rainy days and rain-free intervals (defined as days with rainfall less than 0.1 mm). Rainfall intensity (mm h^{-1}) is calculated as the total rainfall during each sampling period divided by its duration of rainfall. All statistical analyses were performed with R 4.4.1. Before statistical analysis, we checked normality and homogeneity of the data and logarithmically transformed them where necessary.

3. Results

3.1. Heavy Metal Concentrations

We observed an opposite pattern in the concentrations of heavy metals between throughfall and stemflow regardless of forest types (Figure 3 and Table 1), throughfall is significantly influenced by seasons, while stemflow is notably affected by forest types. The annual concentrations of Cr, Cd, and Pb were 167.6 , 13.8 , and $6180.5 \mu\text{g L}^{-1}$ in the throughfall and were 204.7 , 28.4 , and $2251.1 \mu\text{g L}^{-1}$ in stemflow, respectively. Throughfall had higher Cr concentration during rainy than dry seasons (Figure 4). Forest type did not affect heavy metal concentrations in throughfall, except for the concentration of Pb during the dry season, but showed significant impacts on Cr, Cd, and Pb concentrations in stemflow in both dry and rainy seasons, with higher concentrations observed in *Cun. lanceolata* plantations.

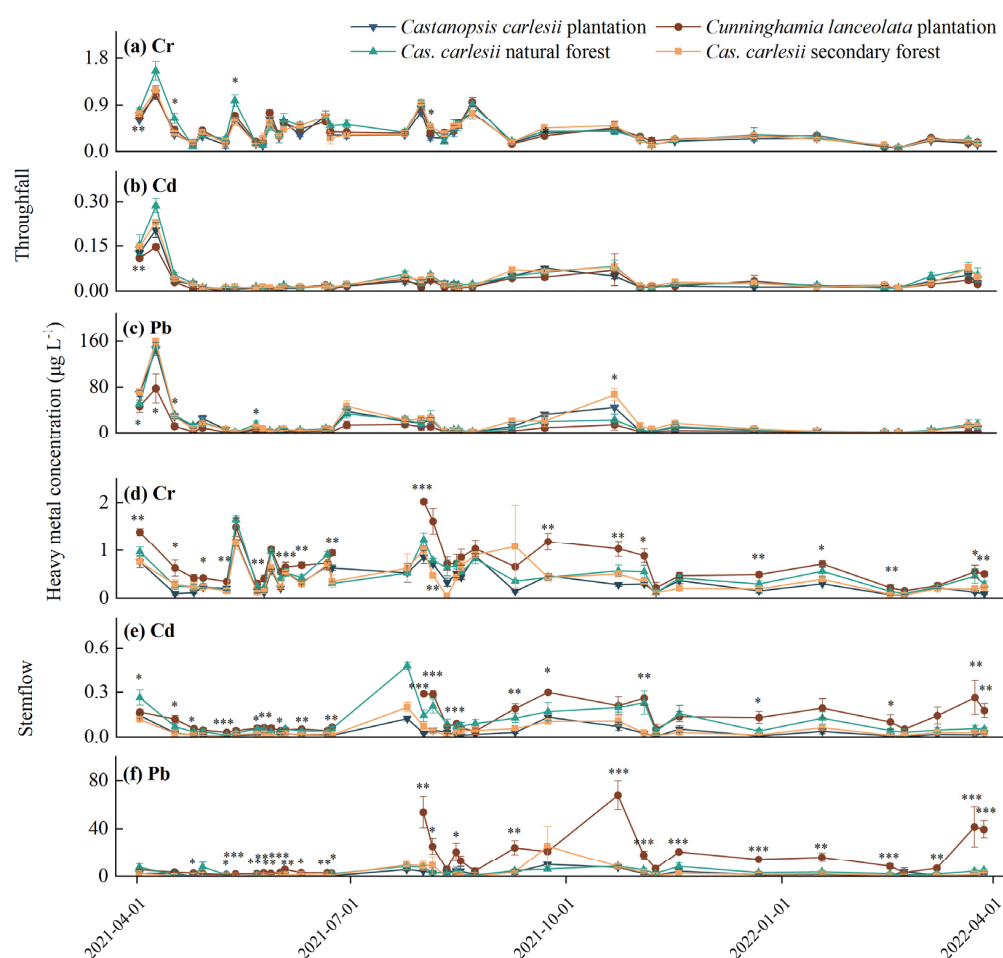


