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**Atmospheric iodine, selenium and caesium depositions in France:  
II. Influence of forest canopies**

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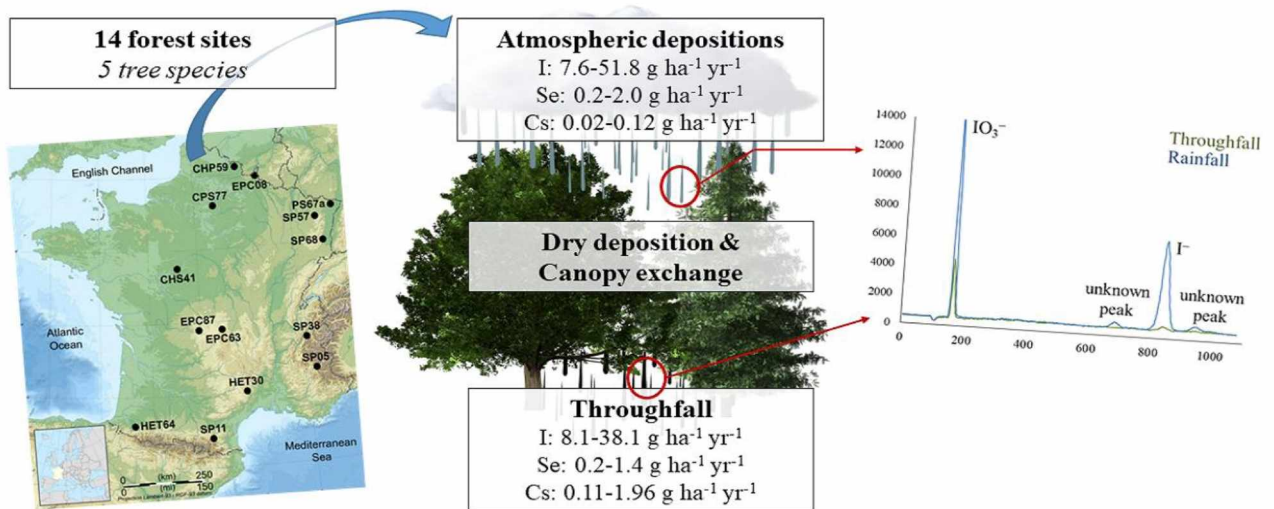
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## Graphical Abstract



## Highlights:

- Higher annual I, Se and Cs concentrations and fluxes in throughfall relative to rain
- Dry deposition contribution led to increased throughfall flux of I and Se
- Canopy budget model involved I and Se sorption by canopies
- Increase of annual flux of Cs in throughfall resulted mainly from canopy leaching
- Loss of inorganic I and Se species caused by forest canopy

## **Abstract**

Estimation of the canopy influence on atmospheric inputs of iodine (I), selenium (Se) and caesium (Cs) in terrestrial ecosystems is an essential condition for appropriate biogeochemical models. However, the processes involved in rain composition modifications after its passage through forest canopy have been barely studied for these elements. We monitored I, Se and Cs concentrations in both rainfall and throughfall of fourteen French forested sites throughout one year, and estimated dry deposition and canopy exchange fluxes for these elements, as well as speciation of I and Se. Comparison of rainfall and throughfall elemental composition highlighted an important impact of forest canopy on both (i) concentrations and fluxes of I, Se and Cs, and (ii) I and Se species. For the three elements, most of their throughfall concentrations were higher than corresponding rainfall. The increase of throughfall elemental fluxes was mostly due to dry deposition for I and Se although the canopy exchange model revealed some sorption within the canopy in most cases; for Cs, foliage leaching was most influencing. Regarding speciation, iodine species in rainfall were highly modified by forest canopy with an important increase of unidentified I proportion in throughfall (on average 49 and 82% in rainfall and throughfall, respectively), possibly due to washoff of dry deposition and/or to transformation into organic forms. Similarly, while rainfall was composed of 26-54% of inorganic Se, inorganic species were undetectable in throughfall. This dataset represents key information to improve modelling of I, Se and Cs cycling within forest ecosystems.

**Keywords:** Iodine; Selenium; Caesium; Throughfall; Speciation; Forest tree species

## 1. Introduction

Fluxes and speciation of atmospheric I, Se and Cs deposited in various terrestrial ecosystems are an essential baseline for understanding environmental conditions that control their persistence in soil and vegetation but their estimates still have large uncertainties. Forest ecosystems are particularly sensitive to atmospheric inputs because of, among others, the large interacting surface area of the forest canopy. The modification of the chemical composition of rain after its passage through the canopy (i.e. throughfall) results from various processes occurring in the phyllosphere: (i) direct absorption of elements and/or (ii) their release (leaching) by leaves or needles, (iii) accumulation then washoff of dry deposition (DD) and/or (iv) modification of physicochemical properties of rainfall leading to changes in element speciation (Lovett and Lindberg, 1984). Canopies could consequently influence the quantity and quality of water incoming to the forest floor. In order to evaluate canopy interaction with atmospheric inputs, the study of elemental compositions in both rainfall and throughfall is usually performed on major ions or nutrients (e.g. Adriaenssens et al., 2012; Staelens et al., 2008; Talkner et al., 2010). For example, it is generally assumed that the uptake or leaching of  $\text{Na}^+$ ,  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  by forest canopy is negligible (e.g. Adriaenssens et al., 2012; Lindberg et al. 1986; Talkner et al., 2010; Thimonier et al., 2008). Although enrichment of  $\text{Na}^+$  in throughfall has been shown to be mainly due to the accumulation of dry deposition on the canopy, the main reason of this enrichment for  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  is still unclear. Indeed, some authors have shown that part of these ions in throughfall can originate from foliage leaching (e.g. Neary and Gizyn, 1994; Staelens et al. 2007); while  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  may also originate from dry deposition (e.g. Adriaenssens et al., 2012; Talkner et al., 2010). For potassium, many authors agreed on its canopy leaching (e.g. Adriaenssens et al., 2012; Berger et al., 2006; Pajuste et al., 2006; Talkner et al., 2010). In spite of their environmental interest, including the existence of their radionuclide analogues, the interaction of trace elements such as iodine and

selenium with tree canopy have been barely studied. Indeed, to date, only five publications reported measurements of stable iodine in throughfall in U.K. (Neal et al., 2007), France (Roulier et al., 2018), Germany (Epp et al., 2020) and Japan (Takeda et al., 2016; Xu et al., 2016). Similarly, studies on Se canopy exchanges are scarce (Di Tullo, 2015; Tyler and Olsson, 2006). Due to the similarities in the chemistry (halogens and chalcogens) and atmospheric sources between I and Cl, and, Se and S, processes reported to act within the canopy for I and S may thus also affect fluxes and/or speciation of I and Se. More data are available for radiocaesium in rainfall and throughfall following Fukushima and Chernobyl accidents (e.g. Kato et al., 2017; Loffredo et al., 2015; Sombré et al., 1994; Yoschenko et al., 2018). As its chemical analogue potassium (e.g. Goor and Thiry, 2004; Varga et al., 2009), canopy leaching may thus act as the main process controlling Cs behaviour.

The persistence of iodine and selenium in terrestrial ecosystems depends on their incoming amounts but also on their speciation. In rain, the major inorganic iodine species are iodide ( $I^-$ ) and iodate ( $IO_3^-$ ), but iodine exists also as organic compounds (e.g. review of Fuge and Johnson, 2015). Similarly, selenium exists in several oxidation states, selenite ( $SeO_3^{2-}$ ) and selenate ( $SeO_4^{2-}$ ) being the major inorganic species detected in rainwaters (e.g. Alfthan et al., 1995; Cutter and Church, 1986; Floor et al., 2011; Robberecht et al., 1983; Suess et al., 2019). To date and to our knowledge, only Xu et al. (2016) provided information about iodine speciation in throughfall for a single Cedar site sampling; whereas Se speciation has never been surveyed in throughfall waters.

We reported in Part I. of this study the results of one year monitoring of I, Se and Cs concentrations in rainfall at 27 forest sites all over France (Roulier et al., 2020). In this paper, Part II., 14 of these forest sites representative of various tree species (i.e. Oak, Beech, Spruce, Pine, Silver fir), and equipped for throughfall sampling, were monthly monitored in order to examine the interaction of iodine, selenium and caesium with tree canopies. Iodine and

selenium speciation modification following canopy exchange was also investigated. The aim of this Part II. is to quantify atmospheric flux modifications induced by forest canopies for these trace species, and identify acting processes within the canopy leading to these changes.

## **2. Material and methods**

### **2.1. Study sites and sampling procedure**

The study sites and sampling procedure were already presented in Roulier et al., 2020. Briefly, sites are part of the French RENECOFOR network (i.e. National Network for the long-term Monitoring of Forest Ecosystem) managed by ONF (i.e. National Forest Board) ([www.onf.fr/renecofor](http://www.onf.fr/renecofor)). They are representative of five temperate forest species: Oak (CHP: Pedunculate Oak; CHS: Sessile Oak; CPS: Pedunculate/Sessile Oak), Spruce (EPC), Beech (HET), Scots Pines (PS) and Silver Fir (SP); and four climates: oceanic, transition, continental and mountainous (Figure 1 and SI Table S1). Fourteen sites are equipped with weather stations to sample both rainfall and throughfall waters. Rainfall and throughfall were weekly collected from September 2016 to September 2017 with bulk collectors (including dry deposition). Stemflow being not collected at these sites, it was thus not considered in this study. Composite samples per 28 days period were analysed (weight pondered average sample from the 4 weekly collected samples). The monthly basis composites were not acidified to prevent losses of I by volatilization and stored in the dark at 4°C after filtration at 0.45 µm (cellulose nitrate membrane filters). Hereafter, the four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

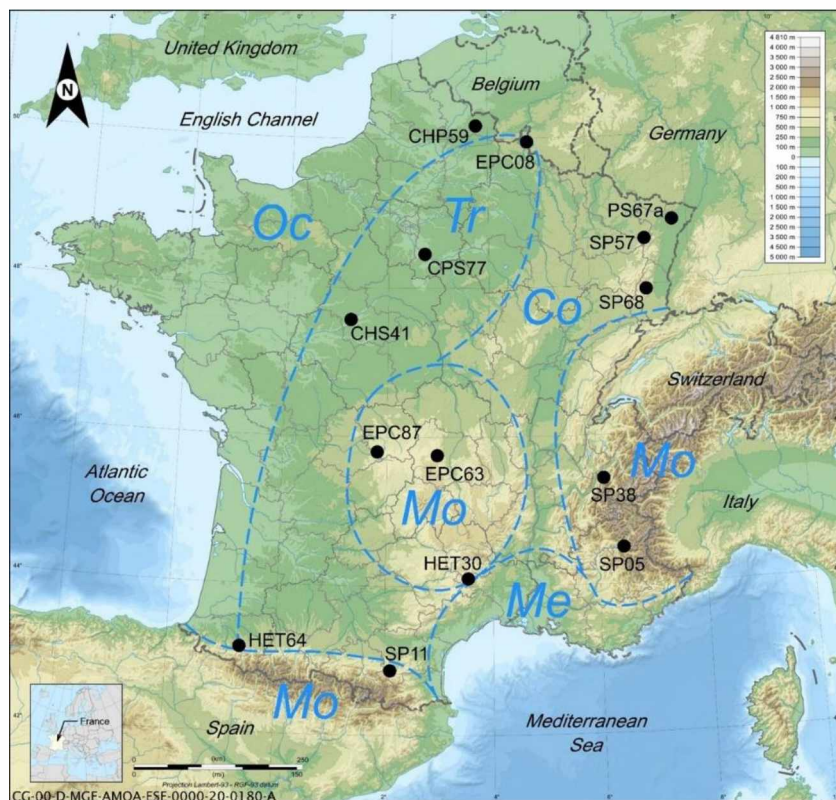


Figure 1. French climates and studied sites in the French RENECOFOR network (n = 14). Oc: Oceanic; Tr: Transition; Co: Continental; Mo: Mountainous; Me: Mediterranean; CHP=Pedunculate Oak; CHS=Sessile Oak; CPS=Sessile/Ped. Oak; EPC=Spruce; HET=Beech; PS=Scots Pine; SP=Silver fir. Base map: Eric Gaba (<https://commons.wikimedia.org/w/index.php?curid=4899458>).

## 2.2. Chemical analyses of rainfall and throughfall waters

The chemical analyses of waters for pH, dissolved organic carbon (DOC) and concentrations of Na (used to estimate dry deposition), K (analogue of Cs), I, Se and Cs were already detailed in Roulier et al. (2020). Briefly, Na and K were analyzed by ICP-AES with quantification limits of 0.01 and 0.05 mg L<sup>-1</sup>, respectively. Total soluble concentrations of I, Se and Cs were measured by inductively coupled plasma mass spectrometry (ICP-MS) (I and Cs: Agilent 7500ce; Se: Agilent 7900, Agilent Technologies, Japan). Quantification limits (I: 0.04 µg L<sup>-1</sup>; Cs: 0.6 ng L<sup>-1</sup>; Se: 2 ng L<sup>-1</sup>) were obtained from external calibrations in ultrapure water with analytical precision <5%. An Agilent 1200 series HPLC pump was coupled to ICP-MS (Agilent 7900) for speciation analyses of iodine (iodide I<sup>-</sup> and iodate IO<sub>3</sub><sup>-</sup>) and selenium



(selenite  $\text{SeO}_3^{2-}$  and selenate  $\text{SeO}_4^{2-}$ ). Chromatographic separation was carried out with a mobile phase of ammonium nitrate delivered at  $1 \text{ mL min}^{-1}$  flow rate (Agilent G3154-65001 column and G3154-65002 guard column, Agilent Technologies, Japan;  $20 \text{ mmol L}^{-1} \text{ NH}_4\text{NO}_3$ , 2.5% MeOH, pH 8.5). This method has quantification limits of 5 (I- $\text{IO}_3^-$  and Se- $\text{SeO}_4^{2-}$ ) and  $10 \text{ ng L}^{-1}$  (I- and Se- $\text{SeO}_3^{2-}$ ). The unidentified fraction (denoted “other” iodine and selenium:  $\text{I}_{\text{other}}$  and  $\text{Se}_{\text{other}}$ , respectively) was calculated as the difference between total and inorganic species concentrations (i.e.  $\text{I}_{\text{other}} = \text{I} - (\text{I}^- + \text{IO}_3^-)$  and  $\text{Se}_{\text{other}} = \text{Se} - (\text{SeO}_3^{2-} + \text{SeO}_4^{2-})$ ), and was always larger than zero.

## 2.3. Calculation and data analysis

### 2.3.1. Methods for calculation

According to Aussenac (1968), the amount of water reaching the forest floor (i.e. throughfall + stemflow) is generally less than the amount of water received in the field (i.e. rainfall). Several factors influence this interception, such as rainfall intensity, direct evaporation from crowns, foliage uptake and saturation capacity according to forest species, degree of tree defoliation and stand density (Ulrich et al., 1995). In our study, without stemflow collection, this interception was calculated as follow (Equation 1) with throughfall and rainfall water fluxes considered for the same time period (monthly or yearly basis):

$$\text{Interception (\%)} = \left(1 - \frac{\text{throughfall water flux}}{\text{rainfall water flux}}\right) \times 100 \quad (1)$$

To take into account the influence of sample volume on concentrations, the annual volume-weighted mean (AVWM) concentrations were calculated from elemental or species concentrations ( $c_i$ ;  $\mu\text{g L}^{-1}$ ) and the volume ( $V_i$ ; mm) of monthly basis composite samples for the analysed time period (13 periods of 28-days) as follow (Equation 2):

$$\text{AVWM} = \frac{\sum_i (c_i \times V_i)}{\sum_i V_i} \quad (2)$$

Rainfall (RF) and throughfall (TF) annual elemental fluxes ( $\text{g ha}^{-1} \text{ yr}^{-1}$ ) were calculated as the sum of monthly elemental concentrations in waters multiplied by their corresponding monthly water fluxes (provided by ONF-RENECOFOR). The net throughfall flux (NTF;  $\text{g ha}^{-1} \text{ yr}^{-1}$ ) was calculated as the difference between throughfall and rainfall fluxes. Since rainfall was collected with bulk collectors, including thus dry deposition, the NTF could be underestimated.

### 2.3.2. Concentration Ratios and Enrichment Factors

In order to compare elemental composition in throughfall and rainfall, the concentration ratios of the considered element (i.e. Na, I, Se and Cs) were determined from their AVWM concentrations (CR, Equation 3) as follow:

$$\text{CR}(i) = \frac{[i]_{\text{throughfall}}}{[i]_{\text{rainfall}}} \quad (3)$$

Where:  $i$ : considered element;  $[i]$ : AVWM elemental concentration ( $\mu\text{g L}^{-1}$ ).

In order to determine elemental enrichment in throughfall compared to rainfall, the enrichment factors of considered element (i.e. Na, I, Se and Cs) were determined from their RF and TF annual fluxes (EF, Equation 4) as follow:

$$\text{EF}(i) = \frac{\text{TF}(i)}{\text{RF}(i)} \quad (4)$$

Where:  $i$ : considered element; TF and RF: rainfall and throughfall elemental fluxes ( $\text{g ha}^{-1} \text{ yr}^{-1}$ ).

### 2.3.3. Canopy budget model

Interaction of the forest canopy with atmospheric inputs induces modification of the chemical composition of the rains under forest canopy (e.g. Adriaenssens et al., 2012; Gandois et al., 2010). In addition to wet deposition crossing the canopy, dry deposition (e.g. particles and adsorbed gases) could accumulate on the canopy and be further washed off. Moreover, an element can interact with the canopy (canopy exchange processes (CE)) including either

absorption ( $CE < 0$ ) or leaching ( $CE > 0$ ) of the element by the canopy. In the present study, dry deposition washed off by throughfall ( $DD_{TF}$ ) and canopy exchange fluxes were calculated with the canopy budget model (Staelens et al., 2008; Talkner et al., 2010). This model simulates the interaction of ions with forest canopy based on throughfall and rainfall measurements with the aim to distinguish relative importance of  $DD_{TF}$  and  $CE$  in their NTF fluxes (Equation 5).

$$NTF = TF - RF = DD_{TF} + CE \quad (5)$$

In the model, if element canopy exchange is assumed nil, the calculated  $DD_{TF}$  equals NTF flux (Staelens et al., 2008). Direct measurement of dry deposition is quite impossible in forest ecosystems because of the complexity of the forest canopy area, hence their estimation is done through the “filtering approach” using sodium as an inert reference ion (Adriaenssens et al., 2012; Staelens et al., 2008; Ulrich, 1983). Indeed, as no interaction of Na with canopy (low uptake and leaching by tree) is supposed, its NTF is only due to dry deposition. In this approach, particles and aerosols containing sodium and considered elements are assumed to be deposited with the same efficiency onto the forest canopy. Therefore, the dry deposition of an element washed off by throughfall ( $DD_{TF}(i)$ ;  $g\ ha^{-1}\ yr^{-1}$ ) was calculated using Equation 6:

$$DD_{TF}(i) = RF(i) \times \frac{NTF(Na)}{RF(Na)} \quad (6)$$

Where:  $RF(i)$ : rainfall element flux ( $g\ ha^{-1}\ yr^{-1}$ );  $NTF(Na)$ : net throughfall Na flux ( $g\ ha^{-1}\ yr^{-1}$ ) and  $RF(Na)$ : rainfall Na flux ( $g\ ha^{-1}\ yr^{-1}$ ).

#### **2.3.4. Statistical analyses**

Prior to the statistical analyses, data normality was evaluated using Shapiro-Wilk test. When normality was fulfilled, relationships between pairs of continuous variables (i.e. water interception; rainfall and throughfall amounts; NTF of Na, I, Se and Cs; CR, DD and CE of I, Se and Cs) were tested with Pearson's correlations. When normality was not met, Spearman's correlations were performed. Given the small number of sites per tree species (Oak, n = 3; Beech, n = 2; Spruce, n = 3; Scots Pines, n = 1; Silver Fir, n = 5), statistical analyses were restricted to the comparison between the two functional tree types namely deciduous (Oak and Beech; n = 5) and coniferous (Spruce, Scots Pines and Silver Fir; n = 9). Hence, comparison between functional tree types were performed using Two-Samples Wilcoxon tests. A Kruskal-Wallis test was performed between NTF of Na and the climate type (the only oceanic site was grouped with transition sites: transition, n = 5; continental, n = 3; mountainous, n = 6). Statistical analyses were performed using the R programming environment (R Core Team, 2013, version 3.4.3). Simple linear regressions were performed using Excel software.

### 3. Results and discussion

Detailed data of (a) annual water amount, annual pH mean, AVWM DOC, Na and K concentrations, (b) AVWM total I, Se and Cs concentrations, and I and Se species proportions in RF and TF, (c) monthly water amount and I, Se, Cs, Na and K concentrations in RF and TF for the 14-studied sites are available in SI Tables S2 to Table S9.

#### 3.1. Impact of the canopy on rainfall water fluxes and their physicochemical properties

On an annual basis, the throughfall amount represented between 44 and 86% of the rainfall amount (SI Figure S1), with no difference between functional tree types (Wilcoxon test,  $p = 0.364$ ). These results correspond to average values reported for temperate French forests (Aussenac, 1968; Gandois et al., 2010; Ulrich et al., 1995). For deciduous trees, the intensity of interception seemed not different according to seasons considering uncertainties (SI Figure S2). Only the beech HET64 site showed lower interception in spring-summer than autumn-winter seasons ( $R^2 = 0.91$  and  $0.97$ , slopes of regression lines TF versus RF =  $0.71 \pm 0.03$  and  $0.59 \pm 0.02$ , respectively).

