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# **Influence of tree species on selenium and iodine partitioning in an experimental forest ecosystem**

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## 1 **Abstract**

2 Storage of selenium and iodine can greatly vary between forest ecosystems, but the influence  
3 of tree species on partitioning and recycling of those elements remains elusive. In this study,  
4 contents of Se and I were measured in tree compartments, litterfall, humus, and soil horizons  
5 in monospecific stands of Douglas fir, pine, spruce, beech, and oak under identical climatic  
6 and edaphic conditions. The cycle of each element was characterized in terms of stocks and  
7 fluxes. Lowest concentrations were in wood (Se: 8 - 13  $\mu\text{g kg}^{-1}$ ; I: < 16.5  $\mu\text{g kg}^{-1}$ ). Senescing  
8 organs had higher Se and I content, than the living parts of trees due to direct exposure to  
9 atmospheric deposition, with some variation between coniferous and deciduous trees. For all  
10 stands, low amounts of Se and I were involved in biological cycle as reflected by low root  
11 uptake. In humus, the enrichment of elements greatly increased with the stage of organic  
12 matter (OM) degradation with average factors of 10 and 20 for Se and I. OM degradation and  
13 element persistence in humus was influenced by tree species. Deciduous trees, with low  
14 biomass, and fast degradation of OM stored less Se and I in humus compared to fir and  
15 spruce with high humus biomass. Interestingly, tree species did not affect soil reserves of Se  
16 and I. Concentration ranges were 331-690  $\mu\text{g Se kg}^{-1}$  and 4.3 - 14.5  $\text{mg I kg}^{-1}$ . However, the  
17 divergent vertical profiles of the elements in the soil column indicated greater mobility of I.  
18 Selenium concentrations regularly decreased with depth in correlation with OM and Fe  
19 oxides content. For iodine, the maximum iodine concentration at a soil depth of 15 to 35 cm  
20 was caused by a parallel precipitation/sorption behavior of aluminium and organic iodine  
21 dissolved in the topsoil.

22

23

## 24 **1. Introduction**

25 Selenium and iodine are omnipresent in the environment and essential elements for  
26 mammals. Forests which cover ~33% of land in Europe (Alberdi et al., 2015) can absorb,  
27 recycle, and accumulate these elements. Tree canopies interact with dry and wet deposition  
28 (De Schrijver et al., 2007) and in throughfall concentrations and speciation relative to that  
29 deposited may be changed (Roulier et al., 2020a). Selenium and iodine can remain on the leaf  
30 surface and/or be integrated into the biomass through foliar absorption (Cakmak et al., 2017;  
31 Kikkert and Berkelaar, 2013; Roulier et al., 2020b). After interaction with foliar surfaces,  
32 these elements are deposited to topsoil via precipitation and litterfall, and subsequently  
33 recycled through root uptake (Kato et al., 2013; Li et al., 2008). Decomposers of decaying  
34 material (earthworm, fungi and bacteria communities) influence the biological, physical, and  
35 chemical properties of soils, thus affecting cycling of elements, e.g. C, N, K, Ca, Mg, and Na,  
36 in forest (Aponte et al., 2013; Berger and Berger, 2012; Mareschal et al., 2010; Prescott and  
37 Grayston, 2013). It has been demonstrated that both elements strongly interact with the  
38 organic and mineral soil phases (Coppin et al., 2009; Pisarek et al., 2021; Roulier et al., 2019;  
39 Tolu et al., 2014; Xu et al., 2016). Previous studies assessed the influence of different forest  
40 types on the distribution of Se and I and showed similar contents in litterfall, while  
41 differences were observed for humus and soil (Pisarek et al., 2021; Roulier et al., 2019).  
42 However, in these studies the different tree species, climate, and geochemical conditions  
43 made it difficult to determine the dominant variables influencing observations (Pisarek et al.,  
44 2021; Roulier et al., 2019). In this study we postulated that the quantitative description of Se  
45 and I cycle in monospecific forest stands developing under identical climatic and edaphic  
46 conditions may help in characterizing the influence of tree vegetation.. For that purpose, the  
47 concentrations of Se and I were quantified in the aboveground tree compartments (wood,  
48 bark, branches, leaves), litterfall (leaves, branches), forest floor/humus and soil horizons for

49 five tree species: Douglas fir, pine, spruce, beech and oak. The objectives were, for the five  
50 tree species, (i) to identify the differences in Se and I distribution and recycling, (ii) to  
51 establish Se and I retention times and accumulation rates in humus, (iii) to investigate  
52 differences in Se and I soil profiles.