Figure 3. Dynamics of heavy metal concentrations in throughfall and stemflow in different types of forests. Values are means with standard error (SE), and asterisks indicate significant differences among forest types at * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. Chromium: Cr, cadmium: Cd, lead: Pb.

Table 1. Comparison of Cr, Cd, and Pb concentrations ($\mu\text{g L}^{-1}$) in throughfall and stemflow.

Composition of Rainfall	Forest Type	Cr			Cd			Pb		
		Mean $\pm$ SE	CV	<i>p</i> Value	Mean $\pm$ SE	CV	<i>p</i> Value	Mean $\pm$ SE	CV	<i>p</i> Value
Throughfall	CCP	0.35 $\pm$ 0.02	0.71	0.264	0.03 $\pm$ 0.00	1.35	0.101	15.8 $\pm$ 2.63	1.73	0.018
	CLP	0.39 $\pm$ 0.02	0.64		0.03 $\pm$ 0.01	1.29		7.73 $\pm$ 1.57	2.12	
	NF	0.42 $\pm$ 0.03	0.77		0.04 $\pm$ 0.00	1.40		15.4 $\pm$ 2.59	1.74	
	SF	0.38 $\pm$ 0.02	0.67		0.04 $\pm$ 0.00	1.23		18.2 $\pm$ 2.86	1.63	
Stemflow	CCP	0.37 $\pm$ 0.03	0.79	<0.001	0.03 $\pm$ 0.00	1.28	<0.001	2.38 $\pm$ 0.29	1.21	<0.001
	CLP	0.74 $\pm$ 0.05	0.62		0.12 $\pm$ 0.01	0.79		13.4 $\pm$ 0.68	1.24	
	NF	0.51 $\pm$ 0.04	0.70		0.1 $\pm$ 0.01	1.06		3.37 $\pm$ 0.33	0.97	
	SF	0.42 $\pm$ 0.55	0.92		0.04 $\pm$ 0.00	1.19		2.94 $\pm$ 0.68	2.30	

CCP: *Castanopsis carlesii* plantation, CLP: *Cunninghamia lanceolata* plantation, NF: *Castanopsis carlesii* natural forest, SF: *Castanopsis carlesii* secondary forest. Values are means with standard errors (SE), and CV represents the coefficient of variation value. Differences among forest types are indicated by *p*-values.