Rainfall and throughfall pH values were mostly acidic, with mean annual values of  $6.0 \pm 0.5$  and  $6.2 \pm 0.5$ , respectively (SI Table S2). Dissolved organic carbon (DOC) concentrations in rainfall were very low (AVWM median =  $0.40 \text{ mg L}^{-1}$ ), while they were much higher in throughfall (AVWM median =  $16 \text{ mg L}^{-1}$ ) in agreement with literature (Gandois et al., 2010; Karavoltzos et al., 2017). Indeed, dissolved organic carbon in throughfall comes from foliar or needles leaching and microbiological activities in the phyllosphere as well as from soluble organic deposits washed off from the canopy surface (Mellec et al., 2010; Michalzik et al., 2001).

### **3.2. Impact of canopies on content and/or speciation of iodine, selenium and caesium atmospheric inputs**

#### **3.2.1. Sodium: indicator of dry deposition accumulated on canopies**

Sodium was a major element in rainfall with AVWM concentrations ranging from 0.20 to 0.82 mg L<sup>-1</sup> (SI Table S2; median = 0.34 mg L<sup>-1</sup>). In throughfall, AVWM Na concentrations were higher than in corresponding rainfall (range and median = 0.29-1.65 and 1.08 mg L<sup>-1</sup>, respectively), with concentration ratios ranging from 1.2 to 4.5 (SI Table S2). Throughfall Na concentrations led to annual fluxes from ~2.4 to 15.5 kg ha<sup>-1</sup> yr<sup>-1</sup> (Table 1; median = 5.2 kg ha<sup>-1</sup> yr<sup>-1</sup>), generally superior to rainfall ones (Figure 2 (a); EF(Na) mean = 1.78) with the exception of sites HET30 and SP38 whose TF were not significantly different from RF. Sodium enrichments in throughfall obtained from annual fluxes were confirmed by comparison of monthly TF and RF fluxes (SI Figure S3 and Figure S4) that showed TF enrichment can occur throughout the year. Since sodium is considered to be hardly taken up and leached by trees, exchange of this element with canopy is assumed to be negligible, thus its net throughfall is expected to equal its dry deposition (Adriaenssens et al., 2012; Gandois et al., 2010; Staelens et al., 2008; Talkner et al., 2010). Therefore, sites with high NTF(Na) are especially influenced by dry deposition. Hence, for our study period, the sites EPC08, EPC63 and SP11 were highly impacted by dry deposition (Table 1) compared to the sites HET30, SP05, SP38 and SP68 with low dry deposition fluxes (NTF(Na) < 1 kg ha<sup>-1</sup> yr<sup>-1</sup>). Although conifers are generally more efficient in trapping dry deposition than deciduous trees because of their aerodynamic properties and constant leaf area throughout the year (Bergkvist et al., 1989), in our study sodium enrichment (and thus the dry deposition flux) did not solely depend on functional tree types (Wilcoxon test,  $p = 0.699$ ). Furthermore, rainfall amount can impact dry deposition by their washing-off from canopy, but here no link between NTF(Na) and RF amount was found (Spearman's correlation,  $p = 0.492$ ). Moreover, NTF(Na) did not correlate neither with TF amount nor interception (Spearman's correlation,  $p = 0.952$  and  $0.916$ , respectively). The

influence of marine aerosol input on sodium NTF was tested according to climate type and no link was found (Kruskal-Wallis test,  $p = 0.693$ ). However, these different dry deposition fluxes in the various sites could be explained by numerous other factors (e.g. canopy structure, tree density in the stand, surface wetness chemistry, local meteorology; Adriaenssens et al., 2012; Gandois et al., 2010), that could not be tested based on the current data set.

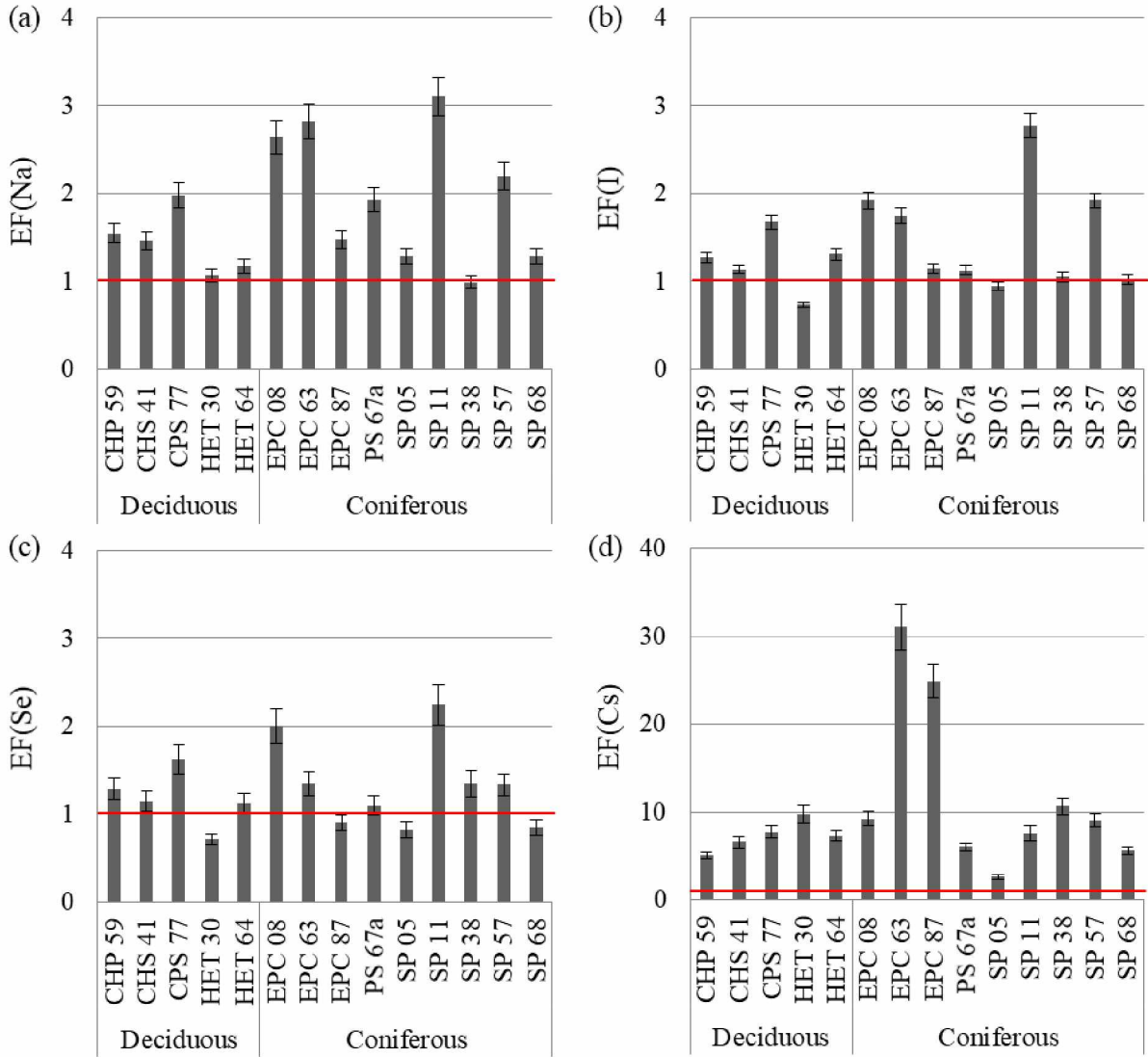


Figure 2. Enrichment Factors (EF) from rainfall and throughfall annual fluxes of (a) Na, (b) I, (c) Se and (d) Cs. Error bars based on error propagation of annual fluxes in rainfall and throughfall. Red lines correspond to EF equal to one. CHP: Pedunculate Oak; CHS: Sessile Oak; CPS: Pedunculate/Sessile Oak; HET: Beech; EPC: Spruce; PS: Scots Pines; SP: Silver Fir.

Table 1. Annual values and associated error of rainfall (RF), throughfall (TF), net throughfall (NTF), dry deposition washed off by throughfall (DD<sub>TF</sub>) and canopy exchange (CE) fluxes of Na, I, Se and Cs on the fourteen studied sites.

		Na	I	Se	Cs
		(kg ha <sup>-1</sup> yr <sup>-1</sup> )	(g ha <sup>-1</sup> yr <sup>-1</sup> )	(g ha <sup>-1</sup> yr <sup>-1</sup> )	(g ha <sup>-1</sup> yr <sup>-1</sup> )
CHP 59	RF	3.6 ± 0.2	9.9 ± 0.4	0.34 ± 0.02	0.042 ± 0.003
	TF	5.5 ± 0.3	12.7 ± 0.4	0.43 ± 0.03	0.21 ± 0.01
	NTF	1.9 ± 0.3	2.7 ± 0.5	0.10 ± 0.04	0.17 ± 0.01
	DD <sub>TF</sub>		5.4 ± 1.0	0.18 ± 0.03	0.023 ± 0.004
	CE		-2.7 ± 1.1	-0.09 ± 0.05	0.15 ± 0.01
CHS 41	RF	3.2 ± 0.2	10.2 ± 0.4	0.28 ± 0.02	0.031 ± 0.002
	TF	4.7 ± 0.2	11.6 ± 0.3	0.32 ± 0.02	0.20 ± 0.01
	NTF	1.5 ± 0.3	1.4 ± 0.5	0.04 ± 0.03	0.17 ± 0.01
	DD <sub>TF</sub>		4.7 ± 1.0	0.13 ± 0.03	0.014 ± 0.003
	CE		-3.4 ± 1.1	-0.09 ± 0.04	0.16 ± 0.01
CPS 77	RF	2.1 ± 0.1	8.2 ± 0.3	0.25 ± 0.02	0.028 ± 0.002
	TF	4.2 ± 0.2	13.8 ± 0.3	0.40 ± 0.03	0.21 ± 0.01
	NTF	2.1 ± 0.2	5.5 ± 0.5	0.15 ± 0.04	0.19 ± 0.01
	DD <sub>TF</sub>		8.1 ± 1.0	0.24 ± 0.03	0.027 ± 0.004
	CE		-2.5 ± 1.1	-0.09 ± 0.05	0.16 ± 0.01
EPC 08	RF	3.9 ± 0.2	12.5 ± 0.5	0.50 ± 0.03	0.048 ± 0.004
	TF	10.3 ± 0.5	24.0 ± 0.7	1.01 ± 0.08	0.44 ± 0.02
	NTF	6.4 ± 0.5	11.5 ± 0.8	0.50 ± 0.08	0.40 ± 0.02
	DD <sub>TF</sub>		20.5 ± 2.2	0.82 ± 0.10	0.08 ± 0.01
	CE		-9.0 ± 2.3	-0.32 ± 0.13	0.32 ± 0.02
EPC 63	RF	2.6 ± 0.1	9.9 ± 0.4	0.28 ± 0.02	0.063 ± 0.004
	TF	7.4 ± 0.4	17.3 ± 0.5	0.38 ± 0.03	1.96 ± 0.09
	NTF	4.7 ± 0.4	7.4 ± 0.7	0.10 ± 0.03	1.89 ± 0.09
	DD <sub>TF</sub>		18.0 ± 1.9	0.51 ± 0.06	0.11 ± 0.01
	CE		-10.6 ± 2.0	-0.41 ± 0.07	1.78 ± 0.09
EPC 87	RF	6.4 ± 0.3	16.4 ± 0.7	0.46 ± 0.03	0.075 ± 0.005
	TF	9.5 ± 0.5	18.8 ± 0.5	0.42 ± 0.03	1.87 ± 0.08
	NTF	3.1 ± 0.6	2.4 ± 0.8	-0.04 ± 0.04	1.80 ± 0.08
	DD <sub>TF</sub>		7.8 ± 1.5	0.22 ± 0.05	0.04 ± 0.01
	CE		-5.4 ± 1.8	-0.26 ± 0.06	1.76 ± 0.08
HET 30	RF	14.6 ± 0.7	51.8 ± 1.5	2.01 ± 0.10	0.12 ± 0.01
	TF	15.5 ± 0.8	38.1 ± 1.1	1.45 ± 0.10	1.16 ± 0.06
	NTF	0.9 ± 1.1	-13.7 ± 1.9	-0.57 ± 0.14	1.04 ± 0.06
	DD <sub>TF</sub>		3.2 ± 3.8	0.12 ± 0.15	0.01 ± 0.01
	CE		-16.9 ± 4.2	-0.69 ± 0.20	1.03 ± 0.06



HET 64	RF	$11.1 \pm 0.6$	$27.1 \pm 1.1$	$0.70 \pm 0.05$	$0.045 \pm 0.003$
	TF	$13.1 \pm 0.7$	$35.4 \pm 0.9$	$0.78 \pm 0.06$	$0.33 \pm 0.02$
	NTF	$1.9 \pm 0.9$	$8.3 \pm 1.4$	$0.08 \pm 0.08$	$0.28 \pm 0.02$
	DD <sub>TF</sub>		$4.7 \pm 2.1$	$0.12 \pm 0.05$	$0.008 \pm 0.004$
	CE		$3.6 \pm 2.5$	$-0.04 \pm 0.09$	$0.28 \pm 0.02$
PS67a	RF	$1.6 \pm 0.1$	$7.6 \pm 0.3$	$0.31 \pm 0.02$	$0.043 \pm 0.002$
	TF	$3.1 \pm 0.2$	$8.6 \pm 0.3$	$0.34 \pm 0.02$	$0.26 \pm 0.01$
	NTF	$1.5 \pm 0.2$	$1.0 \pm 0.4$	$0.03 \pm 0.03$	$0.22 \pm 0.01$
	DD <sub>TF</sub>		$7.1 \pm 0.9$	$0.28 \pm 0.04$	$0.04 \pm 0.01$
	CE		$-6.1 \pm 1.0$	$-0.25 \pm 0.05$	$0.18 \pm 0.01$
SP 05	RF	$2.0 \pm 0.1$	$12.4 \pm 0.5$	$0.24 \pm 0.02$	$0.042 \pm 0.004$
	TF	$2.6 \pm 0.1$	$11.5 \pm 0.3$	$0.20 \pm 0.01$	$0.11 \pm 0.01$
	NTF	$0.6 \pm 0.2$	$-0.9 \pm 0.6$	$-0.04 \pm 0.02$	$0.07 \pm 0.01$
	DD <sub>TF</sub>		$3.6 \pm 1.0$	$0.07 \pm 0.02$	$0.012 \pm 0.004$
	CE		$-4.5 \pm 1.2$	$-0.11 \pm 0.03$	$0.06 \pm 0.01$
SP 11	RF	$3.5 \pm 0.2$	$11.7 \pm 0.5$	$0.31 \pm 0.02$	$0.022 \pm 0.002$
	TF	$10.8 \pm 0.5$	$32.4 \pm 0.9$	$0.70 \pm 0.05$	$0.16 \pm 0.01$
	NTF	$7.3 \pm 0.6$	$20.7 \pm 1.0$	$0.39 \pm 0.06$	$0.14 \pm 0.01$
	DD <sub>TF</sub>		$24.6 \pm 2.5$	$0.65 \pm 0.08$	$0.05 \pm 0.01$
	CE		$-3.9 \pm 2.7$	$-0.27 \pm 0.09$	$0.10 \pm 0.01$
SP 38	RF	$2.7 \pm 0.1$	$12.3 \pm 0.5$	$0.41 \pm 0.03$	$0.07 \pm 0.01$
	TF	$2.7 \pm 0.1$	$12.9 \pm 0.4$	$0.55 \pm 0.04$	$0.75 \pm 0.03$
	NTF	$0.0 \pm 0.2$	$0.6 \pm 0.6$	$0.14 \pm 0.05$	$0.68 \pm 0.03$
	DD <sub>TF</sub>		$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$
	CE		$0.6 \pm 0.6$	$0.14 \pm 0.05$	$0.68 \pm 0.03$
SP 57	RF	$2.3 \pm 0.1$	$10.8 \pm 0.4$	$0.45 \pm 0.03$	$0.077 \pm 0.005$
	TF	$5.0 \pm 0.2$	$20.7 \pm 0.4$	$0.60 \pm 0.04$	$0.70 \pm 0.03$
	NTF	$2.7 \pm 0.3$	$9.9 \pm 0.6$	$0.15 \pm 0.05$	$0.62 \pm 0.03$
	DD <sub>TF</sub>		$12.9 \pm 1.5$	$0.54 \pm 0.07$	$0.09 \pm 0.01$
	CE		$-3.0 \pm 1.6$	$-0.39 \pm 0.08$	$0.53 \pm 0.03$
SP 68	RF	$2.0 \pm 0.1$	$8.0 \pm 0.3$	$0.29 \pm 0.02$	$0.12 \pm 0.01$
	TF	$2.4 \pm 0.1$	$8.1 \pm 0.3$	$0.23 \pm 0.02$	$0.64 \pm 0.03$
	NTF	$0.4 \pm 0.2$	$0.2 \pm 0.4$	$-0.06 \pm 0.03$	$0.52 \pm 0.03$
	DD <sub>TF</sub>		$1.5 \pm 0.6$	$0.05 \pm 0.02$	$0.02 \pm 0.01$
	CE		$-1.3 \pm 0.7$	$-0.12 \pm 0.04$	$0.50 \pm 0.03$

### 3.2.2. Modification of iodine and selenium composition of atmospheric inputs

AVWM concentrations of I (SI Table S3; range and median = 1.37-4.97 and 2.85  $\mu\text{g L}^{-1}$ ) and Se (range and median = 31-149 and 90  $\text{ng L}^{-1}$ ) in throughfall were higher than in corresponding rainfall (range and median = 0.85-2.18 and 1.23  $\mu\text{g L}^{-1}$ , and 23-85 and 41  $\text{ng L}^{-1}$ ; for I and Se, respectively). The concentration ratios were in the same order of magnitude for both elements (SI Table S3; range and median = 0.9-4.0 and 2.0, and 0.8-3.3 and 1.8; for I and Se, respectively). Only the beech site HET30 presented CR below one for both elements. Focusing on functional types, no difference was detected between functional tree types for both CR(I) or CR(Se) (Wilcoxon test,  $p = 0.298$  and  $0.438$ , respectively). CR(I) values were in the same range as those found in the literature in European and Japanese forests (Neal et al., 2007; Roulier et al., 2018; Xu et al., 2016). Annual rainfall and throughfall fluxes of I varied respectively from 7.6 to 51.8 and from 8.1 to 38.1  $\text{g ha}^{-1} \text{yr}^{-1}$  (Table 1). Annual throughfall fluxes of I were not different to rainfall ones considering uncertainties in the sites SP05, SP38 and SP68 (Table 1), and inferior to rainfall ones only in the site HET30 with net throughfall of  $-13.7 \pm 1.9 \text{ g ha}^{-1} \text{yr}^{-1}$ . An iodine enrichment in throughfall compared to rainfall occurred for all other sites (Figure 2 (b); EF(I) mean = 1.56), involving positive NTF(I). For selenium, annual rainfall and throughfall fluxes varied respectively from 0.24 to 2.01 and from 0.20 to 1.45  $\text{g ha}^{-1} \text{yr}^{-1}$  (Table 1). Selenium enrichment of throughfall was observed for seven sites considering uncertainties (Figure 2 (c); EF(Se) mean = 1.60), resulting in positive NTF(Se) for these sites; while for the remaining sites, throughfall fluxes were not different to rainfall, excepted HET30 with negative net throughfall. No significant difference was found for the annual NTF(I) or NTF(Se) neither between functional tree types (Wilcoxon test,  $p > 0.05$ ), nor according to annual RF and TF amounts (Spearman's correlation,  $p > 0.05$ ). Regarding monthly fluxes, TF fluxes of iodine and selenium were predominantly higher or equal to those of RF consistent with results obtained at annual scale for sites showing positive annual NTF (SI Figure

S5 and Figure S6 for I; SI Figure S7 and Figure S8 for Se). Moreover, no seasonal or phenological stages dependence was clearly observed for both elements. The site HET30 presented the lower (negative) annual NTF(I) and NTF(Se) due mainly to two months with high precipitations (> 400 mm) and maximum negative monthly NTF fluxes. Hence, iodine and selenium showed similar behaviour as highlighted by significant positive correlations between NTF of I, Se and Na (Figure 3 (a)(b)(d); Spearman's correlation,  $p < 0.05$ ), and a strong positive correlation between concentration ratios of Se and I (SI Figure S9; Pearson's correlation,  $p < 0.001$ ).