## 53 **2. Methods**

### 54 **2.1 Study site**

55 The study site is situated in the forest of Breuil-Chenu in Bourgogne, France (lat 47°18'10"  
56 N; long 4°4'44" E). The climate is temperate with a warm summer without a dry season  
57 (Cornelis et al., 2010). The experimental site, managed by INRAE-BEF, is part of the  
58 ANAEE France Platform ([www.anaee-france.fr](http://www.anaee-france.fr) - breuil). In 1975 the original coppice forest  
59 was removed. Monospecific plots (each 1000 m<sup>2</sup>) were then planted in 1976 with five tree  
60 species: i) coniferous: Douglas fir (*Pseudotsuga menziesii*), Corsican pine (*P. nigra* var.  
61 *corsicana*), Norway spruce (*Picea abies*); ii) deciduous: European beech (*Fagus sylvatica*),  
62 sessile oak (*Quercus sessiliflora*) (Figure SI-1). The soil is an acid brown soil, classified as  
63 Alumnic Cambisol (IUSS Working Group WRB, 2006), derived from granite and very poor  
64 in major cations (0.5% magnesium oxide, 0.6% calcium oxide and 4.4% potassium oxide)  
65 (Montelius et al., 2015). All the introduced tree species decreased soil base saturation and  
66 increased acidification. Douglas fir, Corsican pine and secondarily oak strongly amplified the  
67 acidification process. In these two stands the major soil process shifted from weak  
68 acidification and crypto podzolisation to strong soil acidification (Legout et al., 2016). Soil  
69 characteristics under each tree stands are presented in Table SI-1.

### 70 **2.2 Sampling**

71 One composite sample of wood, bark, and branches from 10 trees of each stand collected in  
72 2001 was used. Annual composite samples of green leaves, litterfall-leaves and litterfall-

73 branches were obtained from 2002 to 2006. Humus samples included 8 replicates of bulk  
74 humus (sampled in 2005) for each stand and single samples of four humus horizons  
75 (collected in 2006): *oln* (organic, litter, new), *olv* (organic, litter, old), *of* (organic,  
76 fragmented) and *oh* (organic, humified). For each stand, the mineral soil profiles were  
77 sampled in 2006 at three sampling points at the following depths (cm): 0 – 7.5; 7.5 – 15; 15 –  
78 30; 30 – 45; 45 – 60 and 60 – 75. Only the soil profile of the oak stand was collected at one  
79 sampling point and different depths: 0 – 5; 5 – 10; 10 – 15; 15 – 25; 25 – 40; 40 – 55; 55 – 70  
80 cm. For comparison, concentrations in oak soil were thus calculated for horizons similar to  
81 those used for the other soils. Soils were sieved to < 2 mm and dried. The 2 - 5 mm soil  
82 fraction of the deepest layer (60 – 75 cm, Douglas fir) was considered to be representative of  
83 the soil parent material. All samples of plant material, litterfall, humus, and soils were dried  
84 and grounded.

#### 85 **2.4 Total element extraction and determination**

86 Acidic extraction was used to extract total Se. Briefly, soil samples (~0.25 g) were mixed  
87 with 0.5 mL of 38% HCl and 1.5 mL of 68% HNO<sub>3</sub>, while humus and litterfall (~0.25 g) were  
88 mixed with 2 mL of 68% HNO<sub>3</sub>, 0.5 mL of 30 % H<sub>2</sub>O<sub>2</sub> and 3 mL of ultrapure water (Milli-Q  
89 System, 18.2 MΩ cm; Elix, Millipore). Samples were heated at 90 °C for 3 hours in hot block  
90 digestion system (DigiPREP MS, SCP Science). Digested samples were diluted to 50 mL  
91 with ultrapure water, filtered on 0.45 μm acetate membrane and stored at 4 °C until analysis.  
92 Blanks were added in each digestion run.