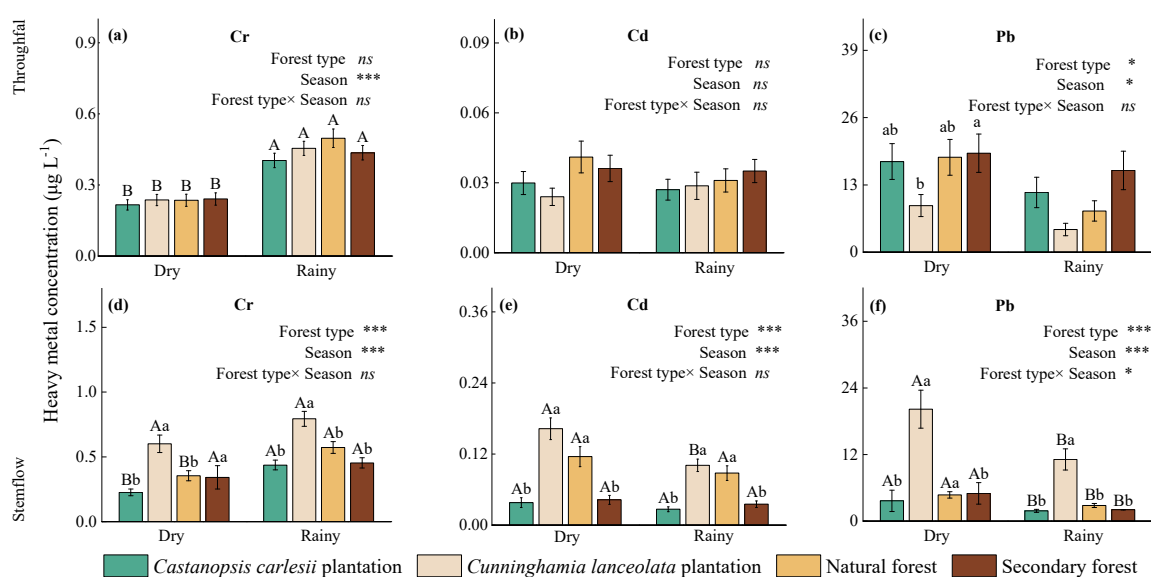


Figure 4. Heavy metal concentrations in throughfall and stemflow among different seasons and forest types. Values are means with standard error (SE). Different capital letters represent the statistical differences between dry season and rainy season, and different lowercase letters indicate differences between forest types. * $p < 0.05$, *** $p < 0.001$. ns indicates no statistically significant difference. Chromium: Cr, cadmium: Cd, lead: Pb.

3.2. Fluxes of Heavy Metals

The dynamics of heavy metal fluxes showed a similar pattern with their concentrations, and the annual fluxes of Cr, Cd, and Pb through the throughfall were 29.3, 2.4, and 847.7 g ha^{-1} , respectively, and through the stemflow were 1.7, 0.2, and 12.7 g ha^{-1} , respectively (Figure 5). The fluxes of heavy metal in the *Cas. carlesii* natural forests showed larger variations compared with those in other forest types and showed lowest variations in the *Cas. carlesii* plantations. Similar with heavy metal concentrations, the fluxes of Cr and Cd in throughfall were not affected by forest type, while throughfall Pb flux significantly varied among forest types in both dry and rainy seasons, with the lowest flux observed in *Cun. lanceolata* plantations (Figure 6). Forest type significantly affected the fluxes of stemflow Cr, Cd, and Pb in both dry and rainy season, with the highest values observed in *Cas. carlesii* natural forests.