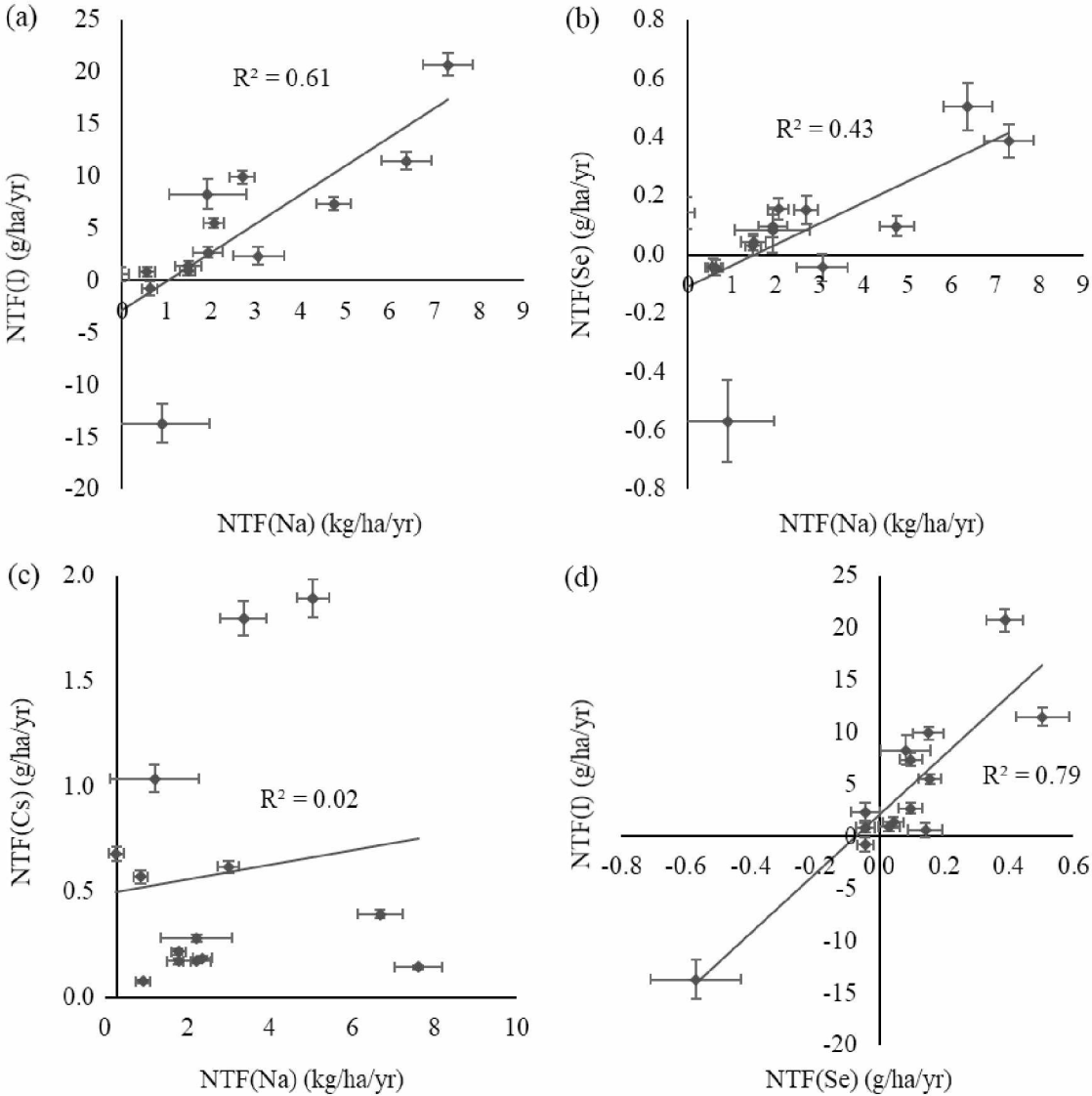


Figure 3. Correlations between net throughfall (NTF) of Na and those of (a) I, (b) Se and (c) Cs. (d) Correlation between NTF(I) and those of Se. Error bars correspond to standard errors.

Estimated  $DD_{TF}$  fluxes of I and Se were of  $6.23$  and  $0.20 \text{ g ha}^{-1} \text{ yr}^{-1}$  (median values) (Table 1) and accounted respectively for  $47 \pm 32$  and  $55 \pm 38\%$  of TF. The sites EPC08, EPC63, SP11 and SP57 presented higher  $DD_{TF}(I)$  and  $DD_{TF}(Se)$  fluxes ( $>12.9$  and  $>0.51 \text{ g ha}^{-1} \text{ yr}^{-1}$ , respectively) compared to overall other sites ( $4.6 \pm 2.7$  and  $0.14 \pm 0.09 \text{ g ha}^{-1} \text{ yr}^{-1}$ , respectively). No significant difference was found for  $DD_{TF}(I)$  or  $DD_{TF}(Se)$  neither between functional tree types (Wilcoxon test,  $p > 0.05$ ), nor according to annual RF and TF amounts (Spearman's correlation,  $p > 0.05$ ). Importance of the canopy exchange (CE) varied greatly from one stand to another (Table 1), but it did not seem to depend on (i) functional tree types (Wilcoxon test,  $p > 0.05$ ), and (ii) RF amount or interception (Spearman's correlation,  $p > 0.05$ ). Iodine canopy exchange was positive only for the site HET64 ( $3.6 \pm 2.5 \text{ g ha}^{-1} \text{ yr}^{-1}$ ), involving leaching of I by trees of this stand; whereas it was negative for other sites involving iodine (ab)sorption by tree canopy (excepted for the site SP38 with CE equal to zero considering uncertainty). In case of Se, only the site SP38, for which  $DD_{TF}$  flux was equal to zero, shown positive CE. Such results indicated that leaves or needles (ab)sorbed selenium in the majority of stands. Although iodine and selenium (ab)sorption could be overestimated, it seems that uptake of these elements occurred by foliar surface and/or related living organism. Indeed, in our previous study on those sites (Roulier et al., 2019), we found a strong correlation between forest litters and rainfall iodine concentrations, involving a potential iodine sorption by leaves and needles returned to soil after their senescence. Some other studies highlighted the foliar sorption of iodine by vegetables after spraying the leaves with radioiodine or following atmospheric radioiodine inputs (e.g. Collins et al., 2004; Hasegawa et al., 2014; Hurtevent et al., 2013; Ota et al., 2020; Tschiersch et al., 2009). In our previous study (Roulier et al., 2018), the washing of beech leaves suggested that the majority of I was incorporated inside the leaves. Same, several studies showed that foliar sorption of selenium was possible for some species following foliar

application of Se solutions (Se(IV) or Se(VI)) (Haygarth et al., 1994; Hu et al., 2002; Hurtevent et al., 2013; Kápolna et al., 2009).

### 3.2.3. Modification of caesium composition of atmospheric inputs

AVWM Cs concentrations in throughfall showed a greater variability (range = 17-310 ng L<sup>-1</sup>) between sites than I and Se (SI Table S3; median = 61 ng L<sup>-1</sup>). As for I and Se, caesium concentrations in throughfall were higher than in corresponding rainfall (range and median = 2.3-13.2 and 5.4 ng L<sup>-1</sup>) with the greatest concentration ratios from 4.2 to 48.9 (SI Table S3; mean = 16.2), especially for the spruce sites EPC63 and EPC87 (CR = 48.9 and 43.3, respectively). Annual throughfall Cs fluxes varied greatly from one site to another by a factor 16 (Table 1; median = 0.39 g ha<sup>-1</sup> yr<sup>-1</sup>), and comparison with RF at the annual scale (median = 0.05 g ha<sup>-1</sup> yr<sup>-1</sup>) confirmed large enrichment for Cs (Figure 2 (d); EF range and mean = 2.7-31.1 and 10.2) compared to I and Se. Therefore, net throughfall of Cs was always positive, values ranging between +0.1 to +1.9 g ha<sup>-1</sup> yr<sup>-1</sup> (median = 0.3 g ha<sup>-1</sup> yr<sup>-1</sup>). Highest net throughfall fluxes were quantified at beech HET30 and spruce EPC63 and EPC87 sites. This throughfall enrichment occurred in all seasons and sites comparing monthly RF and TF fluxes of Cs (SI Figure S10 and Figure S11). Caesium behaviour differed from that of Na, I and Se, as highlighted by the lack of correlation between annual NTF(Cs) and NTF(Na) (Figure 3 (c); Spearman's correlation,  $p = 0.892$ ).

Indeed, fluxes of DD<sub>TF</sub>(Cs) accounted to only  $9 \pm 8$  % of Cs in TF (Table 1) and they therefore did not explain the Cs enrichment in throughfall. Once DD<sub>TF</sub> was subtracted from NTF, Cs canopy exchange was always positive. Hence, throughfall enrichment in Cs would be linked to its leaching from leaves/needles internal tissues. That assumption is supported by significant and positive correlations between monthly NTF(Cs) and NTF(K) (SI Figure S12 and Figure S13,  $0.44 < R^2 < 0.98$ , expected for the sites HET64, PS67a and SP11). Indeed, the

contribution of  $K^+$  foliar leaching in forest is well known (e.g. Adriaenssens et al., 2012; Kopacek et al., 2009), and caesium is considered as a biological analogue of K (e.g. Thiry et al., 2009; Yoshihara et al., 2019).

#### **3.2.4. Focus on iodine and selenium speciation modification**

As no Se-containing peak was detected in throughfall chromatograms, only iodine speciation results will be discussed. However, since rainfall was composed of 26-54% of inorganic Se compounds (SI Table S3), Se speciation was thus strongly modified in throughfall compared to rainfall with complete loss of inorganic species.

An important modification of iodine speciation from rainfall to throughfall was also detected. In throughfall, proportions of  $I^-$ ,  $IO_3^-$  and “other” iodine were respectively: 3-25, 1-13 and 65-92% (SI Table S3; mean = 14, 4 and 82%, respectively), reflecting a consequent increase of “other” iodine proportion and a drastic decrease of inorganic iodine species proportion for most sites (in rainfall, range and mean = 19-44 and 29, 16-32 and 23, 39-60 and 49%, for  $I^-$ ,  $IO_3^-$  and  $I_{\text{other}}$  respectively). Furthermore, unknown iodine fraction in throughfall was at least in part different from the one in rainfall as no I-containing peak was detected in throughfall samples contrariwise to rainfall (SI Figure S14). Concomitantly, DOC concentrations in throughfall dramatically increased compared to rainfall ones (cf. §3.1.; SI Table S2). Actually, the delta of the “other” iodine concentration (i.e. corresponds to throughfall  $I_{\text{other}}$  concentration – rainfall  $I_{\text{other}}$  concentration) was overall positively correlated to those of DOC (SI Figure S15 and Figure S16;  $0.43 < R^2 < 0.83$ ). DOC in throughfall could come from (i) foliar or needles leaching and microbiological activities in the phyllosphere and/or (ii) from soluble organic deposits washed off from the canopy surface (Mellec et al., 2010; Michalzik et al., 2001). Considering the significant dry deposition of iodine estimated above, these correlations could support the assumption that the amount of iodine and its speciation in rainfall

were highly modified after crossing the canopy by washing-off of dry deposition. Alongside that, inorganic iodine fluxes decreased clearly, especially those of iodate (Figure 4;  $EF(IO_3^-)$  range and median = 0.1-0.6 and 0.2;  $EF(I^-)$  range and median = 0.1-1.5 and 0.7), according to modification of iodine speciation due to rainfall composition change and/or absorption by leaves and needles of these iodine species. No difference was found in the enrichment factors of  $I^-$  or  $IO_3^-$  between functional tree types (Wilcoxon's test,  $p = 0.190$  and  $0.147$ , respectively). Furthermore, no relationship was found between negatives CE(I) (involving iodine (ab)sorption by canopies) and the decrease of inorganic iodine fluxes (Spearman's test,  $p = 0.868$  and  $0.892$  for  $EF(I^-)$  and  $EF(IO_3^-)$ , respectively), probably due to dry deposition contribution whose exact role in iodine speciation modification is unknown. To date and to our knowledge, only Xu et al. (2016) provided information about iodine speciation in throughfall which was collected in March 2014 under deciduous and coniferous forest in Fukushima (Japan). They also found a modification of throughfall iodine speciation for both forests, exclusively composed of "other" iodine (iodine unidentified = 100%) contrary to rainfall with an "other" iodine fraction of 71.5%.

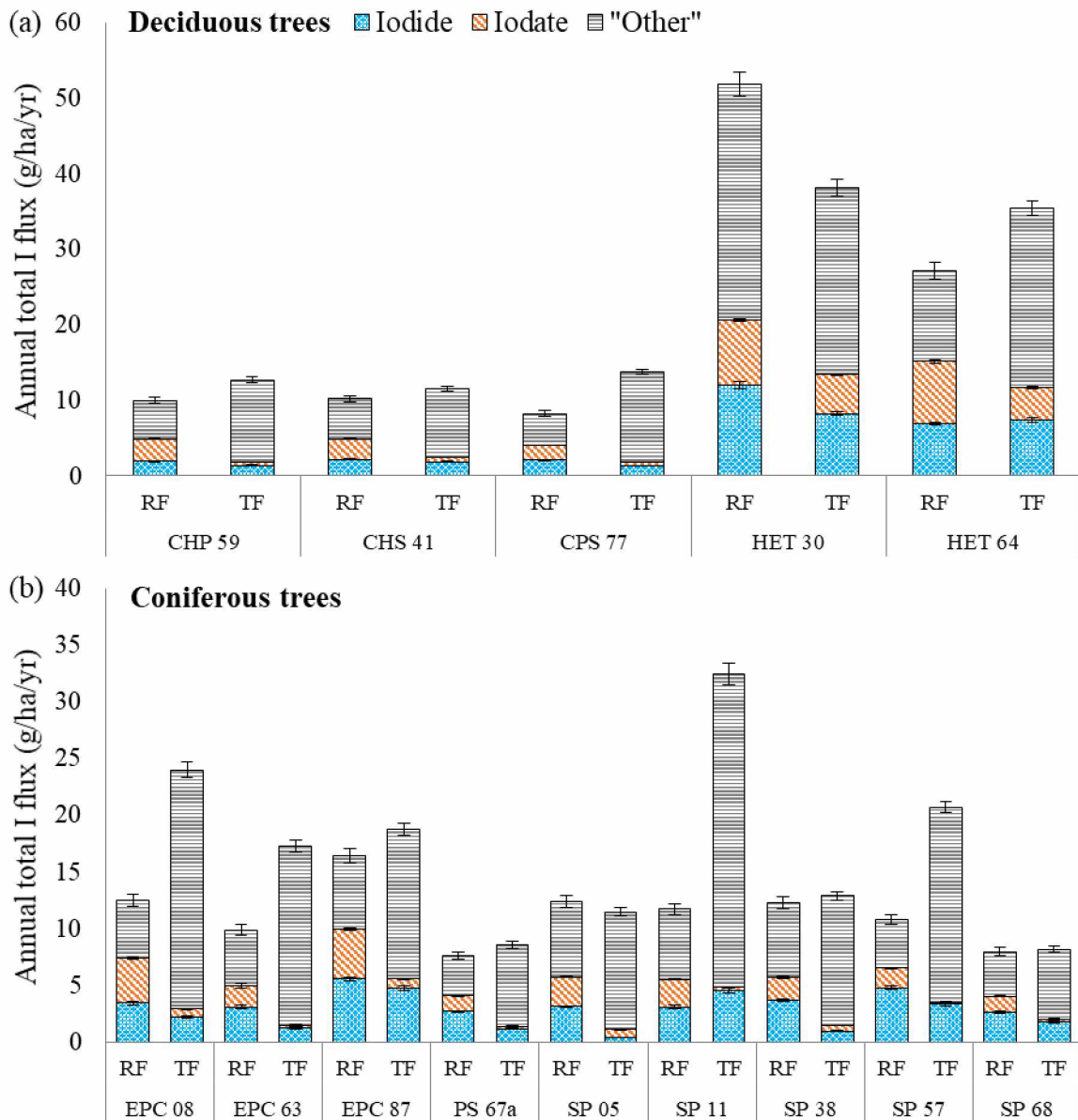


Figure 4. Proportions of iodide, iodate and “other” iodine in annual total I flux from rainfall (RF) and throughfall (TF) for (a) deciduous and (b) coniferous trees. CHP: Pedunculate Oak; CHS: Sessile Oak; CPS: Pedunculate/Sessile Oak; HET: Beech; EPC: Spruce; PS: Scots Pines; SP: Silver Fir. Error bars correspond to standard errors.



#### **4. Conclusion**

In the current study, dry deposition and canopy exchange fluxes of I, Se and Cs were estimated for fourteen forest sites representative of five tree species. Iodine and selenium speciation modifications in throughfall compared to rainfall were also studied. The results of one year monitoring showed that: (i) the three elements were enriched in throughfall compared to corresponding rainfall (mean concentration ratios = 2.2, 2.0 and 16.2 for I, Se and Cs respectively); (ii) the increase of throughfall elemental fluxes were estimated to be mostly due to dry deposition for I and Se, and to leaching from foliage for Cs; (iii) application of the canopy exchange budget model indicated that I and Se were sorbed within canopy in most cases; (iv) important modifications of I and Se speciation from rainfall to throughfall resulted in increased proportion of unidentified species (from 39-60 to 65-92% for iodine; from 46-74% to almost 100% for selenium), inorganic species were thus either minor compounds or undetectable. This study provides a better understanding of the processes acting within tree canopy and affecting atmospheric inputs of I, Se and Cs towards forest soils and their recycling by forest, and should thus help improving long-term predictions of the behaviour of these elements in forest ecosystems.

## **Credit author statement**

Roulier Marine: Conceptualization, Investigation, Validation, Formal analysis, Writing - original draft, Writing - review & editing; Maité Bueno: Conceptualization, Investigation, Validation, Funding acquisition, Writing - review & editing, Supervision; Frédéric Coppin: Conceptualization, Validation, Funding acquisition, Writing - review & editing, Supervision; Manuel Nicolas: Resources, Writing - review & editing; Yves Thiry: Conceptualization, Funding acquisition, Writing - review & editing; François Rigal: Formal analysis, Validation, Writing - review & editing; Isabelle Le Hécho: Conceptualization, Writing - review & editing, Supervision; Florence Pannier: Conceptualization, Writing - review & editing, Supervision.

## **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## **Appendix A. Supplementary data**

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2020.128952>.

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## Supporting Information

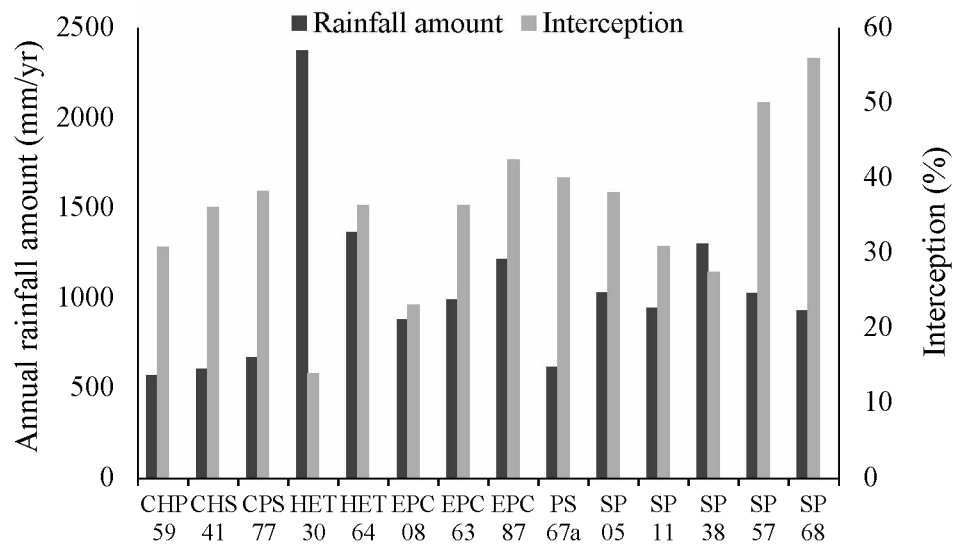


Figure S1. Annual rainfall amount and its interception by canopy (n = 14 sites). CHP: Pedunculate Oak; CHS: Sessile Oak; CPS: Pedunculate/Sessile Oak; HET: Beech; EPC: Spruce; PS: Scots Pines; SP: Silver Fir.

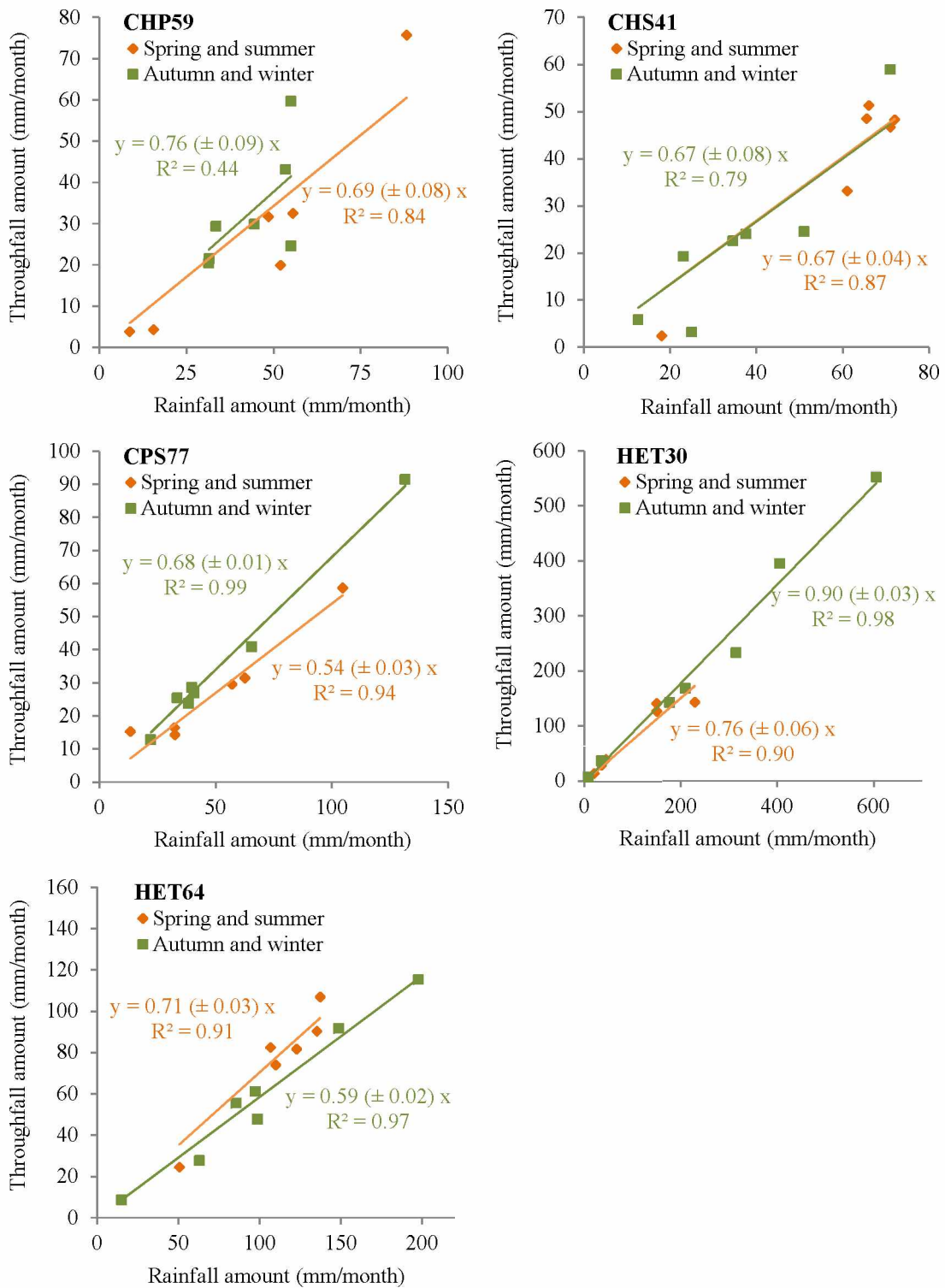


Figure S2. Monthly throughfall amount as function of those of rainfall for each deciduous trees stand and according to season. HET: Beech; CHP: Pedunculate Oak; CHS: Sessile Oak; CPS: Pedunculate/Sessile Oak. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).