93 Total iodine was extracted with a previously established protocol (Roulier et al., 2018; Watts  
94 et al., 2015; Watts and Mitchell, 2009). Approximately 0.2 g of soil or humus, or 0.4 g of  
95 plant material were mixed with 5 mL of 5 % tetramethylammonium hydroxide (TMAH).  
96 Digestion was performed in hot block digestion system at 85 °C for 3 hours. Digested  
97 samples were diluted to 10 mL with Milli-Q water, centrifuged at 4 000 rpm for 10 min,

98 filtered on 0.45  $\mu\text{m}$  acetate membrane and stored at 4  $^{\circ}\text{C}$  until analysis. Before analysis,  
99 digested soils and humus were diluted 50-fold and plant materials 25-fold. Blanks were added  
100 in each digestion run.

101 Selenium and iodine concentrations were determined by inductively coupled plasma mass  
102 spectrometry (ICP-MS; 7500ce, Tokyo, Japan). For Se analysis, collision/reaction cell gas  
103 flow was 5  $\text{mL min}^{-1}$   $\text{H}_2$ . When sample replicates were available (i.e. for bulk humus  
104 ( $n=8/\text{stand}$ ), soil ( $n=3/\text{depth}/\text{stand}$ ), annual samples of litterfall and leaves ( $n=5/\text{stand}$ )), each  
105 replicate was digested and analyzed once. Results are given as the mean value with the  
106 combined uncertainties of the analysis and sampling (weighted by the biomass produced  
107 during the year for foliage and litterfall). When only one sample was available (i.e. for wood,  
108 bark, branch, and humus horizons), it was digested and analyzed in triplicate and result is  
109 given as the mean value with the analytical uncertainty of the mean. Total Se quantification  
110 of Certified Reference Materials ( $118 \pm 9 \mu\text{g kg}^{-1}$  for DC 73032,  $191 \pm 15 \mu\text{g kg}^{-1}$  for DC  
111 73030,  $24 \pm 3 \mu\text{g kg}^{-1}$  for ERM-CD281,  $23 \pm 2 \mu\text{g kg}^{-1}$  for BCR-129) agreed well with the  
112 certified values ( $140 \pm 20 \mu\text{g kg}^{-1}$  for DC 73032,  $200 \pm 30 \mu\text{g kg}^{-1}$  for DC 73030,  $23 \pm 4 \mu\text{g}$   
113  $\text{kg}^{-1}$  for ERM-CD281, 25  $\mu\text{g kg}^{-1}$  (information value) for BCR-129). Furthermore,  
114 experimental results of iodine concentrations on certified materials ( $9.2 \pm 0.3 \text{mg kg}^{-1}$  for DC  
115 73022,  $6.3 \pm 0.1 \text{mg kg}^{-1}$  for DC 73030 and  $137 \pm 11 \mu\text{g kg}^{-1}$  for BCR-129) confirmed the  
116 accuracy of the methods (certified values:  $9.4 \pm 1.2 \text{mg kg}^{-1}$  for DC 73022,  $6.4 \pm 0.5 \text{mg kg}^{-1}$   
117 for DC 73030,  $167 \pm 24 \mu\text{g kg}^{-1}$  for BCR-129). The instrumental limit of detection (LOD)  
118 depended from daily sensitivity and varied between 0.01-0.03  $\mu\text{g L}^{-1}$  for selenium and  
119 between 0.02-0.08  $\mu\text{g L}^{-1}$  for iodine, leading to detection limit in solid samples of 1.6  $\mu\text{g kg}^{-1}$   
120 for Se and 8.5-16.5  $\mu\text{g kg}^{-1}$  for I.