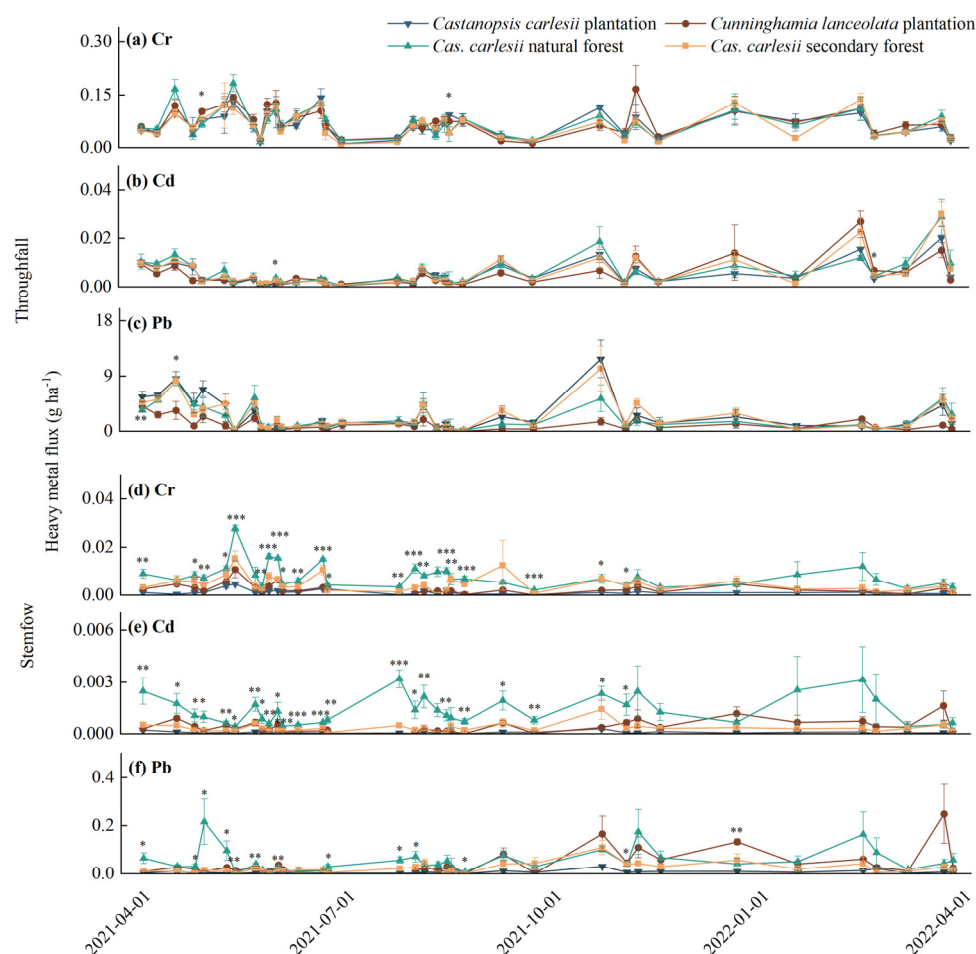


Figure 5. Dynamics of heavy metal fluxes in throughfall and stemflow in different forest types. Values are means with standard error (SE), and asterisks indicate significant differences among forest types at * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. Chromium: Cr, cadmium: Cd, lead: Pb.