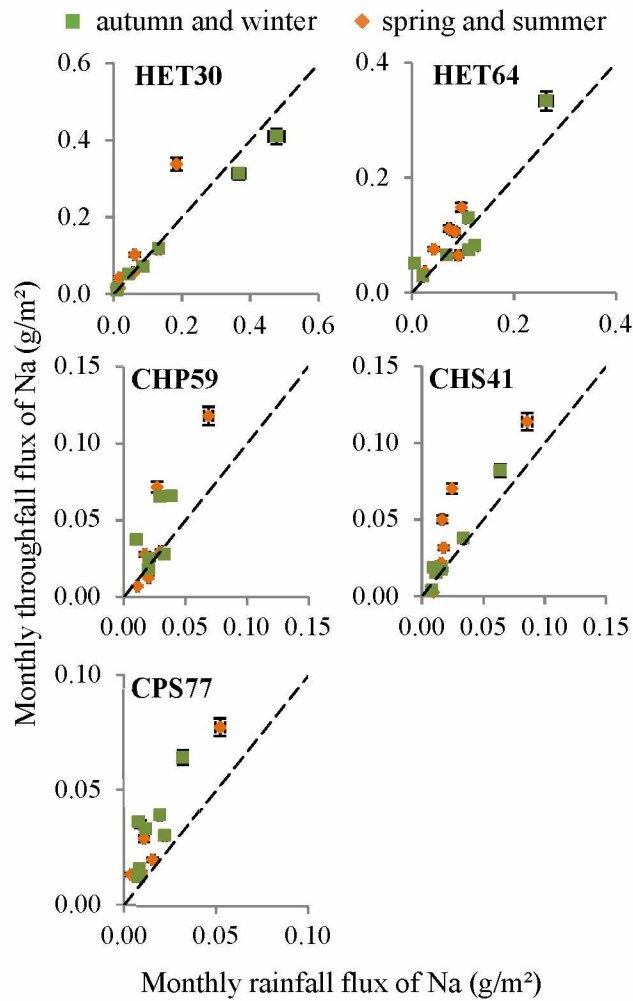


Figure S3. Monthly throughfall flux of Na as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. HET=Beech; CHP=Pedunculate Oak; CHS=Sessile Oak; CPS=Sessile/Ped. Oak. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

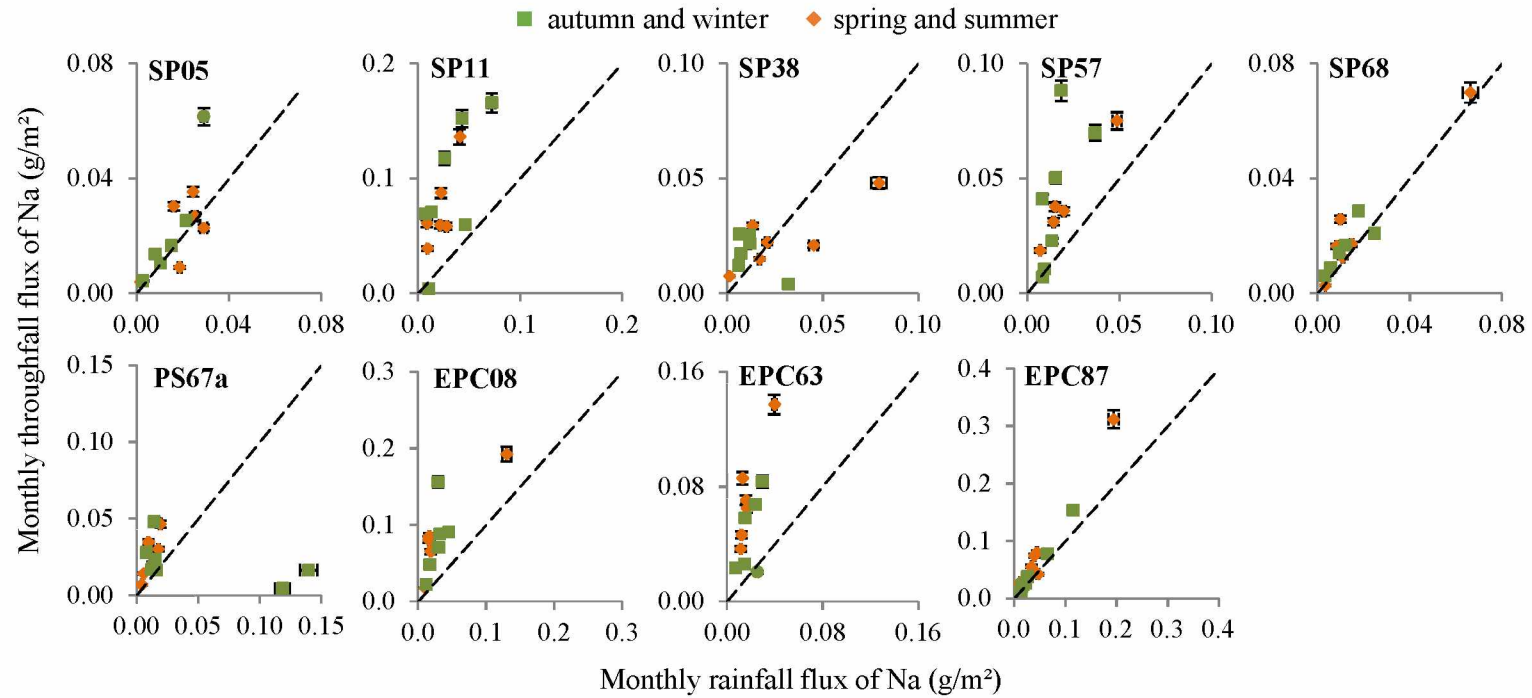


Figure S4. Monthly throughfall flux of Na as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. SP=Silver fir; PS=Scots Pine; EPC=Spruce. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

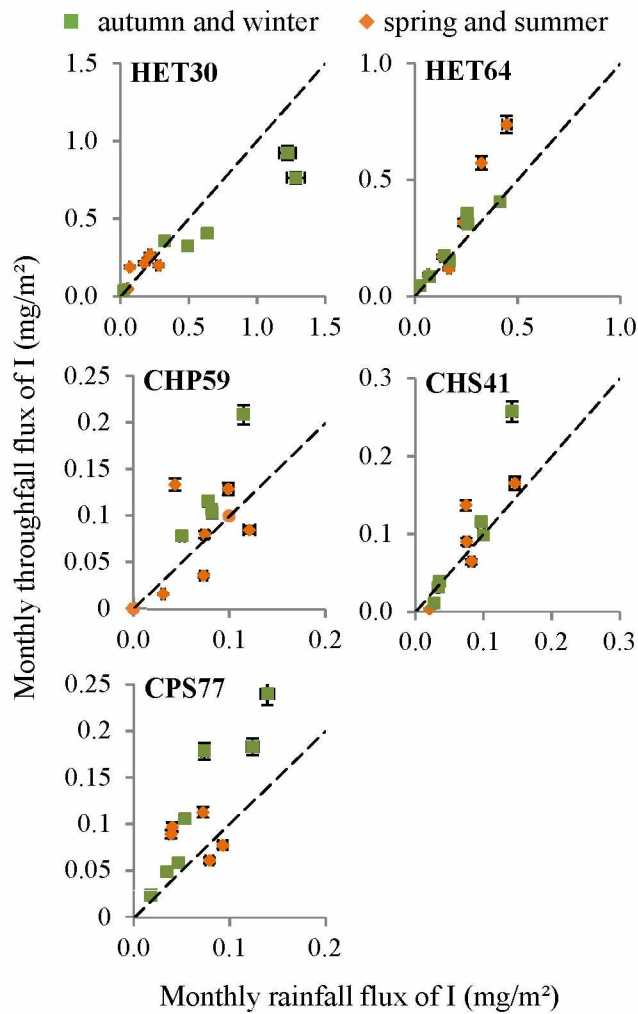


Figure S5. Monthly throughfall flux of I as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. HET=Beech; CHP=Pedunculate Oak; CHS=Sessile Oak; CPS=Sessile/Ped. Oak. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

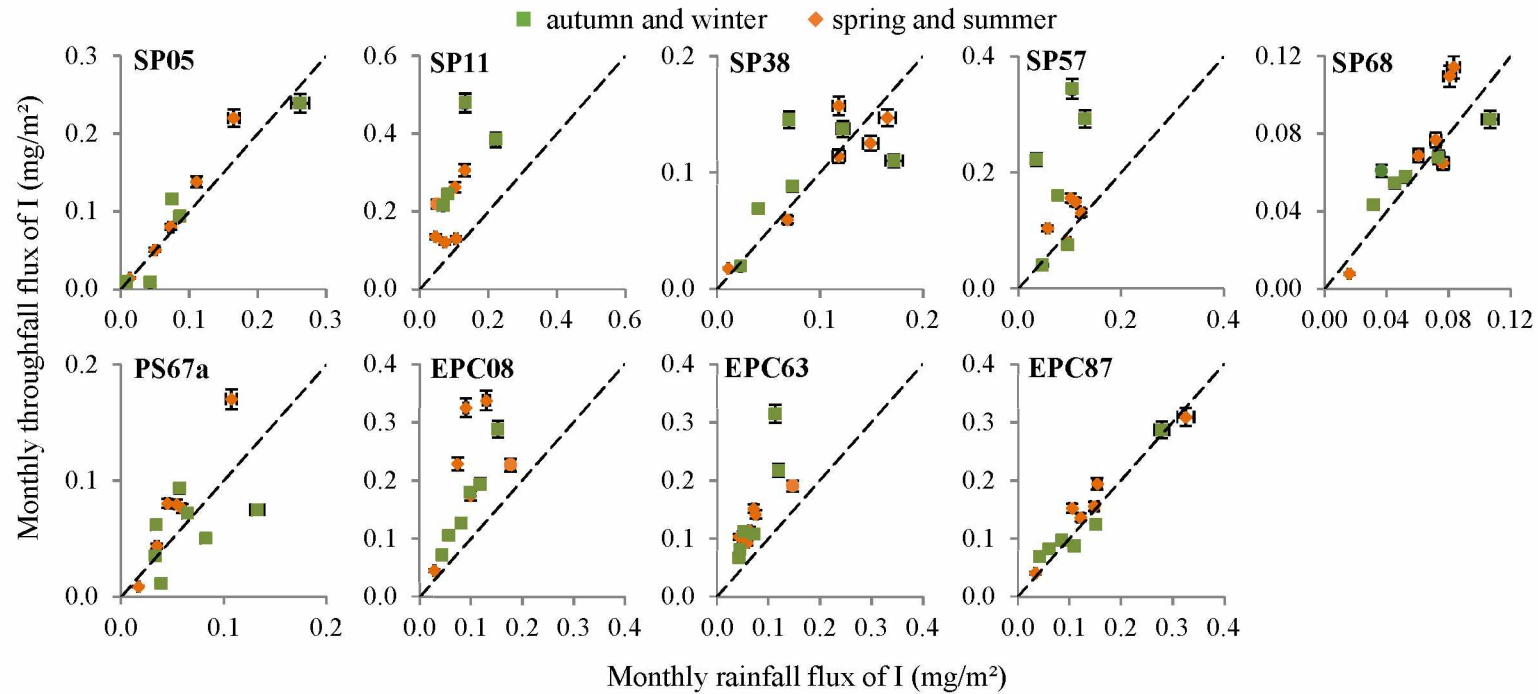


Figure S6. Monthly throughfall flux of I as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. SP=Silver fir; PS=Scots Pine; EPC=Spruce. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

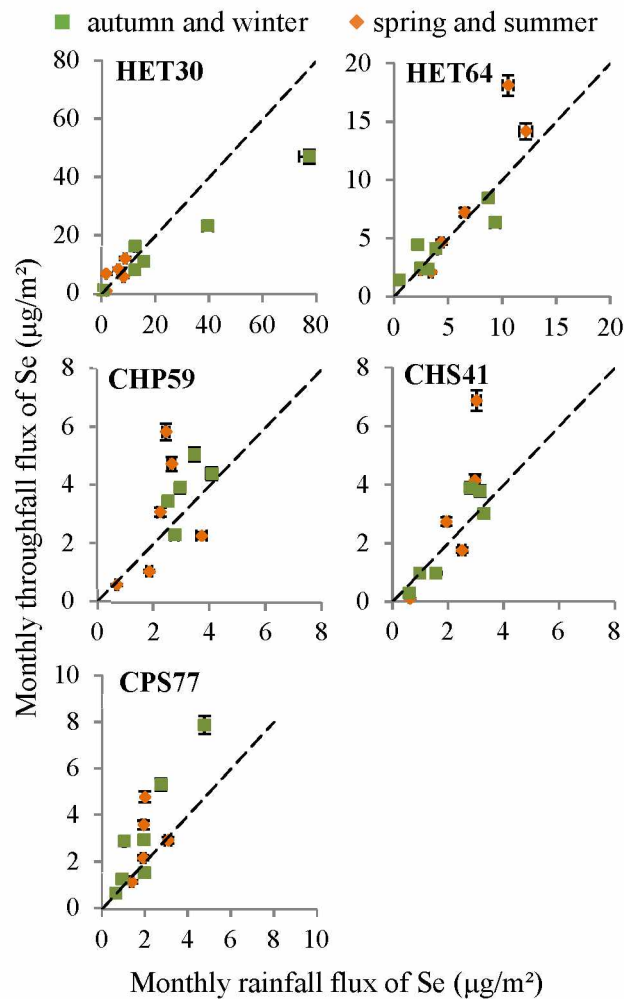


Figure S7. Monthly throughfall flux of Se as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. HET=Beech; CHP=Pedunculate Oak; CHS=Sessile Oak; CPS=Sessile/Ped. Oak. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

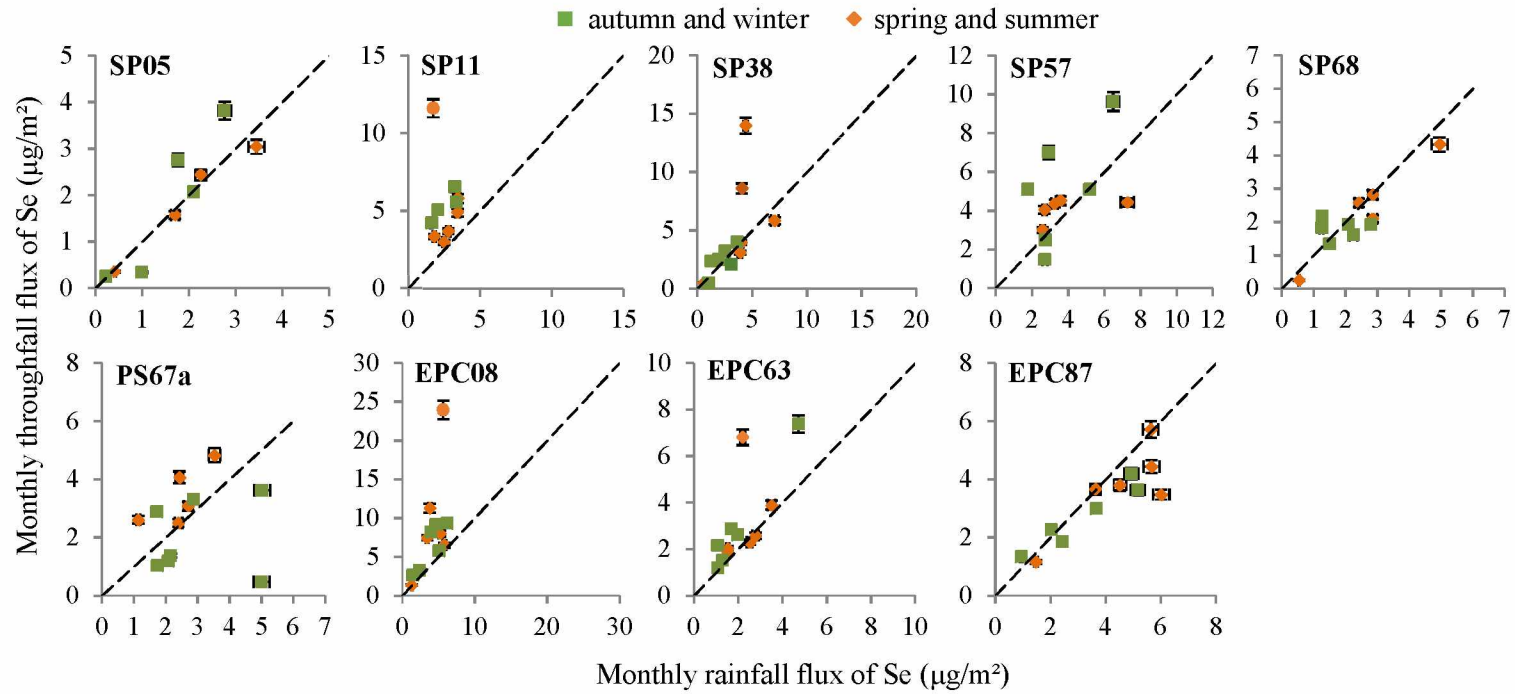


Figure S8. Monthly throughfall flux of Se as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. SP=Silver fir; PS=Scots Pine; EPC=Spruce. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

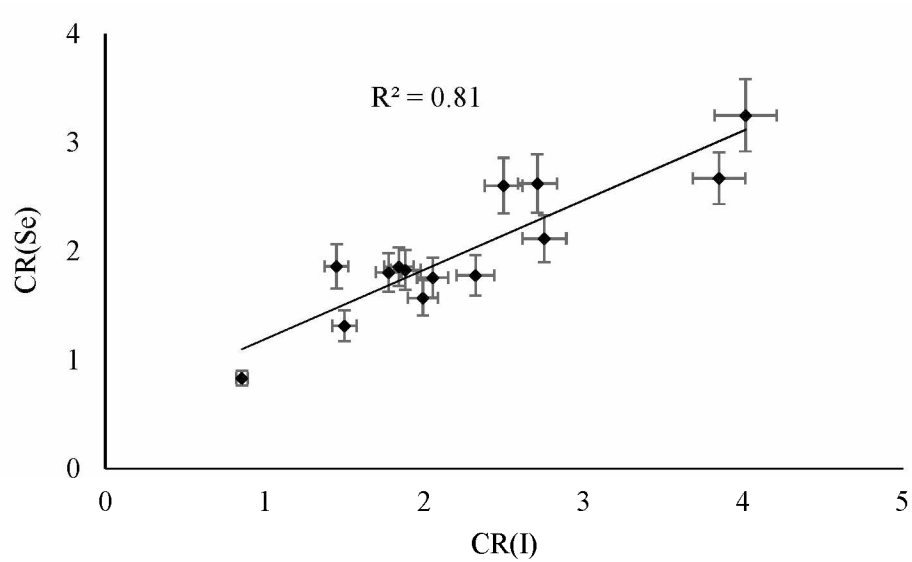


Figure S9. Concentration Ratios (CR) from AVWM concentrations of Se as function of those of I (n = 14 sites). Error bars correspond to standard errors.

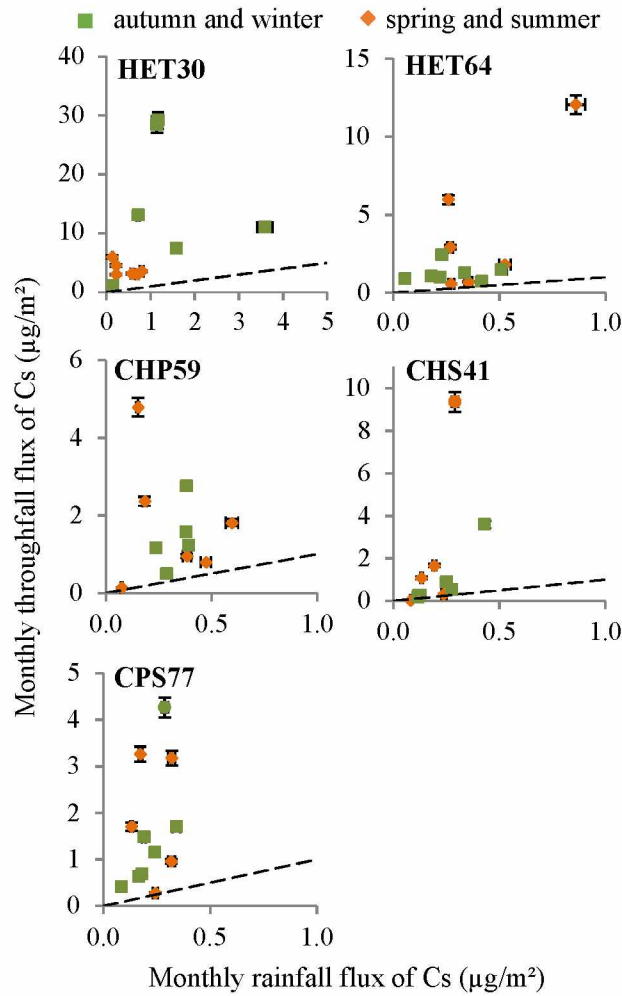


Figure S10. Monthly throughfall flux of Cs as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. HET=Beech; CHP=Pedunculate Oak; CHS=Sessile Oak; CPS=Sessile/Ped. Oak.

The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).