## 121 **2.4 Calculations**

122 The biomass values of standing tree compartments (needles, branches, bole-bark, bole-wood)  
123 were taken from Montelius et al. (2015). The biomass of broadleaves was estimated at  
124 121.6% of litterfall mass to account for the average loss of mass of living foliage during  
125 senescence (Vergutz et al., 2012). The total masses of humus ( $\text{kg ha}^{-1}$ ) were estimated from  
126 the weighing of a reference surface of  $0.5 \text{ m}^2$  (Mareschal et al., 2010). Annual litterfall ( $\text{kg}$   
127  $\text{ha}^{-1} \text{ yr}^{-1}$ ) was estimated using 5 collectors covering a total of  $2.5 \text{ m}^2$  (collected every 3  
128 months, from 2002 to 2006). The annual biomass production was calculated as the average of  
129 annual increments of standing biomass over the period 2001 - 2006. Biomass pools and  
130 annual productivities are presented in Table SI-2 and Table SI-3.

131 The stocks of elements in each soil layer ( $\text{g ha}^{-1}$ ) were calculated by multiplying the element  
132 concentrations by the bulk density and the thickness of the soil layer. Element stocks in tree  
133 compartments and humus ( $\text{g ha}^{-1}$ ) were estimated by multiplying element concentrations by  
134 the corresponding biomass pool. Annual fluxes through litterfall ( $\text{g ha}^{-1} \text{ yr}^{-1}$ ) and production  
135 of biomass from tree compartments ( $\text{g ha}^{-1} \text{ yr}^{-1}$ ) were estimated by multiplying the element  
136 concentrations by average annual productivities. As iodine was not detected in wood  
137 compartment ( $< 16.5 \mu\text{g kg}^{-1}$ ), a value of half the detection limit with an uncertainty of 100%  
138 was used for the calculation of iodine stock and annual immobilization in wood.

139 The cycle of elements in tree biomass was estimated with three main annual fluxes:  
140 requirement (R); uptake (U) and translocation (T) according to equations presented in Table  
141 1. This model, with some modifications, has already been used for Se, I and many other  
142 elements (Di Tullo, 2015; Goor and Thiry, 2004; Ranger and Colin-Belgrand, 1996; Roulier  
143 et al., 2018). Requirement includes the total element pool implemented during annual  
144 aboveground biomass production. Uptake corresponds to the amount of element absorbed

145 from the soil by the roots calculated as the amount accumulated in tree compartments and  
146 returns of the element to soil through litterfall and crown leaching. Crown leaching was  
147 shown to have negligible influence on Se and I budgets (Di Tullo, 2015; Roulier et al., 2018),  
148 thus it was neglected in this model. Translocation reflects the internal element transfer from  
149 senescing foliage to the rest of the tree, calculated as the difference between the element  
150 content in living foliage and litterfall.

151 Selenium and iodine accumulation rates, X acc.rate, in humus were calculated according to  
152 Equation 1 from Redon et al., 2011:

$$153 \quad \mathbf{X \text{ acc. rate}} = \frac{([\mathbf{X}]_{\text{humus}} - [\mathbf{X}]_{\text{litter}}) \times \mathbf{DM}_{\text{humus}}}{t_{\text{resDM}}} \quad (\text{g ha}^{-1} \text{ yr}^{-1}) \quad (1)$$

154 Where  $[\mathbf{X}]_{\text{humus}}$  and  $[\mathbf{X}]_{\text{litter}}$  are Se or I concentrations in humus and litterfall.  $\mathbf{DM}_{\text{humus}}$  is the  
155 humus dry mass,  $t_{\text{resDM}}$  is the average dry matter residence time in humus.  $t_{\text{resDM}}$  was  
156 estimated according to Equation 2:

$$157 \quad \mathbf{t_{resDM}} = \frac{\mathbf{DM}_{\text{humus}}}{\mathbf{LF} \times (\mathbf{1} - \mathbf{\text{litter fraction mineralized}})} \quad (\text{years}) \quad (2)$$

158 where LF is the annual litterfall dry mass and ‘litter fraction mineralized’ is the average  
159 decrease in the mass of the litter through the transformation of OM (43% in oak, 65% in  
160 beech, 70% in Douglas fir, 61% in spruce, 85% in pine forests; from Osono and Takeda,  
161 2005).