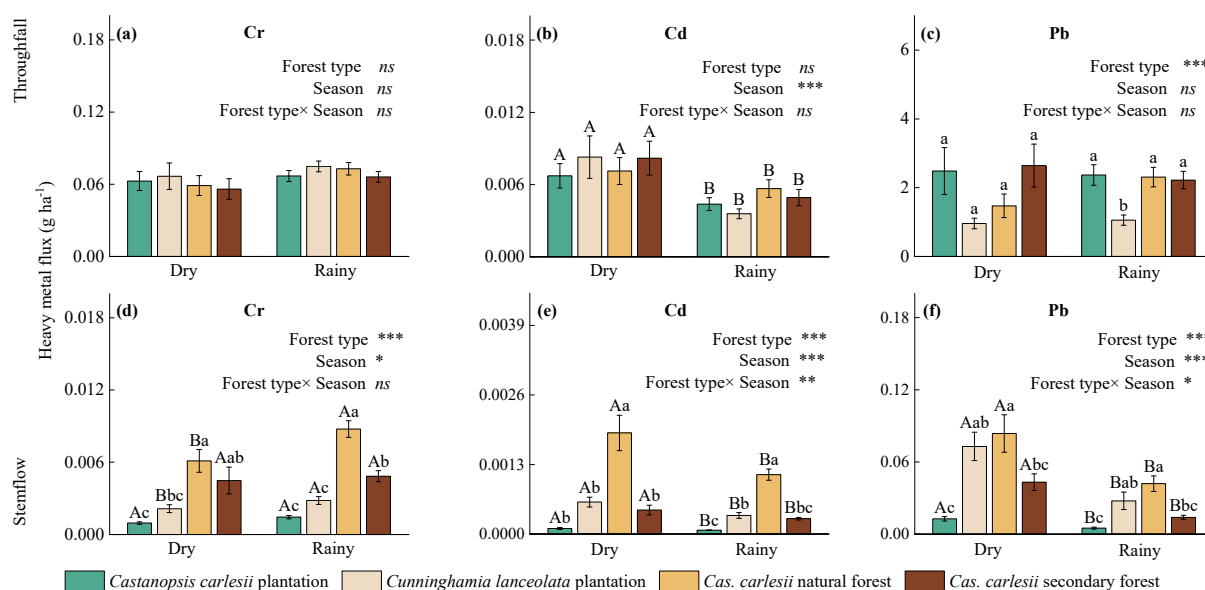


Figure 6. Heavy metal fluxes in throughfall and stemflow in different seasons and forest types. Values are means with standard error (SE). Different capital letters represent the statistical differences between dry season and rainy season, and different lowercase letters indicate differences between forest types. * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. ns indicates no statistically significant difference. Chromium: Cr, cadmium: Cd, lead: Pb.

3.3. Factors Affecting the Concentrations and Fluxes of Heavy Metals

Climate showed strong impacts of the concentrations and fluxes of heavy metals in both throughfall and stemflow. Specifically, the fluxes of Cr, Cd, and Pb fluxes in throughfall showed a significant influence with rainfall intensity, while the Pb flux in throughfall was lowest, but the rainfall intensity only affected the Pb flux along with stemflow in *Cas. carlessii* natural forest (Figure 7). Understory wind speed negatively affected throughfall Cr flux, while both atmospheric wind speed and understory wind speed positively affected the throughfall Cd and Pb fluxes (Table 2). Atmospheric air temperature, understory temperature, atmospheric air relative moisture, and understory relative moisture all showed significant impacts on the concentrations and fluxes of heavy metal fluxes in throughfall and stemflow, but their effects varied among heavy metals. The rainfall pH positively affected the throughfall and stemflow heavy metal fluxes, except stemflow Cd flux. The rainfall duration positively influenced the heavy metal fluxes, except stemflow Cr flux.