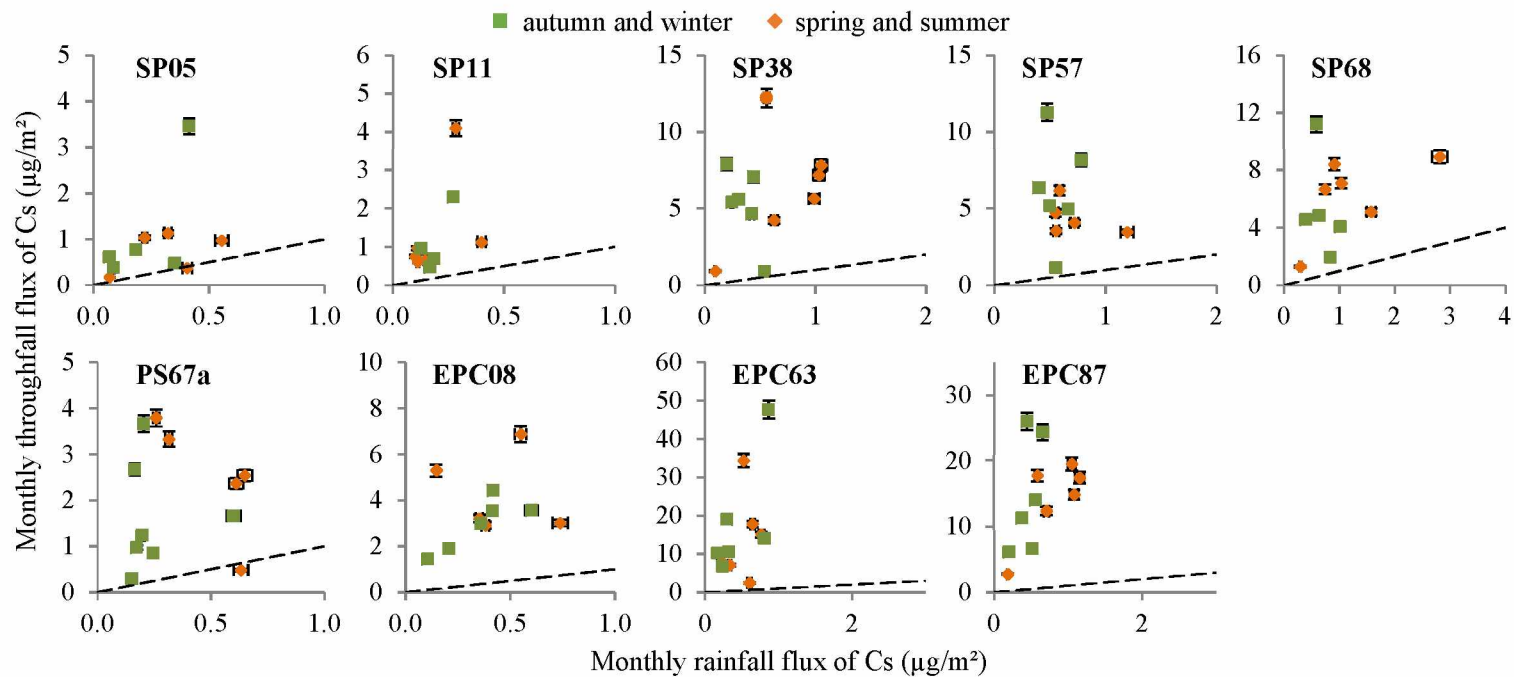


Figure S11. Monthly throughfall flux of Cs as function of those of rainfall according to season. Error bars correspond to standard errors. The dotted line corresponds to a 1:1 ratio. SP=Silver fir; PS=Scots Pine; EPC=Spruce. The four seasons were defined as follows: September 2016 to November 2016 and September 2017 (Autumn), December 2016 to February 2017 (Winter), March 2017 to May 2017 (Spring), and June 2017 to August 2017 (Summer).

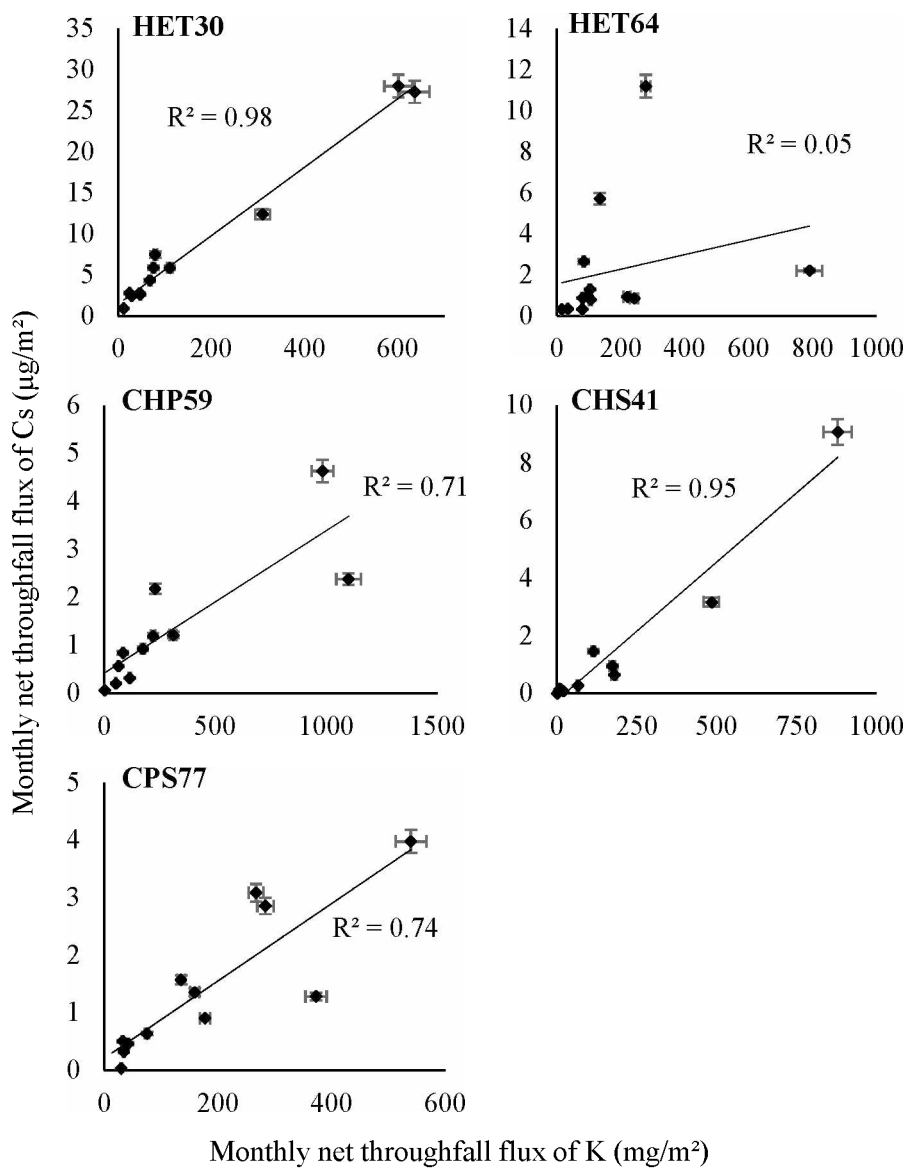


Figure S12. Monthly net throughfall flux of Cs as function of those of K for deciduous trees. Error bars correspond to standard errors. HET=Beech; CHP=Pedunculate Oak; CHS=Sessile Oak; CPS=Sessile/Ped.

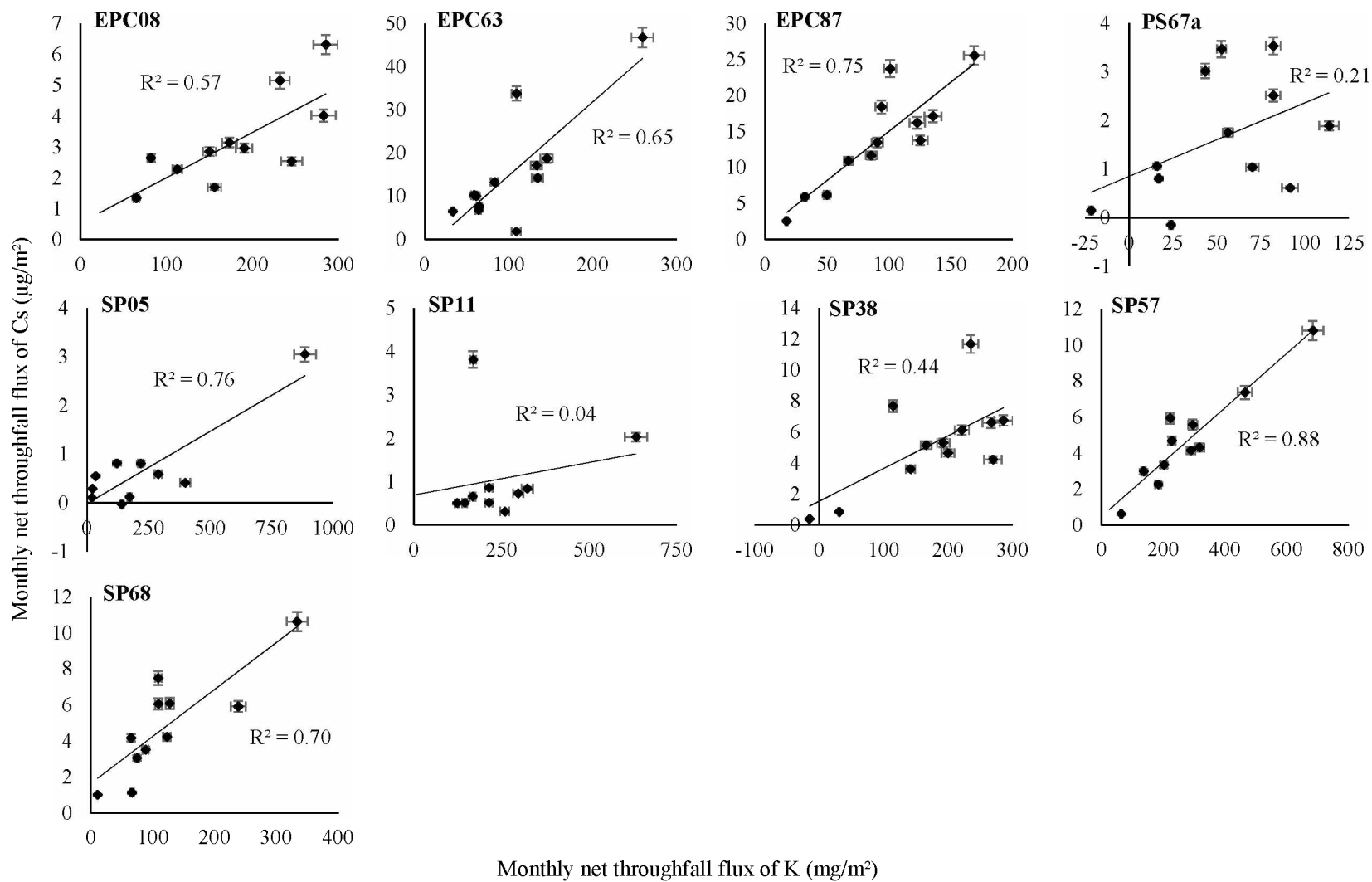


Figure S13. Monthly net throughfall flux of Cs as function of those of K for coniferous trees. Error bars correspond to standard errors. EPC=Spruce; PS=Scots Pine; SP=Silver fir.

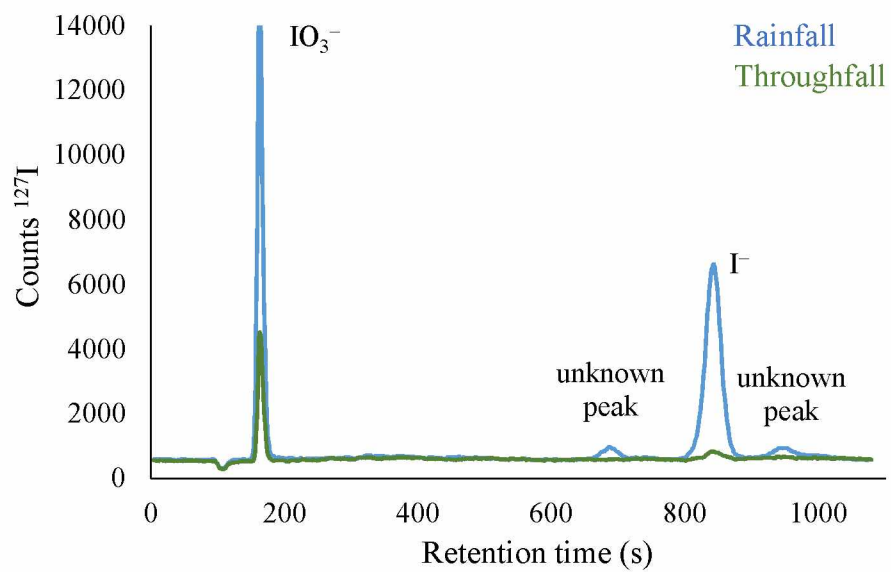


Figure S14. Example of IC-ICP-MS chromatograms of inorganic iodine species in rainfall and throughfall (sample EPC08, February 2017).

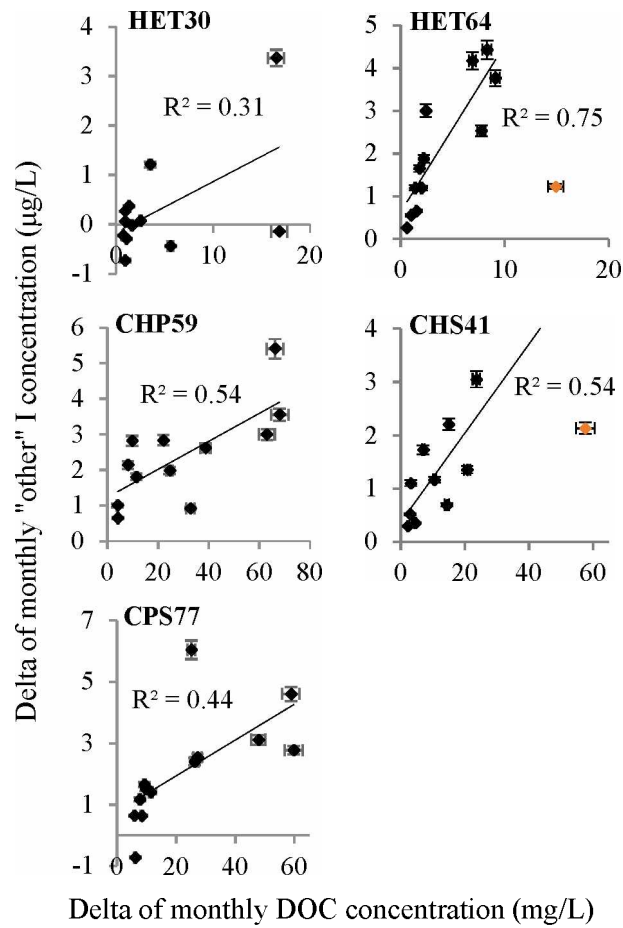


Figure S15. Delta of monthly “other” iodine concentration (= throughfall I concentration – rainfall I concentration) as function of those of DOC for deciduous trees. Orange dots correspond to excluded data. Error bars correspond to standard errors. HET=Beech; CHP=Pedunculate Oak; CHS=Sessile Oak; CPS=Sessile/Ped.

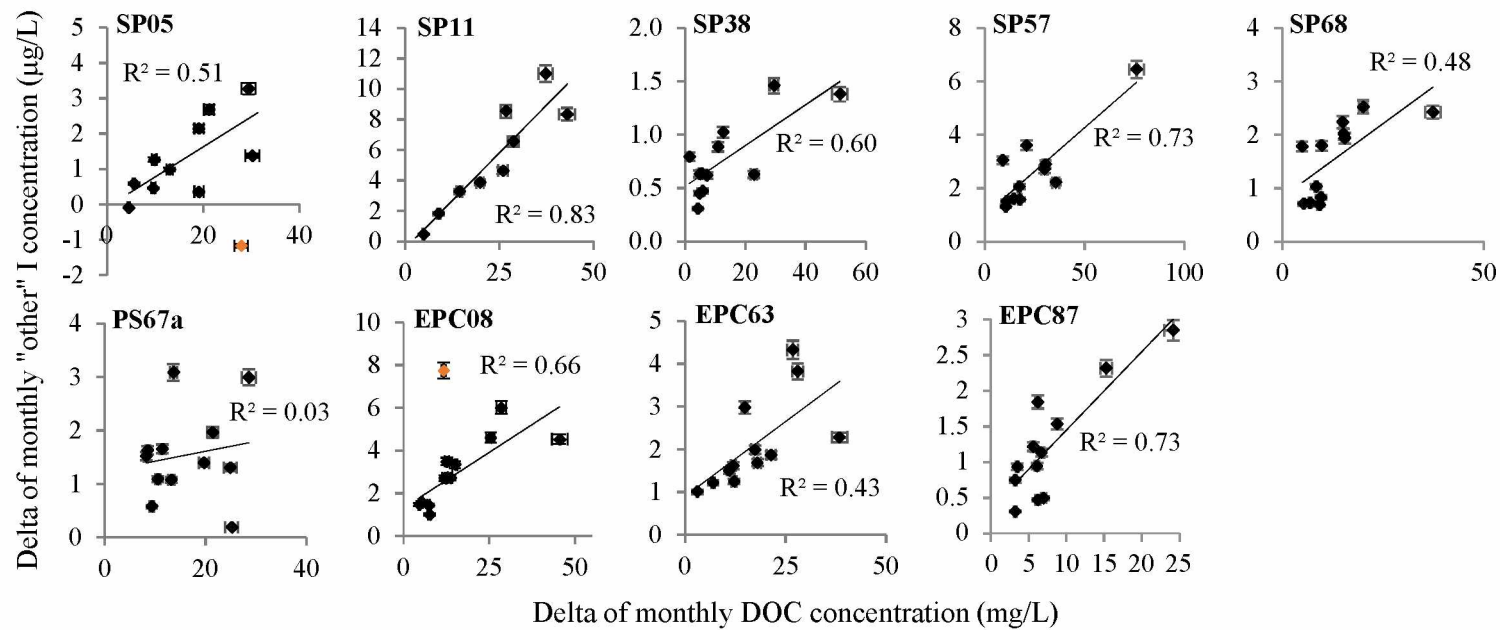


Figure S16. Delta of monthly "other" iodine concentration (= throughfall I concentration – rainfall I concentration) as function of those of DOC for coniferous trees. Orange dots correspond to excluded data. Error bars correspond to standard errors. EPC=Spruce; PS=Scots Pine; SP=Silver fir.

Table S1. Environmental conditions characteristics of studied sites (n = 14 sites).

Forest site*	Dominant tree species*	Functional tree types	Climate*	Tree height (m)**	Tree diameter (cm)**
CHP59	Pedunculate Oak	Deciduous	Oceanic	26.0	45.1
CHS41	Sessile Oak	Deciduous	Transition	26.6	42.2
CPS77	Sessile/Ped. Oak	Deciduous	Transition	30.0	49.2
EPC08	Spruce	Coniferous	Transition	27.6	38.4
EPC63	Spruce	Coniferous	Mountainous	27.5	35.5
EPC87	Spruce	Coniferous	Transition	23.9	31.1
HET30	Beech	Deciduous	Mountainous	18.7	32.7
HET64	Beech	Deciduous	Mountainous	29.0	29.8
PS67a	Scots Pine	Coniferous	Continental	25.9	32.1
SP05	Silver fir	Coniferous	Mountainous	30.9	36.9
SP11	Silver fir	Coniferous	Mountainous	32.4	47.1
SP38	Silver fir	Coniferous	Mountainous	29.9	41.5
SP57	Silver fir	Coniferous	Continental	28.4	40.8
SP68	Silver fir	Coniferous	Continental	32.6	51.5

\*: published in Redon et al. 2011; \*\*measured in 2014

Table S2. Annual water amount, annual pH mean, annual volume-weighted mean (AVWM) concentrations and concentration ratios (CR) of dissolved organic carbon (DOC), Na and K in rainfall and throughfall of studied sites (n = 14 sites). Uncertainties correspond to standard errors.