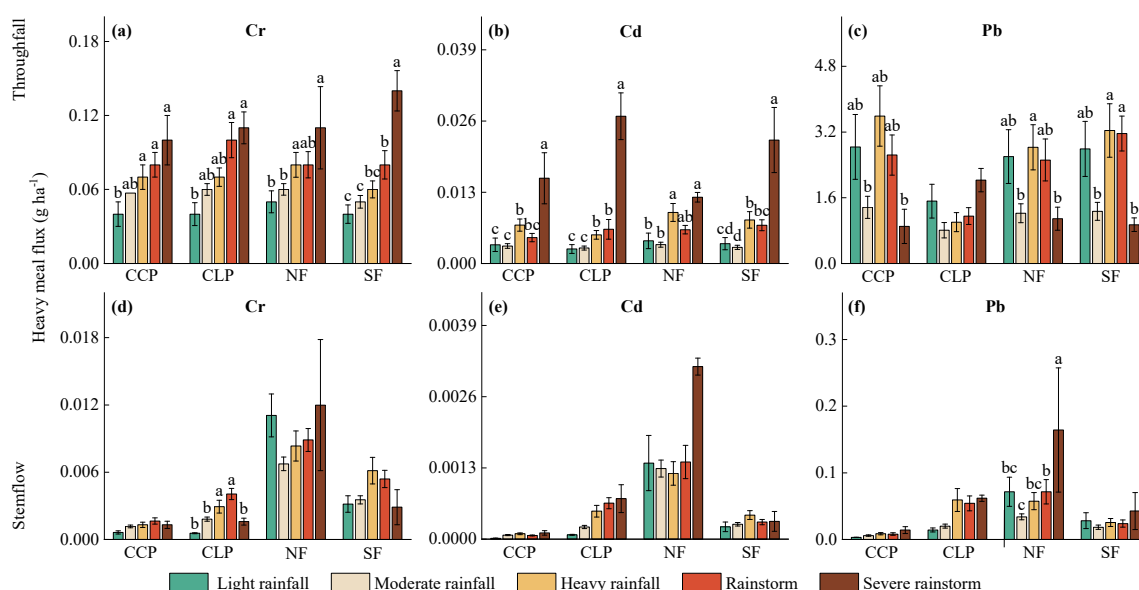


Figure 7. Heavy metal fluxes in throughfall and stemflow across different types of forests under varying rainfall intensities. CCP: *Castanopsis carlessii* plantation, CLP: *Cunninghamia lanceolata* plantation, NF: *Castanopsis carlessii* natural forest, SF: secondary forest of *Castanopsis carlessii* secondary forest. Values are means with standard error (SE), and different lowercase letters indicate differences between rainfall intensities. Chromium: Cr, cadmium: Cd, lead: Pb.

Table 2. Effects of environment factors on concentrations and fluxes of Cr, Cd, and Pb in throughfall and stemflow, as assessed using linear fixed-effect mode.

Influence Factors	Throughfall						Stemflow					
	Cr Conc	Cd Conc	Pb Conc	Cr Flux	Cd Flux	Pb Flux	Cr Conc	Cd Conc	Pb Conc	Cr Flux	Cd Flux	Pb Flux
Atmospheric wind speed (m s^{-1})	1.096 **	0.423 ***	11.218	−0.045	0.061 ***	4.439 ***	1.754 ***	0.724 ***	7.157 ***	0.002	0.001	0.050
Understory wind speed (m s^{-1})	0.104	0.079 **	3.289 ***	−0.051	0.009 *	1.067 *	0.631 ***	0.257 ***	2.114 **	−0.003	0.001	−0.040
Atmospheric temperature ($^{\circ}\text{C}$)	0.243 ***	0.008	0.858 ***	0.001	− 0.005 ***	0.061	0.330 ***	0.027 **	0.097	0.001 *	− 0.001 *	− 0.021 **
Understory temperature ($^{\circ}\text{C}$)	0.243 ***	0.007	0.796 ***	−0.001	− 0.005 ***	0.029	0.322 ***	0.027 **	0.125	0.001 *	− 0.001 *	− 0.020 **
Atmospheric relative moisture (%)	0.235 ***	−0.015	−0.236	0.031 *	− 0.007 ***	− 0.492 *	0.309 ***	0.003	−0.601 *	0.004 **	−0.001	−0.024
Understory relative moisture (%)	0.278 ***	0.012	1.000 ***	0.015 *	− 0.005 ***	0.175	0.327 ***	0.019	−0.162	0.002 ***	− 0.001 *	− 0.025 ***
pH of rainfall	−0.009	− 0.066 ***	− 2.485 ***	− 0.050 ***	0.012 ***	− 0.868 ***	− 0.489 ***	0.019	1.596 ***	− 0.008 ***	0.001	0.054 **
pH of throughfall/stemflow	− 0.646 ***	− 0.121 ***	− 3.404 ***	− 0.058 *	0.011 *	−0.714	− 0.712 ***	− 0.082 **	−0.542	−0.001	0.001	0.034
Rainfall intensity (mm h^{-1})	− 1.198 ***	− 0.023 ***	− 0.568 ***	0.009	− 0.004 ***	− 0.262 ***	0.100 ***	−0.015	− 0.485 ***	0.002 ***	− 0.001 *	− 0.022 ***
Rainfall duration (h)	0.020	− 0.023 ***	−0.123	0.005 *	0.002 ***	0.096 **	− 0.099 ***	0.001	0.139 **	−0.001	0.001 **	0.012 ***
Tree high (m)	−0.001	0.003	0.208	−0.004	0.001	0.056	−0.315	−0.039	−0.026	0.001	−0.001	−0.003
DHB (cm)	0.001	0.001	0.058	−0.003	0.001	−0.032	−0.004	−0.021	−0.077	0.001	−0.001	−0.006
Stand density (stem ha^{-1})	−0.009	0.001	0.151	−0.001	0.001	0.100	−0.077	−0.026	−0.383	−0.001	−0.001	−0.010

Atmospheric wind speed and temperature refer to the wind speed and temperature measured on open ground outside the forest. The effects of each variable were assessed individually, estimates of the slope were reported, and bold indicates significant effects. * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. Bold indicates significant effects. Conc represents concentration.

4. Discussion

Our results showed that the heavy metal concentrations and fluxes in throughfall were higher than those in stemflow. Comparing the average concentrations and fluxes of Cr, Cd, and Pb in this study with those in stemflow and throughfall from studies in the US, Spain, and other regions in China [23,46–49], it was found that both the concentrations and fluxes of heavy metals were higher in this study. The elements carried by rainfall undergo changes as they pass through the forest canopy [50,51]. Previous studies have shown higher nutrient concentrations in stemflow than in throughfall [32,52], but the opposite result was observed in this study. This discrepancy may be due to the fact that Cr, Cd, and Pb are not essential nutrients for plant growth, and trees differ in their ability to absorb and utilize these metals. Moreover, the differences in the concentrations and fluxes of Cr, Cd, and Pb between throughfall and stemflow varied across forest types and seasons. There are distinct differences in canopy structure and morphological characteristics between different types of forests. For example, the *Cas. carlesii* is part of the evergreen broadleaf forest, with a canopy that has a gradually pointed top and occasionally a slightly tilted base. Its large, elliptical leaves facilitate the collection of atmospheric deposition and direct the water accumulated on branches towards the trunk. In contrast, *Cun. lanceolata* belongs to the coniferous forest, with a conical or pyramidal crown. The leaves radiate from the main branches, and the lateral branches have a wide angle relative to the trunk, which prevents water from concentrating on the trunk and reduces the accumulation of atmospheric deposition. In addition, the leaves of *Cun. lanceolata* are waxy, whereas those of *Cas. carlesii* are leathery, and the substances secreted by *Cas. carlesii* leaves are more water-soluble [53]. Compared to monoculture forests, mixed and *Cas. carlesii* natural forests tend to have a rougher canopy structure [54], and the diverse tree species in mixed forests facilitate the canopy complementarity [55,56]. In this study, the roughness of the bark of *Cas. carlesii* and *Cun. lanceolata* differs. A rough bark demonstrates enhanced ion adsorption compared to smooth bark [57]. Also, the duration of throughfall is shorter than that of stemflow, which may affect ion movement. Changes in element concentrations are related to rainfall timing and interval [58,59], as well as retention thresholds of the canopy and bark [53]. The bark of *Cun. lanceolata* plantations are characterized by their thickness, roughness, and numerous cracks. The resin secreted by *Cun. lanceolata* plantations have a waxy nature, which facilitates the capture of atmospheric particles. This may explain the concentration of heavy metals in the stemflow of *Cun. lanceolata* plantations is higher than those of the other three forest types.

The concentrations of the three heavy metals vary greatly between throughfall and stemflow, the underlying mechanism may be related to differences in the solubility of metals from different sources and their mobility through hydrological pathways, as well as the timescale of precipitation. Existing studies suggest that, in the early stages of precipitation, rainwater primarily washes down dry deposition, while later, it further leaches soluble ions [13]. Pinos Juan (2020) found that, compared to throughfall, stemflow has a longer residence time and slower flow rate, which promotes ion enrichment in stemflow relative to atmospheric precipitation, especially under high-temperature conditions [60]. In addition, the throughfall volume (3569.7 mm) in this study is 22 times greater than the stemflow (161.6 mm). The concentration of heavy metals in stemflow is constrained by the volume of stemflow, and therefore, its concentration is lower than that of throughfall. Heavy metals are primarily influenced by climate factors, with concentrations being higher in the dry season compared to the rainy season, suggesting that precipitation dilutes the concentration of heavy metals. This indicates that the main source of heavy metals may be atmospheric deposition. For heavy metal elements primarily sourced from natural processes, such as soil weathering, like Pb, their particles tend to be larger, more insoluble, and less mobile