Forest site	Water amount (mm yr <sup>-1</sup> )		pH		AVWM DOC conc. (mg L <sup>-1</sup> )		CR(DOC)	AVWM Na <sup>+</sup> conc. (mg L <sup>-1</sup> )		CR(Na)	AVWM K <sup>+</sup> conc. (mg L <sup>-1</sup> )		CR(K)
	Rainfall*	Throughfall	Rainfall*	Throughfall	Rainfall*	Throughfall		Rainfall*	Throughfall		Rainfall*	Throughfall	
	CHP59	573	396	6.1 ± 0.6	6.5 ± 0.5	0.68 ± 0.03	25.8 ± 1.3	38 ± 3	0.62 ± 0.03	1.39 ± 0.07	2.2 ± 0.2	0.35 ± 0.02	9.86 ± 0.49
CHS41	608	388	6.1 ± 0.3	6.3 ± 0.4	0.58 ± 0.03	20.4 ± 1.0	35 ± 3	0.53 ± 0.03	1.21 ± 0.06	2.3 ± 0.2	0.12 ± 0.01	5.96 ± 0.30	49 ± 3
CPS77	673	416	6.0 ± 0.3	6.1 ± 0.5	0.50 ± 0.03	21.8 ± 1.1	44 ± 3	0.31 ± 0.02	1.00 ± 0.05	3.2 ± 0.2	0.13 ± 0.01	5.42 ± 0.27	41 ± 3
EPC08	882	678	6.0 ± 0.5	6.2 ± 0.4	0.37 ± 0.02	14.2 ± 0.7	38 ± 3	0.44 ± 0.02	1.51 ± 0.08	3.4 ± 0.2	0.08 ± 0.00	3.21 ± 0.16	41 ± 3
EPC63	993	631	6.2 ± 0.4	5.9 ± 0.4	0.25 ± 0.01	17.6 ± 0.9	70 ± 4	0.26 ± 0.01	1.16 ± 0.06	4.4 ± 0.3	0.16 ± 0.01	2.22 ± 0.11	14 ± 1
EPC87	1218	701	5.8 ± 0.2	5.8 ± 0.3	0.40 ± 0.02	7.9 ± 0.4	20 ± 1	0.53 ± 0.03	1.35 ± 0.07	2.6 ± 0.2	0.10 ± 0.01	1.77 ± 0.09	17 ± 1
HET30	2372	2041	5.9 ± 0.6	6.1 ± 0.7	0.12 ± 0.01	2.0 ± 0.1	17 ± 2	0.62 ± 0.03	0.76 ± 0.04	1.2 ± 0.1	0.07 ± 0.00	1.10 ± 0.05	15 ± 1
HET64	1365	868	6.2 ± 0.3	6.6 ± 0.5	0.30 ± 0.02	3.6 ± 0.2	12 ± 1	0.82 ± 0.04	1.50 ± 0.08	1.8 ± 0.1	0.11 ± 0.01	2.78 ± 0.14	25 ± 2
PS67a	619	371	6.2 ± 0.7	6.0 ± 0.5	0.61 ± 0.03	17.0 ± 0.8	28 ± 2	0.26 ± 0.01	0.83 ± 0.04	3.2 ± 0.2	0.24 ± 0.01	2.09 ± 0.10	9 ± 1
SP05	1032	639	6.4 ± 0.5	6.7 ± 0.4	0.55 ± 0.03	17.2 ± 0.9	31 ± 2	0.20 ± 0.01	0.41 ± 0.02	2.1 ± 0.2	0.15 ± 0.01	5.65 ± 0.28	37 ± 3
SP11	945	653	5.9 ± 0.3	6.2 ± 0.3	0.55 ± 0.03	14.8 ± 0.7	27 ± 2	0.37 ± 0.02	1.65 ± 0.08	4.5 ± 0.3	0.08 ± 0.00	4.71 ± 0.24	57 ± 4
SP38	1301	943	6.0 ± 0.5	6.1 ± 0.7	0.32 ± 0.02	11.7 ± 0.6	37 ± 3	0.21 ± 0.01	0.29 ± 0.01	1.4 ± 0.1	0.17 ± 0.01	2.66 ± 0.13	15 ± 1
SP57	1029	514	5.6 ± 0.5	5.8 ± 0.6	0.41 ± 0.02	22.0 ± 1.1	54 ± 4	0.22 ± 0.01	0.97 ± 0.05	4.4 ± 0.3	0.09 ± 0.00	6.29 ± 0.31	72 ± 5
SP68	931	411	5.7 ± 0.4	6.3 ± 0.3	0.27 ± 0.01	10.1 ± 0.5	37 ± 2	0.22 ± 0.01	0.58 ± 0.03	2.7 ± 0.2	0.07 ± 0.00	4.10 ± 0.21	56 ± 4
<b>Mean</b>	<b>1039</b>	<b>689</b>	<b>6.0</b>	<b>6.2</b>	<b>0.42</b>	<b>15</b>	<b>35</b>	<b>0.40</b>	<b>1.04</b>	<b>2.8</b>	<b>0.14</b>	<b>4.13</b>	<b>34</b>
<b>Median</b>	<b>969</b>	<b>635</b>	<b>6.0</b>	<b>6.2</b>	<b>0.40</b>	<b>16</b>	<b>36</b>	<b>0.34</b>	<b>1.08</b>	<b>2.6</b>	<b>0.12</b>	<b>3.66</b>	<b>32</b>
<b>Min</b>	<b>573</b>	<b>371</b>	<b>5.6</b>	<b>5.8</b>	<b>0.12</b>	<b>2</b>	<b>12</b>	<b>0.20</b>	<b>0.29</b>	<b>1.2</b>	<b>0.07</b>	<b>1.10</b>	<b>9</b>
<b>Max</b>	<b>2372</b>	<b>2041</b>	<b>6.4</b>	<b>6.7</b>	<b>0.68</b>	<b>26</b>	<b>70</b>	<b>0.82</b>	<b>1.65</b>	<b>4.5</b>	<b>0.35</b>	<b>9.86</b>	<b>72</b>

\*published in Roulier et al. submitted



Table S3. Annual volume-weighted mean (AVWM) concentrations and concentration ratios (CR) of total iodine, selenium and caesium, and proportions of I and Se species in rainfall and throughfall of studied sites (n = 14 sites). Uncertainties correspond to standard errors.

Forest site	AVWM I conc. ( $\mu\text{g L}^{-1}$ )								CR(I)
	Rainfall*				Throughfall				
	I	$\Gamma$ (%)	$\text{IO}_3^-$ (%)	$\text{I}_{\text{other}}$ (%)	I	$\Gamma$ (%)	$\text{IO}_3^-$ (%)	$\text{I}_{\text{other}}$ (%)	
CHP59	1.74 ± 0.07	19 ± 1	30 ± 2	51 ± 4	3.19 ± 0.10	11 ± 1	3.0 ± 0.1	86 ± 4	1.8 ± 0.1
CHS41	1.68 ± 0.06	21 ± 1	27 ± 1	52 ± 4	2.98 ± 0.07	16 ± 1	4.9 ± 0.2	79 ± 3	1.8 ± 0.1
CPS77	1.23 ± 0.05	25 ± 1	24 ± 1	51 ± 4	3.32 ± 0.08	10 ± 1	3.1 ± 0.1	87 ± 3	2.7 ± 0.1
EPC08	1.42 ± 0.05	27 ± 1	32 ± 2	41 ± 4	3.54 ± 0.10	9.2 ± 0.5	2.8 ± 0.1	88 ± 4	2.5 ± 0.1
EPC63	0.99 ± 0.04	31 ± 2	19 ± 2	50 ± 5	2.73 ± 0.08	7 ± 1	1 ± 1	92 ± 4	2.8 ± 0.1
EPC87	1.35 ± 0.05	34 ± 2	27 ± 1	39 ± 4	2.68 ± 0.07	25 ± 1	4.4 ± 0.2	71 ± 3	2.0 ± 0.1
HET30	2.18 ± 0.06	23 ± 1	17 ± 1	60 ± 4	1.87 ± 0.05	22 ± 1	13 ± 1	65 ± 4	0.9 ± 0.0
HET64	1.99 ± 0.08	25 ± 1	30 ± 1	44 ± 5	4.07 ± 0.10	21 ± 1	12 ± 1	67 ± 3	2.1 ± 0.1
PS67a	1.23 ± 0.05	35 ± 2	19 ± 1	46 ± 4	2.31 ± 0.07	14 ± 1	2.4 ± 0.1	84 ± 4	1.9 ± 0.1
SP05	1.20 ± 0.05	25 ± 1	21 ± 1	54 ± 5	1.79 ± 0.05	3.2 ± 0.3	6.2 ± 0.3	91 ± 4	1.5 ± 0.1
SP11	1.24 ± 0.05	26 ± 1	21 ± 1	53 ± 5	4.97 ± 0.14	14 ± 1	0.98 ± 0.04	85 ± 4	4.0 ± 0.2
SP38	0.94 ± 0.04	30 ± 2	17 ± 1	53 ± 5	1.37 ± 0.04	7.3 ± 0.4	3.7 ± 0.2	89 ± 4	1.4 ± 0.1
SP57	1.05 ± 0.04	44 ± 2	16 ± 1	40 ± 4	4.03 ± 0.08	16 ± 1	0.59 ± 0.03	83 ± 3	3.8 ± 0.2
SP68	0.85 ± 0.03	33 ± 2	18 ± 1	49 ± 5	1.98 ± 0.06	21 ± 1	3.5 ± 0.2	75 ± 4	2.3 ± 0.1
<b>Mean</b>	<b>1.36</b>	<b>29</b>	<b>23</b>	<b>49</b>	<b>2.92</b>	<b>14</b>	<b>4</b>	<b>82</b>	<b>2.2</b>
<b>Median</b>	<b>1.23</b>	<b>27</b>	<b>21</b>	<b>50</b>	<b>2.85</b>	<b>14</b>	<b>3</b>	<b>85</b>	<b>2.0</b>
<b>Min</b>	<b>0.85</b>	<b>19</b>	<b>16</b>	<b>39</b>	<b>1.37</b>	<b>3</b>	<b>1</b>	<b>65</b>	<b>0.9</b>
<b>Max</b>	<b>2.18</b>	<b>44</b>	<b>32</b>	<b>60</b>	<b>4.97</b>	<b>25</b>	<b>13</b>	<b>92</b>	<b>4.0</b>

Forest site	AVWM Se conc. ( $\text{ng L}^{-1}$ )				CR(Se)	AVWM Cs conc. ( $\text{ng L}^{-1}$ )		CR(Cs)	
	Rainfall*			Throughfall		Rainfall*	Throughfall		
	Se	$\text{SeO}_3^{2-}$ (%)	$\text{SeO}_4^{2-}$ (%)	$\text{Se}_{\text{other}}$ (%)					Se
CHP59	59 ± 4	13 ± 1	14 ± 1	74 ± 8	109 ± 8	1.9 ± 0.2	7.3 ± 0.5	54 ± 2	7 ± 1
CHS41	46 ± 3	8 ± 1	21 ± 2	72 ± 8	83 ± 6	1.8 ± 0.2	5.0 ± 0.4	52 ± 3	10 ± 1
CPS77	37 ± 3	9 ± 1	17 ± 2	74 ± 9	97 ± 7	2.6 ± 0.3	4.1 ± 0.3	51 ± 2	13 ± 1
EPC08	57 ± 4	10 ± 1	18 ± 2	72 ± 8	149 ± 11	2.6 ± 0.3	5.4 ± 0.4	65 ± 3	12 ± 1
EPC63	28 ± 2	11 ± 1	16 ± 1	73 ± 8	60 ± 5	2.1 ± 0.2	6.3 ± 0.4	310 ± 14	49 ± 4
EPC87	38 ± 2	10 ± 1	33 ± 3	57 ± 8	60 ± 5	1.6 ± 0.2	6.2 ± 0.4	268 ± 12	43 ± 3
HET30	85 ± 4	20 ± 2	34 ± 3	46 ± 6	71 ± 5	0.8 ± 0.1	5.0 ± 0.4	57 ± 3	11 ± 1
HET64	51 ± 3	6.9 ± 0.5	31 ± 2	62 ± 8	90 ± 7	1.8 ± 0.2	3.3 ± 0.2	38 ± 2	11 ± 1
PS67a	49 ± 3	8 ± 1	37 ± 3	55 ± 8	90 ± 6	1.8 ± 0.2	6.9 ± 0.4	70 ± 3	10 ± 1
SP05	23 ± 2	10 ± 1	20 ± 2	70 ± 9	31 ± 2	1.3 ± 0.1	4.1 ± 0.3	17 ± 1	4 ± 0
SP11	33 ± 2	8 ± 1	30 ± 3	62 ± 9	107 ± 8	3.3 ± 0.3	2.3 ± 0.3	25 ± 1	11 ± 1
SP38	31 ± 2	10 ± 1	26 ± 2	64 ± 9	58 ± 5	1.9 ± 0.2	5.4 ± 0.4	80 ± 3	15 ± 1
SP57	44 ± 3	8 ± 1	21 ± 2	72 ± 7	118 ± 8	2.7 ± 0.2	7.5 ± 0.5	136 ± 5	18 ± 1
SP68	31 ± 2	11 ± 1	31 ± 3	58 ± 9	56 ± 4	1.8 ± 0.2	13.2 ± 0.8	156 ± 7	12 ± 1
<b>Mean</b>	<b>44</b>	<b>16</b>	<b>25</b>	<b>59</b>	<b>84</b>	<b>2.0</b>	<b>5.9</b>	<b>98</b>	<b>16</b>
<b>Median</b>	<b>41</b>	<b>16</b>	<b>24</b>	<b>61</b>	<b>90</b>	<b>1.8</b>	<b>5.4</b>	<b>61</b>	<b>12</b>
<b>Min</b>	<b>23</b>	<b>10</b>	<b>14</b>	<b>45</b>	<b>31</b>	<b>0.8</b>	<b>2.3</b>	<b>17</b>	<b>4</b>
<b>Max</b>	<b>85</b>	<b>22</b>	<b>40</b>	<b>70</b>	<b>149</b>	<b>3.3</b>	<b>13.2</b>	<b>310</b>	<b>49</b>

\*published in Roulier et al. submitted

Table S4. Monthly water amount of rainfall and throughfall in study sites (n = 14 sites).

Forest site	Water amount (mm month <sup>-1</sup> )												
	Rainfall												
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17
CHP59	45	31	54	32	55	34	88	9	16	49	52	56	55
CHS41	35	13	71	25	23	38	72	18	66	66	71	61	51
CPS77	38	40	66	22	34	41	105	13	32	57	63	32	132
EPC08	15	35	88	30	63	84	134	13	43	96	89	71	122
EPC63	84	74	125	46	43	56	86	57	99	72	136	55	60
EPC87	107	73	159	18	45	90	157	49	96	135	143	90	58
HET30	176	314	403	208	35	604	149	149	228	35	44	20	8
HET64	98	63	198	15	148	85	123	135	51	110	137	107	97
PS67a	47	47	67	11	29	47	69	6	41	53	64	66	74
SP05	61	117	269	0	17	80	72	23	148	60	68	73	45
SP11	53	57	151	16	125	80	74	111	61	55	49	59	57
SP38	70	77	169	11	41	119	137	13	188	99	133	160	85
SP57	75	64	118	22	43	136	168	19	94	84	87	76	43
SP68	37	45	130	3	37	89	195	11	124	82	75	61	45
Forest site	Water amount (mm month <sup>-1</sup> )												
	Throughfall												
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17
CHP59	30	20	43	21	60	29	76	4	4	32	20	32	25
CHS41	23	6	59	3	19	24	48	2	51	49	47	33	25
CPS77	24	29	41	13	25	27	59	15	16	30	31	14	91
EPC08	9	22	75	22	59	74	107	5	29	59	67	50	100
EPC63	57	46	77	12	43	37	66	46	55	41	89	37	27
EPC87	30	49	97	7	30	41	99	33	54	80	95	51	35
HET30	144	234	396	169	38	553	142	128	145	30	40	15	7
HET64	48	28	115	9	92	56	82	90	25	74	107	82	61
PS67a	33	31	39	3	15	35	39	2	23	33	36	35	47
SP05	21	76	211	0	8	48	39	14	79	35	42	32	36
SP11	41	36	117	3	95	65	57	108	35	28	18	23	28
SP38	63	32	143	9	20	108	89	14	172	65	70	92	67
SP57	46	21	68	5	40	81	80	6	46	45	25	38	11
SP68	14	15	61	0	17	42	106	2	57	39	24	16	17

Table S5. Monthly iodine concentrations in rainfall and throughfall in study sites (n = 14 sites).

Forest site	monthly I conc. ( $\mu\text{g L}^{-1}$ )														
	Rainfall														
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17		
CHP59	0.08 ± 0.00	0.07 ± 0.00	0.11 ± 0.00	- ± -	0.08 ± 0.00	0.05 ± 0.00	0.10 ± 0.00	0.03 ± 0.00	0.07 ± 0.00	0.04 ± 0.00	0.07 ± 0.00	0.12 ± 0.01	0.08 ± 0.00		
CHS41	0.10 ± 0.00	- ± -	0.14 ± 0.00	0.03 ± 0.00	0.04 ± 0.00	0.03 ± 0.00	0.08 ± 0.00	0.02 ± 0.00	0.07 ± 0.00	0.07 ± 0.00	0.15 ± 0.01	0.11 ± 0.01	0.10 ± 0.00		
CPS77	0.07 ± 0.00	0.05 ± 0.00	0.12 ± 0.00	0.02 ± 0.00	0.05 ± 0.00	0.03 ± 0.00	0.09 ± 0.00	0.01 ± 0.00	0.04 ± 0.00	0.04 ± 0.00	0.07 ± 0.00	0.08 ± 0.00	0.14 ± 0.00		
EPC08	0.04 ± 0.00	- ± -	0.15 ± 0.00	0.06 ± 0.00	0.08 ± 0.00	0.10 ± 0.00	0.18 ± 0.01	0.03 ± 0.00	0.10 ± 0.00	0.07 ± 0.00	0.13 ± 0.01	0.09 ± 0.01	0.12 ± 0.00		
EPC63	0.11 ± 0.00	0.07 ± 0.00	0.12 ± 0.00	- ± -	0.04 ± 0.00	0.05 ± 0.00	0.04 ± 0.00	0.06 ± 0.00	0.08 ± 0.00	0.06 ± 0.00	0.15 ± 0.01	0.07 ± 0.00	0.05 ± 0.00		
EPC87	0.15 ± 0.00	0.11 ± 0.01	0.28 ± 0.01	0.02 ± 0.00	0.04 ± 0.00	0.06 ± 0.00	0.15 ± 0.01	0.03 ± 0.00	0.11 ± 0.00	0.12 ± 0.01	0.32 ± 0.02	0.15 ± 0.01	0.09 ± 0.00		
HET30	0.32 ± 0.00	0.49 ± 0.02	1.29 ± 0.04	0.64 ± 0.01	0.02 ± 0.00	1.23 ± 0.04	0.21 ± 0.01	0.17 ± 0.01	0.28 ± 0.01	0.02 ± 0.00	0.07 ± 0.00	0.05 ± 0.00	- ± -		
HET64	0.42 ± 0.01	0.14 ± 0.01	0.26 ± 0.01	0.03 ± 0.00	0.17 ± 0.01	0.07 ± 0.00	0.13 ± 0.01	0.17 ± 0.01	0.07 ± 0.00	0.24 ± 0.01	0.45 ± 0.02	0.32 ± 0.02	0.26 ± 0.01		
PS67a	0.06 ± 0.00	0.08 ± 0.00	0.13 ± 0.00	0.04 ± 0.00	0.03 ± 0.00	0.03 ± 0.00	0.04 ± 0.00	0.02 ± 0.00	0.05 ± 0.00	0.03 ± 0.00	0.06 ± 0.00	0.11 ± 0.01	0.06 ± 0.00		
SP05	0.09 ± 0.00	0.24 ± 0.01	0.26 ± 0.01	- ± -	0.01 ± 0.00	0.04 ± 0.00	0.07 ± 0.00	0.01 ± 0.00	0.17 ± 0.00	0.05 ± 0.00	0.11 ± 0.01	0.11 ± 0.01	0.08 ± 0.00		
SP11	0.13 ± 0.00	0.07 ± 0.00	0.22 ± 0.01	0.02 ± 0.00	0.08 ± 0.00	0.06 ± 0.00	0.05 ± 0.00	0.11 ± 0.00	0.05 ± 0.00	0.07 ± 0.00	0.10 ± 0.01	0.13 ± 0.01	0.08 ± 0.00		
SP38	0.07 ± 0.00	0.17 ± 0.01	0.12 ± 0.00	- ± -	0.02 ± 0.00	0.04 ± 0.00	0.12 ± 0.01	0.01 ± 0.00	0.12 ± 0.00	0.07 ± 0.00	0.15 ± 0.01	0.17 ± 0.01	0.07 ± 0.00		
SP57	0.11 ± 0.00	0.10 ± 0.00	0.13 ± 0.00	0.02 ± 0.00	0.04 ± 0.00	0.08 ± 0.00	0.10 ± 0.00	- ± -	0.11 ± 0.00	0.06 ± 0.00	0.10 ± 0.01	0.12 ± 0.01	0.05 ± 0.00		
SP68	0.03 ± 0.00	0.07 ± 0.00	0.11 ± 0.00	- ± -	0.04 ± 0.00	0.05 ± 0.00	0.08 ± 0.00	0.02 ± 0.00	0.08 ± 0.00	0.06 ± 0.00	0.07 ± 0.00	0.08 ± 0.00	0.05 ± 0.00		

Forest site	monthly I conc. ( $\mu\text{g L}^{-1}$ )														
	Throughfall														
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17		
CHP59	0.10 ± 0.00	- ± -	0.21 ± 0.00	0.07 ± 0.00	0.12 ± 0.00	0.08 ± 0.00	0.13 ± 0.01	0.02 ± 0.00	0.04 ± 0.00	0.13 ± 0.00	0.08 ± 0.00	0.09 ± 0.00	0.11 ± 0.00		
CHS41	0.10 ± 0.00	0.05 ± 0.00	0.26 ± 0.01	0.01 ± 0.00	0.04 ± 0.00	0.03 ± 0.00	0.06 ± 0.00	0.00 ± 0.00	0.14 ± 0.00	0.09 ± 0.00	0.17 ± 0.00	- ± -	0.12 ± 0.00		
CPS77	0.18 ± 0.00	0.11 ± 0.00	0.18 ± 0.00	0.02 ± 0.00	0.06 ± 0.00	0.05 ± 0.00	0.08 ± 0.00	- ± -	0.09 ± 0.00	0.10 ± 0.00	0.11 ± 0.00	0.06 ± 0.00	0.24 ± 0.01		
EPC08	0.07 ± 0.00	0.10 ± 0.00	0.29 ± 0.01	0.11 ± 0.00	0.13 ± 0.00	0.18 ± 0.01	0.23 ± 0.00	0.04 ± 0.00	0.17 ± 0.00	0.23 ± 0.00	0.34 ± 0.01	0.32 ± 0.01	0.19 ± 0.01		
EPC63	0.31 ± 0.01	0.11 ± 0.00	0.22 ± 0.01	0.03 ± 0.00	0.07 ± 0.00	0.08 ± 0.00	0.10 ± 0.00	0.11 ± 0.00	0.14 ± 0.00	0.09 ± 0.00	0.19 ± 0.01	0.15 ± 0.01	0.11 ± 0.00		
EPC87	0.12 ± 0.00	0.09 ± 0.00	0.29 ± 0.01	- ± -	0.07 ± 0.00	0.08 ± 0.00	0.16 ± 0.01	0.04 ± 0.00	0.15 ± 0.00	0.14 ± 0.00	0.31 ± 0.01	0.19 ± 0.00	0.10 ± 0.00		
HET30	0.36 ± 0.01	0.32 ± 0.01	0.76 ± 0.03	0.41 ± 0.01	0.03 ± 0.00	0.92 ± 0.03	0.27 ± 0.00	0.21 ± 0.00	0.20 ± 0.01	0.05 ± 0.00	0.19 ± 0.01	0.05 ± 0.00	0.05 ± 0.00		
HET64	0.40 ± 0.00	0.18 ± 0.00	0.36 ± 0.01	0.05 ± 0.00	0.15 ± 0.00	0.08 ± 0.00	0.17 ± 0.01	0.12 ± 0.00	0.09 ± 0.00	0.32 ± 0.01	0.74 ± 0.02	0.57 ± 0.02	0.31 ± 0.01		
PS67a	0.09 ± 0.00	0.05 ± 0.00	0.07 ± 0.00	0.01 ± 0.00	0.03 ± 0.00	0.06 ± 0.00	0.08 ± 0.00	0.01 ± 0.00	0.08 ± 0.00	0.04 ± 0.00	0.08 ± 0.00	0.17 ± 0.01	0.07 ± 0.00		
SP05	0.09 ± 0.00	- ± -	0.24 ± 0.01	- ± -	0.01 ± 0.00	0.01 ± 0.00	0.08 ± 0.00	0.01 ± 0.00	0.22 ± 0.01	0.05 ± 0.00	0.14 ± 0.01	- ± -	0.12 ± 0.00		
SP11	0.48 ± 0.01	0.21 ± 0.00	0.38 ± 0.01	- ± -	- ± -	- ± -	0.13 ± 0.00	0.13 ± 0.00	0.22 ± 0.01	0.12 ± 0.00	0.26 ± 0.01	0.31 ± 0.01	0.24 ± 0.01		
SP38	0.15 ± 0.00	0.11 ± 0.00	0.14 ± 0.00	- ± -	0.02 ± 0.00	0.07 ± 0.00	0.11 ± 0.00	0.02 ± 0.00	0.16 ± 0.00	0.06 ± 0.00	0.13 ± 0.00	0.15 ± 0.01	0.09 ± 0.00		
SP57	0.34 ± 0.00	0.08 ± 0.00	0.29 ± 0.01	- ± -	0.22 ± 0.00	0.16 ± 0.01	0.16 ± 0.00	- ± -	0.15 ± 0.00	0.10 ± 0.00	0.08 ± 0.00	0.13 ± 0.00	0.04 ± 0.00		
SP68	0.04 ± 0.00	0.07 ± 0.00	0.09 ± 0.00	- ± -	0.06 ± 0.00	0.06 ± 0.00	0.11 ± 0.01	0.01 ± 0.00	0.11 ± 0.00	0.07 ± 0.00	0.08 ± 0.00	0.06 ± 0.00	0.05 ± 0.00		

Table S6. Monthly selenium concentrations in rainfall and throughfall in study sites (n = 14 sites).