in water, making them harder to leach and migrate. As a result, the forest canopy has a weaker retention and purification effect on Pb. In contrast, Cr and Cd are more likely to be anthropogenic in origin, possess higher solubility, and have greater reactivity, allowing the forest canopy to effectively retain and filter them. This pattern helps explain the higher concentrations of Pb in the study area. Since the area is relatively far from urban centers and industrial areas and is less affected by human activities, the concentration of Pb—primarily from natural sources—is higher than that of Cr and Cd, which are more influenced by anthropogenic sources. Additionally, Cd typically binds with soil particles, making it less active in the soil [61]. Compared to Cr, Cd is less likely to enter the tree system through throughfall, which is why Cr concentrations are higher than Cd in the study area.

It should be noted that natural forests also exhibit higher concentrations of heavy metals, possibly due to their greater diversity of tree species, older age, and presence of more epiphytic plants, which may lead to more active biochemical reactions within their canopy layers [62]. In the subtropical region, characterized by a humid climate, epiphytic plants are integral components of forest ecosystems. Epiphytes located on branches may adsorb heavy metals via stemflow along tree trunks. The final fluxes of heavy metals in the stemflow are highest in natural forests, primarily because the amount of stemflow in *Cun. lanceolata* plantations is significantly lower than in other forest types, whereas stemflow in the natural forests not only has higher concentrations of heavy metals but also has higher volumes per unit area. In addition, Cr and Cd were leached from the canopies, while Pb showed an opposite pattern, indicating that Pb was absorbed by the canopies. This may be related to the significant retention effect of coniferous forests on Pb [27]; needle leaves are more effective at capturing atmospheric Pb, resulting in higher concentrations of Pb in the stemflow under conditions of rainwater dissolution and cracked bark.

We found that throughfall and stemflow are affected by seasonal changes. The concentration and flux of heavy metals in throughfall are higher in the rainy than in the dry season, primarily due to variations in rainfall and the fluctuations of intercepted particulate matter. The study area features high air quality and limited impact from human activities; typhoons in the rainy season can introduce dust, increasing the deposition of washed-out materials. Additionally, concentrated rainfall during this time improves the removal of these deposits. As a result, the heavy metal fluxes in throughfall increase with rainfall intensity, supporting this conclusion. The concentration and flux of heavy metals in stemflow show a seasonal pattern opposite to that of throughfall. This relationship is linked to the positive effects of temperature and wind speed on heavy metal concentration in trunk sap. In the dry season, higher temperatures and stronger winds enhance the tree's absorption of both water and heavy metals. In addition to the canopy structure examined in this study, leaf leaching is another important factor influencing heavy metal flux. Future research should further explore tree leaf morphology, leaf surface area, and canopy leaf area to better understand the underlying mechanisms.

5. Conclusions

Our results show that heavy metals in throughfall primarily originate from atmospheric deposition, with fluxes mainly driven by meteorological factors. In contrast, stemflow heavy metal fluxes are significantly influenced by forest type, with the highest flux observed in natural forests. This variation is linked to factors such as bark roughness and changes in forest type. Additionally, the dynamics of metal migration differ between elements, with chromium concentrations and fluxes higher in the rainy season, while cadmium and lead exhibit the opposite trend. Overall, rainfall partitioning plays a crucial role in the dynamics of heavy metal fluxes, and forest type changes contribute uniquely to these fluxes. This study provides valuable insights into heavy metal migration and hydrological

processes in forest ecosystems, offering guidance for the sustainable management of forests in the context of land use changes.

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