Forest site	monthly Se conc. (ng L <sup>-1</sup> )													
	Rainfall													
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17	
CHP59	3.0 ± 0.1	1.5 ± 0.1	3.5 ± 0.2	- ± -	4.1 ± 0.2	2.8 ± 0.1	2.3 ± 0.2	0.7 ± 0.1	1.9 ± 0.1	2.5 ± 0.2	2.7 ± 0.2	3.7 ± 0.3	2.5 ± 0.2	
CHS41	3.2 ± 0.1	- ± -	2.8 ± 0.2	0.6 ± 0.0	1.0 ± 0.1	1.6 ± 0.1	2.5 ± 0.1	0.6 ± 0.0	3.0 ± 0.1	1.9 ± 0.2	3.0 ± 0.2	2.1 ± 0.2	3.3 ± 0.3	
CPS77	2.8 ± 0.1	1.0 ± 0.1	2.0 ± 0.1	0.7 ± 0.0	0.9 ± 0.1	2.0 ± 0.1	3.1 ± 0.2	0.4 ± 0.0	1.9 ± 0.1	1.9 ± 0.2	2.0 ± 0.2	1.4 ± 0.1	4.8 ± 0.4	
EPC08	1.5 ± 0.0	- ± -	4.0 ± 0.2	2.4 ± 0.1	5.1 ± 0.3	6.2 ± 0.3	5.2 ± 0.2	1.3 ± 0.1	5.8 ± 0.2	3.8 ± 0.3	5.6 ± 0.5	3.4 ± 0.3	4.7 ± 0.4	
EPC63	4.7 ± 0.2	1.1 ± 0.1	1.7 ± 0.2	- ± -	1.1 ± 0.1	1.3 ± 0.1	1.5 ± 0.1	2.5 ± 0.2	2.8 ± 0.1	1.5 ± 0.1	3.5 ± 0.3	2.2 ± 0.2	2.0 ± 0.2	
EPC87	5.2 ± 0.2	2.0 ± 0.1	4.9 ± 0.4	0.2 ± 0.0	0.9 ± 0.1	2.4 ± 0.2	6.0 ± 0.3	1.5 ± 0.1	3.6 ± 0.1	5.7 ± 0.4	5.6 ± 0.5	4.5 ± 0.3	3.7 ± 0.3	
HET30	12.7 ± 0.4	12.5 ± 0.8	39.6 ± 1.6	15.9 ± 0.9	0.6 ± 0.0	77.4 ± 3.6	8.8 ± 0.4	6.3 ± 0.5	8.1 ± 0.4	0.4 ± 0.0	1.8 ± 0.2	1.7 ± 0.1	- ± -	
HET64	9.4 ± 0.4	3.8 ± 0.3	2.2 ± 0.2	0.5 ± 0.0	2.5 ± 0.2	3.1 ± 0.2	4.4 ± 0.2	3.4 ± 0.3	2.4 ± 0.1	6.5 ± 0.6	10.5 ± 0.8	12.2 ± 0.9	8.7 ± 0.6	
PS67a	2.9 ± 0.1	2.2 ± 0.1	2.1 ± 0.1	5.0 ± 0.3	1.7 ± 0.1	1.7 ± 0.1	1.1 ± 0.1	0.5 ± 0.0	2.7 ± 0.1	2.4 ± 0.2	2.4 ± 0.2	3.5 ± 0.3	5.0 ± 0.4	
SP05	2.1 ± 0.1	2.4 ± 0.1	2.8 ± 0.3	- ± -	0.2 ± 0.0	1.0 ± 0.1	2.3 ± 0.2	0.4 ± 0.0	3.7 ± 0.2	1.7 ± 0.1	3.4 ± 0.3	2.3 ± 0.2	1.8 ± 0.2	
SP11	3.2 ± 0.1	1.6 ± 0.1	3.4 ± 0.2	0.9 ± 0.1	2.4 ± 0.2	1.8 ± 0.1	1.7 ± 0.1	3.4 ± 0.2	1.8 ± 0.1	2.5 ± 0.2	2.8 ± 0.2	3.5 ± 0.3	2.1 ± 0.2	
SP38	2.5 ± 0.1	3.1 ± 0.2	1.9 ± 0.1	- ± -	1.0 ± 0.0	1.3 ± 0.1	4.0 ± 0.2	0.6 ± 0.1	4.1 ± 0.3	3.9 ± 0.3	4.4 ± 0.4	7.0 ± 0.6	3.6 ± 0.3	
SP57	6.5 ± 0.2	2.8 ± 0.2	1.8 ± 0.1	0.6 ± 0.0	2.9 ± 0.2	5.2 ± 0.3	2.7 ± 0.1	- ± -	7.3 ± 0.3	3.6 ± 0.3	2.6 ± 0.3	3.2 ± 0.2	2.7 ± 0.2	
SP68	1.2 ± 0.1	1.5 ± 0.1	1.3 ± 0.2	- ± -	2.2 ± 0.1	2.8 ± 0.3	2.2 ± 0.2	0.5 ± 0.0	5.0 ± 0.3	2.4 ± 0.2	2.9 ± 0.2	2.9 ± 0.2	2.1 ± 0.1	
Forest site	monthly Se conc. (ng L <sup>-1</sup> )													
	Throughfall													
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17	
CHP59	3.9 ± 0.2	- ± -	5.1 ± 0.3	3.0 ± 0.2	4.4 ± 0.3	2.3 ± 0.2	3.1 ± 0.2	0.6 ± 0.1	1.1 ± 0.1	5.9 ± 0.5	4.8 ± 0.4	2.3 ± 0.2	3.5 ± 0.3	
CHS41	3.8 ± 0.2	1.0 ± 0.1	3.9 ± 0.3	0.3 ± 0.0	1.0 ± 0.1	1.0 ± 0.1	1.8 ± 0.1	0.1 ± 0.0	4.1 ± 0.3	2.7 ± 0.3	6.9 ± 0.6	- ± -	3.0 ± 0.3	
CPS77	5.3 ± 0.3	2.9 ± 0.2	3.0 ± 0.2	0.7 ± 0.0	1.3 ± 0.1	1.6 ± 0.1	2.9 ± 0.2	- ± -	2.2 ± 0.2	3.6 ± 0.3	4.8 ± 0.4	1.2 ± 0.1	7.9 ± 0.7	
EPC08	2.6 ± 0.2	3.8 ± 0.2	8.2 ± 0.5	3.3 ± 0.2	5.8 ± 0.4	9.3 ± 0.6	8.0 ± 0.5	1.3 ± 0.1	6.5 ± 0.5	11.3 ± 0.9	23.9 ± 2.0	7.5 ± 0.7	9.2 ± 0.8	
EPC63	7.4 ± 0.4	2.2 ± 0.2	2.9 ± 0.2	0.5 ± 0.0	1.2 ± 0.1	1.5 ± 0.1	1.9 ± 0.1	2.3 ± 0.2	2.6 ± 0.2	2.0 ± 0.2	3.9 ± 0.4	6.8 ± 0.6	2.6 ± 0.2	
EPC87	3.6 ± 0.2	2.3 ± 0.2	4.2 ± 0.3	- ± -	1.3 ± 0.1	1.9 ± 0.1	3.5 ± 0.3	1.2 ± 0.1	3.7 ± 0.2	4.4 ± 0.4	5.7 ± 0.5	3.8 ± 0.3	3.0 ± 0.3	
HET30	16.3 ± 1.0	8.2 ± 0.5	23.3 ± 1.7	11.1 ± 0.7	1.2 ± 0.1	47.1 ± 3.0	12.2 ± 0.8	8.5 ± 0.7	5.8 ± 0.5	1.3 ± 0.1	7.0 ± 0.6	1.1 ± 0.1	1.6 ± 0.1	
HET64	6.3 ± 0.4	4.1 ± 0.3	4.4 ± 0.3	1.4 ± 0.1	2.4 ± 0.2	2.3 ± 0.2	4.6 ± 0.3	2.1 ± 0.2	2.2 ± 0.1	7.2 ± 0.6	18.1 ± 1.5	14.1 ± 1.2	8.4 ± 0.7	
PS67a	3.3 ± 0.2	1.4 ± 0.1	1.2 ± 0.1	0.5 ± 0.0	1.0 ± 0.1	2.9 ± 0.2	2.6 ± 0.2	- ± -	3.1 ± 0.2	2.5 ± 0.2	4.1 ± 0.3	4.8 ± 0.4	3.6 ± 0.4	
SP05	2.1 ± 0.0	- ± -	3.8 ± 0.3	- ± -	0.3 ± 0.0	0.3 ± 0.0	2.4 ± 0.1	0.4 ± 0.0	- ± -	1.6 ± 0.1	3.0 ± 0.3	- ± -	2.8 ± 0.3	
SP11	6.5 ± 0.4	4.2 ± 0.3	5.6 ± 0.4	- ± -	- ± -	- ± -	11.6 ± 0.7	4.9 ± 0.4	3.4 ± 0.3	3.0 ± 0.3	3.7 ± 0.3	5.8 ± 0.5	5.0 ± 0.4	
SP38	3.2 ± 0.2	2.1 ± 0.1	2.5 ± 0.2	- ± -	0.5 ± 0.0	2.4 ± 0.2	4.0 ± 0.3	0.5 ± 0.1	8.6 ± 0.6	3.1 ± 0.3	14.0 ± 1.2	5.8 ± 0.5	4.0 ± 0.4	
SP57	9.6 ± 0.3	2.5 ± 0.2	5.1 ± 0.3	- ± -	7.0 ± 0.4	5.1 ± 0.3	4.0 ± 0.3	- ± -	4.4 ± 0.4	4.5 ± 0.4	3.0 ± 0.3	4.4 ± 0.4	1.5 ± 0.1	
SP68	1.8 ± 0.1	1.4 ± 0.1	2.2 ± 0.2	- ± -	1.6 ± 0.1	1.9 ± 0.1	- ± -	0.3 ± 0.0	4.3 ± 0.3	2.6 ± 0.2	2.8 ± 0.2	2.1 ± 0.2	1.9 ± 0.2	

Table S7. Monthly caesium concentrations in rainfall and throughfall in study sites (n = 14 sites).

Forest site	monthly Cs conc. (ng L <sup>-1</sup> )															
	Rainfall															
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17			
CHP 59	0.24 ± 0.01	0.34 ± 0.02	0.38 ± 0.03	- ± -	0.39 ± 0.02	0.29 ± 0.03	0.38 ± 0.05	0.07 ± 0.00	0.47 ± 0.03	0.15 ± 0.01	0.18 ± 0.02	0.60 ± 0.03	0.38 ± 0.02			
CHS 41	0.25 ± 0.02	- ± -	0.43 ± 0.04	0.12 ± 0.01	0.13 ± 0.01	0.12 ± 0.01	0.24 ± 0.03	0.08 ± 0.01	0.29 ± 0.03	0.13 ± 0.02	0.19 ± 0.02	0.56 ± 0.03	0.28 ± 0.02			
CPS 77	0.24 ± 0.01	0.19 ± 0.01	0.29 ± 0.02	0.08 ± 0.00	0.18 ± 0.01	0.17 ± 0.01	0.32 ± 0.03	0.08 ± 0.01	0.32 ± 0.03	0.17 ± 0.02	0.13 ± 0.01	0.24 ± 0.01	0.34 ± 0.03			
EPC 08	0.11 ± 0.01	- ± -	0.42 ± 0.04	0.21 ± 0.01	0.36 ± 0.02	0.60 ± 0.04	0.35 ± 0.06	0.15 ± 0.01	0.74 ± 0.06	0.15 ± 0.02	0.55 ± 0.02	0.38 ± 0.03	0.41 ± 0.02			
EPC 63	0.87 ± 0.03	0.81 ± 0.05	0.30 ± 0.03	- ± -	0.24 ± 0.02	0.17 ± 0.01	0.23 ± 0.02	0.34 ± 0.03	0.77 ± 0.07	0.65 ± 0.04	0.53 ± 0.04	0.61 ± 0.05	0.32 ± 0.02			
EPC 87	0.65 ± 0.03	0.55 ± 0.05	0.44 ± 0.04	0.10 ± 0.01	0.19 ± 0.01	0.37 ± 0.03	0.70 ± 0.07	0.18 ± 0.01	1.14 ± 0.09	1.07 ± 0.05	0.58 ± 0.03	1.04 ± 0.04	0.51 ± 0.02			
HET 30	0.72 ± 0.05	1.14 ± 0.10	1.17 ± 0.13	1.58 ± 0.10	0.15 ± 0.01	3.59 ± 0.30	0.80 ± 0.09	0.70 ± 0.07	0.58 ± 0.07	0.13 ± 0.01	0.22 ± 0.01	0.22 ± 0.01	- ± -			
HET 64	0.51 ± 0.04	0.34 ± 0.02	0.23 ± 0.03	0.06 ± 0.00	0.22 ± 0.02	0.18 ± 0.01	0.35 ± 0.03	0.27 ± 0.03	0.26 ± 0.03	0.27 ± 0.03	0.53 ± 0.03	0.86 ± 0.05	0.42 ± 0.03			
PS 67a	0.20 ± 0.01	0.25 ± 0.03	0.20 ± 0.02	0.15 ± 0.01	0.17 ± 0.01	0.16 ± 0.01	0.31 ± 0.02	0.10 ± 0.00	0.61 ± 0.04	0.65 ± 0.04	0.26 ± 0.01	0.63 ± 0.03	0.60 ± 0.02			
SP 05	0.35 ± 0.01	0.44 ± 0.06	0.41 ± 0.05	- ± -	0.09 ± 0.01	0.07 ± 0.01	0.22 ± 0.02	0.07 ± 0.01	0.56 ± 0.07	0.41 ± 0.02	0.32 ± 0.03	1.11 ± 0.05	0.18 ± 0.01			
SP 11	0.13 ± 0.01	0.17 ± 0.03	0.27 ± 0.04	0.06 ± 0.01	0.12 ± 0.01	0.08 ± 0.01	0.11 ± 0.02	0.28 ± 0.04	0.15 ± 0.02	0.11 ± 0.01	0.10 ± 0.01	0.40 ± 0.03	0.19 ± 0.02			
SP 38	0.20 ± 0.01	0.42 ± 0.04	0.44 ± 0.06	- ± -	0.54 ± 0.02	0.24 ± 0.03	0.99 ± 0.07	0.09 ± 0.01	1.03 ± 0.10	0.63 ± 0.04	0.56 ± 0.05	1.05 ± 0.05	0.30 ± 0.03			
SP 57	0.78 ± 0.03	0.49 ± 0.03	0.48 ± 0.05	0.15 ± 0.01	0.40 ± 0.03	0.66 ± 0.05	0.55 ± 0.07	- ± -	1.19 ± 0.10	0.59 ± 0.04	0.55 ± 0.03	0.72 ± 0.03	0.55 ± 0.03			
SP 68	0.83 ± 0.02	0.57 ± 0.04	0.59 ± 0.06	- ± -	0.38 ± 0.02	0.63 ± 0.05	0.74 ± 0.06	0.29 ± 0.02	2.81 ± 0.19	1.03 ± 0.05	0.90 ± 0.04	1.57 ± 0.05	1.01 ± 0.05			

Forest site	monthly Cs conc. (ng L <sup>-1</sup> )															
	Throughfall															
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17			
CHP 59	1.17 ± 0.07	- ± -	2.76 ± 0.15	1.59 ± 0.03	1.24 ± 0.05	0.50 ± 0.02	0.95 ± 0.06	0.14 ± 0.00	0.80 ± 0.05	4.79 ± 0.07	2.36 ± 0.14	1.81 ± 0.10	1.58 ± 0.07			
CHS 41	0.90 ± 0.06	0.42 ± 0.02	3.60 ± 0.21	0.16 ± 0.00	0.28 ± 0.01	0.27 ± 0.01	0.31 ± 0.02	0.03 ± 0.00	9.37 ± 0.60	1.08 ± 0.07	1.66 ± 0.06	- ± -	0.54 ± 0.03			
CPS 77	1.15 ± 0.07	1.48 ± 0.07	4.27 ± 0.25	0.41 ± 0.01	0.68 ± 0.03	0.63 ± 0.02	0.96 ± 0.04	- ± -	3.18 ± 0.10	3.26 ± 0.04	1.71 ± 0.04	0.28 ± 0.02	1.70 ± 0.12			
EPC 08	1.45 ± 0.08	1.76 ± 0.10	4.43 ± 0.26	1.91 ± 0.05	3.00 ± 0.08	3.57 ± 0.11	3.21 ± 0.18	- ± -	3.01 ± 0.09	5.30 ± 0.09	6.87 ± 0.29	2.92 ± 0.15	3.56 ± 0.20			
EPC 63	47.66 ± 2.61	14.11 ± 0.67	19.05 ± 1.07	2.68 ± 0.08	6.74 ± 0.40	10.26 ± 0.35	7.85 ± 0.40	7.12 ± 0.13	15.02 ± 0.90	17.80 ± 0.37	34.36 ± 1.47	2.45 ± 0.17	10.61 ± 0.40			
EPC 87	24.39 ± 1.31	14.01 ± 0.71	26.03 ± 1.39	- ± -	6.14 ± 0.24	11.29 ± 0.36	12.35 ± 0.54	2.74 ± 0.06	17.36 ± 1.82	14.83 ± 0.22	17.70 ± 0.30	19.46 ± 0.19	6.69 ± 0.31			
HET 30	13.08 ± 0.76	28.44 ± 1.69	29.15 ± 2.08	7.47 ± 0.20	1.05 ± 0.06	11.04 ± 0.56	3.54 ± 0.13	3.09 ± 0.13	3.19 ± 0.19	5.99 ± 0.18	4.52 ± 0.19	3.01 ± 0.13	2.07 ± 0.06			
HET 64	1.47 ± 0.06	1.25 ± 0.07	2.43 ± 0.16	0.90 ± 0.02	0.99 ± 0.07	1.05 ± 0.04	0.68 ± 0.04	0.57 ± 0.04	5.97 ± 0.39	2.91 ± 0.08	1.80 ± 0.07	12.05 ± 0.54	0.72 ± 0.09			
PS 67a	3.67 ± 0.20	0.86 ± 0.04	1.24 ± 0.08	0.30 ± 0.01	0.98 ± 0.04	2.67 ± 0.12	3.33 ± 0.12	- ± -	2.37 ± 0.19	2.54 ± 0.11	3.79 ± 0.06	0.48 ± 0.05	1.66 ± 0.10			
SP 05	0.48 ± 0.01	- ± -	3.46 ± 0.18	- ± -	0.38 ± 0.01	0.62 ± 0.04	1.04 ± 0.07	0.17 ± 0.01	0.97 ± 0.06	0.38 ± 0.03	1.14 ± 0.08	- ± -	0.78 ± 0.04			
SP 11	0.97 ± 0.07	0.48 ± 0.03	2.30 ± 0.13	- ± -	- ± -	- ± -	0.97 ± 0.06	4.10 ± 0.11	0.65 ± 0.03	0.62 ± 0.02	0.75 ± 0.05	1.13 ± 0.07	0.70 ± 0.04			
SP 38	7.87 ± 0.43	4.64 ± 0.23	7.04 ± 0.40	- ± -	0.92 ± 0.04	5.40 ± 0.12	5.63 ± 0.20	0.94 ± 0.02	7.16 ± 0.45	4.23 ± 0.22	12.23 ± 0.10	7.80 ± 0.41	5.59 ± 0.34			
SP 57	8.15 ± 0.12	5.18 ± 0.28	11.28 ± 0.61	- ± -	6.34 ± 0.23	4.97 ± 0.19	4.71 ± 0.18	- ± -	3.45 ± 0.13	6.16 ± 0.13	3.54 ± 0.18	4.07 ± 0.20	1.17 ± 0.04			
SP 68	1.96 ± 0.11	- ± -	11.22 ± 0.59	- ± -	4.56 ± 0.20	4.85 ± 0.17	6.65 ± 0.28	1.30 ± 0.01	8.90 ± 0.61	7.09 ± 0.23	8.40 ± 0.36	5.09 ± 0.20	4.07 ± 0.17			

Table S8. Monthly sodium concentrations in rainfall and throughfall in study sites (n = 14 sites).

Forest site	monthly Na conc. (mg L <sup>-1</sup> )														
	Rainfall														
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17		
CHP59	0.46 ± 0.02	0.66 ± 0.03	0.55 ± 0.03	0.93 ± 0.05	0.70 ± 0.04	0.31 ± 0.02	0.78 ± 0.04	1.29 ± 0.06	1.30 ± 0.07	0.56 ± 0.03	0.33 ± 0.02	0.54 ± 0.03	0.60 ± 0.03		
CHS41	0.47 ± 0.02	0.69 ± 0.03	0.90 ± 0.05	0.32 ± 0.02	0.51 ± 0.03	0.90 ± 0.05	1.19 ± 0.06	0.52 ± 0.03	0.37 ± 0.02	0.27 ± 0.01	0.22 ± 0.01	0.27 ± 0.01	0.19 ± 0.01		
CPS77	0.22 ± 0.01	0.20 ± 0.01	0.49 ± 0.02	0.36 ± 0.02	0.66 ± 0.03	0.48 ± 0.02	0.50 ± 0.03	0.28 ± 0.01	0.28 ± 0.01	0.19 ± 0.01	0.25 ± 0.01	0.29 ± 0.01	0.09 ± 0.00		
EPC08	0.79 ± 0.04	0.35 ± 0.02	0.36 ± 0.02	1.01 ± 0.05	0.71 ± 0.04	0.35 ± 0.02	0.97 ± 0.05	0.81 ± 0.04	0.51 ± 0.03	0.17 ± 0.01	0.17 ± 0.01	0.26 ± 0.01	0.14 ± 0.01		
EPC63	0.18 ± 0.01	0.20 ± 0.01	0.19 ± 0.01	0.78 ± 0.04	0.59 ± 0.03	0.53 ± 0.03	0.46 ± 0.02	0.23 ± 0.01	0.16 ± 0.01	0.16 ± 0.01	0.12 ± 0.01	0.22 ± 0.01	0.12 ± 0.01		
EPC87	0.21 ± 0.01	0.20 ± 0.01	0.41 ± 0.02	0.77 ± 0.04	0.59 ± 0.03	1.28 ± 0.06	1.24 ± 0.06	0.20 ± 0.01	0.41 ± 0.02	0.25 ± 0.01	0.31 ± 0.02	0.52 ± 0.03	0.30 ± 0.02		
HET30	0.24 ± 0.01	0.27 ± 0.01	0.91 ± 0.05	0.63 ± 0.03	0.31 ± 0.02	0.79 ± 0.04	1.23 ± 0.06	0.41 ± 0.02	0.25 ± 0.01	0.18 ± 0.01	0.37 ± 0.02	0.75 ± 0.04	1.17 ± 0.06		
HET64	1.25 ± 0.06	0.34 ± 0.02	0.56 ± 0.03	0.33 ± 0.02	0.75 ± 0.04	3.09 ± 0.15	0.79 ± 0.04	0.67 ± 0.03	0.49 ± 0.02	0.39 ± 0.02	0.61 ± 0.03	0.68 ± 0.03	0.70 ± 0.04		
PS67a	0.17 ± 0.01	0.32 ± 0.02	2.10 ± 0.11	11.18 ± 0.56	0.43 ± 0.02	0.31 ± 0.02	0.28 ± 0.01	0.67 ± 0.03	0.31 ± 0.02	0.11 ± 0.01	0.15 ± 0.01	0.27 ± 0.01	0.22 ± 0.01		
SP05	0.25 ± 0.01	0.25 ± 0.01	0.08 ± 0.00	- ± 0.00	0.16 ± 0.01	0.13 ± 0.01	0.40 ± 0.02	0.07 ± 0.00	0.17 ± 0.01	0.31 ± 0.02	0.36 ± 0.02	0.22 ± 0.01	0.18 ± 0.01		
SP11	0.49 ± 0.02	0.13 ± 0.01	0.48 ± 0.02	0.63 ± 0.03	0.37 ± 0.02	0.54 ± 0.03	0.55 ± 0.03	0.25 ± 0.01	0.14 ± 0.01	0.17 ± 0.01	0.44 ± 0.02	0.38 ± 0.02	0.23 ± 0.01		
SP38	0.09 ± 0.00	0.15 ± 0.01	0.07 ± 0.00	- ± 0.00	0.78 ± 0.04	0.06 ± 0.00	0.58 ± 0.03	0.08 ± 0.00	0.07 ± 0.00	0.17 ± 0.01	0.34 ± 0.02	0.13 ± 0.01	0.07 ± 0.00		
SP57	0.11 ± 0.01	0.21 ± 0.01	0.13 ± 0.01	0.38 ± 0.02	0.43 ± 0.02	0.27 ± 0.01	0.29 ± 0.01	0.61 ± 0.03	0.16 ± 0.01	0.17 ± 0.01	0.08 ± 0.00	0.26 ± 0.01	0.22 ± 0.01		
SP68	0.09 ± 0.00	0.21 ± 0.01	0.19 ± 0.01	- ± 0.00	0.33 ± 0.02	0.20 ± 0.01	0.34 ± 0.02	0.32 ± 0.02	0.08 ± 0.00	0.18 ± 0.01	0.11 ± 0.01	0.18 ± 0.01	0.13 ± 0.01		

Forest site	monthly Na conc. (mg L <sup>-1</sup> )														
	Throughfall														
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17		
CHP59	0.60 ± 0.03	1.21 ± 0.06	1.52 ± 0.08	2.06 ± 0.10	1.10 ± 0.06	1.27 ± 0.06	1.56 ± 0.08	1.83 ± 0.09	2.91 ± 0.15	2.26 ± 0.11	1.39 ± 0.07	0.91 ± 0.05	1.13 ± 0.06		
CHS41	0.77 ± 0.04	1.17 ± 0.06	1.39 ± 0.07	1.19 ± 0.06	0.78 ± 0.04	1.57 ± 0.08	2.36 ± 0.12	1.05 ± 0.05	1.37 ± 0.07	0.65 ± 0.03	0.46 ± 0.02	1.51 ± 0.08	0.76 ± 0.04		
CPS77	0.65 ± 0.03	1.26 ± 0.06	1.57 ± 0.08	0.93 ± 0.05	1.18 ± 0.06	1.44 ± 0.07	1.32 ± 0.07	0.86 ± 0.04	2.12 ± 0.11	0.97 ± 0.05	0.62 ± 0.03	0.98 ± 0.05	0.36 ± 0.02		
EPC08	2.63 ± 0.13	1.78 ± 0.09	1.17 ± 0.06	3.24 ± 0.16	1.54 ± 0.08	2.10 ± 0.11	1.79 ± 0.09	3.86 ± 0.19	2.46 ± 0.12	1.45 ± 0.07	1.20 ± 0.06	1.30 ± 0.07	0.48 ± 0.02		
EPC63	1.03 ± 0.05	0.57 ± 0.03	0.88 ± 0.04	1.18 ± 0.06	0.48 ± 0.02	2.25 ± 0.11	2.07 ± 0.10	1.86 ± 0.09	1.29 ± 0.06	0.90 ± 0.05	0.73 ± 0.04	1.27 ± 0.06	0.87 ± 0.04		
EPC87	0.91 ± 0.05	0.44 ± 0.02	0.80 ± 0.04	2.01 ± 0.10	1.29 ± 0.06	3.71 ± 0.19	3.16 ± 0.16	0.85 ± 0.04	1.38 ± 0.07	0.70 ± 0.04	0.84 ± 0.04	0.83 ± 0.04	0.69 ± 0.03		
HET30	0.35 ± 0.02	0.30 ± 0.02	0.79 ± 0.04	0.69 ± 0.03	0.33 ± 0.02	0.74 ± 0.04	2.38 ± 0.12	0.80 ± 0.04	0.38 ± 0.02	0.56 ± 0.03	1.06 ± 0.05	1.00 ± 0.05	1.27 ± 0.06		
HET64	1.70 ± 0.09	1.01 ± 0.05	1.12 ± 0.06	5.81 ± 0.29	0.81 ± 0.04	5.99 ± 0.30	1.82 ± 0.09	0.72 ± 0.04	1.49 ± 0.07	1.02 ± 0.05	0.99 ± 0.05	1.35 ± 0.07	1.07 ± 0.05		
PS67a	0.84 ± 0.04	0.75 ± 0.04	0.42 ± 0.02	1.65 ± 0.08	1.12 ± 0.06	1.39 ± 0.07	1.21 ± 0.06	3.30 ± 0.17	0.91 ± 0.05	0.44 ± 0.02	0.97 ± 0.05	0.87 ± 0.04	0.35 ± 0.02		
SP05	0.80 ± 0.04	0.81 ± 0.04	0.12 ± 0.01	- ± 0.00	0.57 ± 0.03	0.22 ± 0.01	0.58 ± 0.03	0.29 ± 0.01	0.34 ± 0.02	0.26 ± 0.01	0.84 ± 0.04	0.94 ± 0.05	0.38 ± 0.02		
SP11	2.88 ± 0.14	1.92 ± 0.10	1.41 ± 0.07	1.42 ± 0.07	0.63 ± 0.03	2.34 ± 0.12	2.38 ± 0.12	0.54 ± 0.03	1.75 ± 0.09	1.40 ± 0.07	3.35 ± 0.17	3.72 ± 0.19	2.56 ± 0.13		
SP38	0.41 ± 0.02	0.78 ± 0.04	0.15 ± 0.01	- ± 0.00	0.20 ± 0.01	0.16 ± 0.01	0.54 ± 0.03	0.55 ± 0.03	0.17 ± 0.01	0.23 ± 0.01	0.30 ± 0.02	0.24 ± 0.01	0.18 ± 0.01		
SP57	0.89 ± 0.04	1.09 ± 0.05	0.74 ± 0.04	1.29 ± 0.06	2.20 ± 0.11	0.86 ± 0.04	0.94 ± 0.05	1.49 ± 0.07	0.81 ± 0.04	0.69 ± 0.03	0.74 ± 0.04	0.93 ± 0.05	0.92 ± 0.05		
SP68	0.44 ± 0.02	0.96 ± 0.05	0.34 ± 0.02	- ± 0.00	0.97 ± 0.05	0.68 ± 0.03	0.66 ± 0.03	1.37 ± 0.07	0.45 ± 0.02	0.44 ± 0.02	0.67 ± 0.03	0.76 ± 0.04	0.52 ± 0.03		

Table S9. Monthly potassium concentrations in rainfall and throughfall in study sites (n = 14 sites).

Forest site	monthly K conc. (mg L <sup>-1</sup> )													
	Rainfall													
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17	
CHP59	0.28 ± 0.01	0.21 ± 0.01	0.21 ± 0.01	2.01 ± 0.10	0.06 ± 0.00	0.06 ± 0.00	0.12 ± 0.01	0.81 ± 0.04	2.09 ± 0.10	0.37 ± 0.02	0.17 ± 0.01	0.31 ± 0.02	0.13 ± 0.01	
CHS41	0.14 ± 0.01	0.33 ± 0.02	0.15 ± 0.01	0.13 ± 0.01	0.06 ± 0.00	0.19 ± 0.01	0.10 ± 0.01	0.12 ± 0.01	0.11 ± 0.01	0.09 ± 0.00	0.18 ± 0.01	0.07 ± 0.00	0.07 ± 0.00	
CPS77	0.15 ± 0.01	0.08 ± 0.00	0.38 ± 0.02	0.14 ± 0.01	0.07 ± 0.00	0.06 ± 0.00	0.06 ± 0.00	0.23 ± 0.01	0.24 ± 0.01	0.21 ± 0.01	0.09 ± 0.00	0.14 ± 0.01	0.06 ± 0.00	
EPC08	0.29 ± 0.01	0.08 ± 0.00	0.05 ± 0.00	0.20 ± 0.01	0.06 ± 0.00	0.00 ± 0.00	0.06 ± 0.00	0.15 ± 0.01	0.34 ± 0.02	0.05 ± 0.00	0.13 ± 0.01	0.09 ± 0.00	0.00 ± 0.00	
EPC63	0.13 ± 0.01	0.44 ± 0.02	0.05 ± 0.00	0.64 ± 0.03	0.00 ± 0.00	0.08 ± 0.00	0.08 ± 0.00	0.12 ± 0.01	0.11 ± 0.01	0.17 ± 0.01	0.12 ± 0.01	0.12 ± 0.01	0.21 ± 0.01	
EPC87	0.06 ± 0.00	0.09 ± 0.00	0.06 ± 0.00	0.22 ± 0.01	0.00 ± 0.00	0.11 ± 0.01	0.09 ± 0.00	0.09 ± 0.00	0.16 ± 0.01	0.13 ± 0.01	0.07 ± 0.00	0.25 ± 0.01	0.13 ± 0.01	
HET30	0.05 ± 0.00	0.07 ± 0.00	0.08 ± 0.00	0.06 ± 0.00	0.00 ± 0.00	0.07 ± 0.00	0.11 ± 0.01	0.07 ± 0.00	0.00 ± 0.00	0.10 ± 0.01	0.21 ± 0.01	0.32 ± 0.02	1.38 ± 0.07	
HET64	0.13 ± 0.01	0.44 ± 0.02	0.06 ± 0.00	0.14 ± 0.01	0.05 ± 0.00	0.20 ± 0.01	0.10 ± 0.01	0.09 ± 0.00	0.17 ± 0.01	0.08 ± 0.00	0.08 ± 0.00	0.13 ± 0.01	0.09 ± 0.00	
PS67a	0.12 ± 0.01	0.16 ± 0.01	0.26 ± 0.01	3.09 ± 0.15	0.06 ± 0.00	0.11 ± 0.01	0.06 ± 0.00	0.36 ± 0.02	0.19 ± 0.01	0.08 ± 0.00	0.10 ± 0.01	0.20 ± 0.01	0.52 ± 0.03	
SP05	0.43 ± 0.02	0.15 ± 0.01	0.05 ± 0.00	- ± 0.00	0.00 ± 0.00	0.08 ± 0.00	0.07 ± 0.00	0.08 ± 0.00	0.10 ± 0.01	0.26 ± 0.01	0.47 ± 0.02	0.10 ± 0.01	0.16 ± 0.01	
SP11	0.23 ± 0.01	0.08 ± 0.00	0.08 ± 0.00	0.32 ± 0.02	0.00 ± 0.00	0.07 ± 0.00	0.09 ± 0.00	0.05 ± 0.00	0.09 ± 0.00	0.09 ± 0.00	0.17 ± 0.01	0.06 ± 0.00	0.07 ± 0.00	
SP38	0.07 ± 0.00	0.17 ± 0.01	0.00 ± 0.00	- ± 0.00	0.74 ± 0.04	0.00 ± 0.00	0.12 ± 0.01	0.10 ± 0.01	0.19 ± 0.01	0.44 ± 0.02	0.32 ± 0.02	0.07 ± 0.00	0.10 ± 0.01	
SP57	0.08 ± 0.00	0.09 ± 0.00	0.05 ± 0.00	0.28 ± 0.01	0.06 ± 0.00	0.06 ± 0.00	0.00 ± 0.00	0.25 ± 0.01	0.11 ± 0.01	0.16 ± 0.01	0.10 ± 0.01	0.15 ± 0.01	0.17 ± 0.01	
SP68	0.09 ± 0.00	0.15 ± 0.01	0.00 ± 0.00	- ± 0.00	0.00 ± 0.00	0.08 ± 0.00	0.00 ± 0.00	0.21 ± 0.01	0.10 ± 0.01	0.10 ± 0.01	0.11 ± 0.01	0.15 ± 0.01	0.12 ± 0.01	
Forest site	monthly K conc. (mg L <sup>-1</sup> )													
	Throughfall													
	Sept.16	Oct.16	Nov.16	Dec.16	Jan.17	Febr.17	March.17	Apr.17	May.17	June.17	July.17	Aug.17	Sept.17	
CHP59	6.27 ± 0.31	11.61 ± 0.58	25.79 ± 1.29	9.38 ± 0.47	1.47 ± 0.07	1.88 ± 0.09	1.00 ± 0.05	2.61 ± 0.13	34.07 ± 1.70	31.60 ± 1.58	11.98 ± 0.60	10.12 ± 0.51	9.24 ± 0.46	
CHS41	8.19 ± 0.41	17.90 ± 0.90	8.40 ± 0.42	2.12 ± 0.11	0.49 ± 0.02	0.73 ± 0.04	0.55 ± 0.03	1.16 ± 0.06	17.28 ± 0.86	3.70 ± 0.19	2.71 ± 0.14	6.22 ± 0.31	2.82 ± 0.14	
CPS77	7.71 ± 0.39	13.15 ± 0.66	13.78 ± 0.69	2.92 ± 0.15	1.38 ± 0.07	1.63 ± 0.08	1.39 ± 0.07	1.11 ± 0.06	17.68 ± 0.88	9.44 ± 0.47	4.48 ± 0.22	2.40 ± 0.12	1.83 ± 0.09	
EPC08	8.13 ± 0.41	5.32 ± 0.27	3.82 ± 0.19	7.43 ± 0.37	1.46 ± 0.07	2.56 ± 0.13	1.47 ± 0.07	5.20 ± 0.26	4.41 ± 0.22	4.04 ± 0.20	4.40 ± 0.22	5.02 ± 0.25	1.73 ± 0.09	
EPC63	4.77 ± 0.24	2.51 ± 0.13	1.97 ± 0.10	2.12 ± 0.11	0.78 ± 0.04	1.76 ± 0.09	1.08 ± 0.05	1.53 ± 0.08	2.65 ± 0.13	3.56 ± 0.18	1.41 ± 0.07	3.15 ± 0.16	2.63 ± 0.13	
EPC87	3.60 ± 0.18	1.99 ± 0.10	1.83 ± 0.09	4.30 ± 0.22	1.08 ± 0.05	1.86 ± 0.09	1.01 ± 0.05	0.66 ± 0.03	2.56 ± 0.13	1.79 ± 0.09	1.53 ± 0.08	2.26 ± 0.11	1.63 ± 0.08	
HET30	2.22 ± 0.11	2.81 ± 0.14	1.60 ± 0.08	0.52 ± 0.03	0.31 ± 0.02	0.22 ± 0.01	0.44 ± 0.02	0.31 ± 0.02	0.33 ± 0.02	3.83 ± 0.19	1.92 ± 0.10	2.05 ± 0.10	5.00 ± 0.25	
HET64	2.26 ± 0.11	8.93 ± 0.45	6.95 ± 0.35	27.73 ± 1.39	1.22 ± 0.06	1.74 ± 0.09	0.56 ± 0.03	0.30 ± 0.02	5.82 ± 0.29	1.26 ± 0.06	1.07 ± 0.05	3.54 ± 0.18	1.44 ± 0.07	
PS67a	1.74 ± 0.09	3.16 ± 0.16	2.21 ± 0.11	4.01 ± 0.20	1.22 ± 0.06	2.52 ± 0.13	1.23 ± 0.06	2.38 ± 0.12	2.79 ± 0.14	3.60 ± 0.18	2.46 ± 0.12	1.06 ± 0.05	1.15 ± 0.06	
SP05	9.58 ± 0.48	11.25 ± 0.56	4.27 ± 0.21	- ± 0.00	2.79 ± 0.14	0.87 ± 0.04	3.23 ± 0.16	1.55 ± 0.08	5.25 ± 0.26	4.48 ± 0.22	5.93 ± 0.30	1.69 ± 0.08	8.28 ± 0.41	
SP11	8.21 ± 0.41	7.34 ± 0.37	5.50 ± 0.28	3.67 ± 0.18	1.37 ± 0.07	5.06 ± 0.25	3.85 ± 0.19	1.61 ± 0.08	4.30 ± 0.22	4.58 ± 0.23	9.87 ± 0.49	12.81 ± 0.64	7.90 ± 0.40	
SP38	1.90 ± 0.10	8.72 ± 0.44	1.86 ± 0.09	- ± 0.00	0.77 ± 0.04	1.54 ± 0.08	2.43 ± 0.12	2.38 ± 0.12	1.49 ± 0.07	2.86 ± 0.14	3.98 ± 0.20	3.22 ± 0.16	2.99 ± 0.15	
SP57	10.21 ± 0.51	11.17 ± 0.56	10.17 ± 0.51	5.73 ± 0.29	5.61 ± 0.28	4.01 ± 0.20	3.63 ± 0.18	6.02 ± 0.30	4.17 ± 0.21	6.85 ± 0.34	5.73 ± 0.29	5.56 ± 0.28	6.23 ± 0.31	
SP68	5.13 ± 0.26	10.48 ± 0.52	5.44 ± 0.27	- ± 0.00	3.79 ± 0.19	3.10 ± 0.16	2.25 ± 0.11	6.47 ± 0.32	2.44 ± 0.12	3.03 ± 0.15	4.80 ± 0.24	5.93 ± 0.30	4.69 ± 0.23	