## 162 **2.5 Statistical analysis**

163 The distribution of data was examined using the Shapiro-Wilk test. Pearson correlations and  
164 ANOVA test were performed for normally distributed data, while the Spearman correlation  
165 and the Kruskal-Wallis rank test were used for data that did not follow normal distribution.  
166 The t-test was performed in order to indicate if the difference in element concentration  
167 between soil layers and forest compartments of monospecific stands are significant. Tukey

168 test and pairwise Wilcoxon test were used to identify the contrast (indicated by letters) within  
169 sub-groups of categorical variables, respectively for normally and not normally distributed  
170 data. The apportionment of element concentrations within categorical sub-groups were  
171 represented with boxplots. Stocks and fluxes were compared between tree species based on t-  
172 tests. Statistical study was performed in R Studio (R Core Team, 2013; version 3.4.3).

### 173 **3. Results: Se and I contents in the compartments of the five soil-tree systems**

#### 174 **3.1 Tree compartments and litterfall**

175 Selenium and iodine concentrations in forest stands of Douglas fir, pine, spruce, beech and  
176 oak are presented in Table 2. The lowest concentrations of iodine ( $< 16.5 \mu\text{g kg}^{-1}$ , i.e. under  
177 LOD) and selenium ( $8\text{-}13 \mu\text{g kg}^{-1}$ ) were found in wood stem of all trees. Similarly, Zhao et  
178 al. (2019) found low I concentrations ( $< 6 \mu\text{g kg}^{-1}$  -  $100 \mu\text{g kg}^{-1}$ ) in spruce tree rings, the  
179 fluctuation of which was due to the variation of I content in the atmosphere over the years.  
180 The ranges of selenium concentrations in bole-bark ( $16\text{-}47 \mu\text{g kg}^{-1}$ ) and branches ( $18\text{-}37 \mu\text{g}$   
181  $\text{kg}^{-1}$ ) compartments were comparable. Selenium concentrations in beech foliage ( $43 \pm 5 \mu\text{g}$   
182  $\text{kg}^{-1}$ ) and bole-bark ( $47 \pm 5 \mu\text{g kg}^{-1}$ ) were slightly higher than those reported by Di Tullo  
183 (2015) in 54 years-old beech forest ( $30 \pm 1$  and  $28 \pm 6 \mu\text{g kg}^{-1}$ , respectively). Iodine contents  
184 in leaves ( $86\text{-}223 \mu\text{g kg}^{-1}$ ), branches ( $41\text{-}165 \mu\text{g kg}^{-1}$ ) and bole-barks ( $59 - 226 \mu\text{g kg}^{-1}$ )  
185 corresponded well to the concentrations reported in the literature ( $92 - 241$ ,  $16\text{-}186$ ,  $180\text{-}329$   
186  $\mu\text{g kg}^{-1}$ , respectively; from Korobova, 2010 and Roulier et al., 2018).

187 In woody compartments, differences were noticeable (t-test,  $p < 0.05$ ):

- 188 - For Se, in branches: [Douglas fir, spruce] > [oak, beech, pine] and, in bole-bark:  
189 [beech] > [Douglas fir] > [spruce] > [pine, oak],
- 190 - For I, in branches: [spruce] > [Douglas fir] > [pine] > [oak] > [beech] and, in bole-  
191 bark: [beech] > [Douglas fir] > [spruce, pine] > [oak].

192 In average, considering functional tree types, the five-year survey showed that leaves from  
193 deciduous trees were richer in Se and I (Se mean: 39  $\mu\text{g kg}^{-1}$ ; I mean: 212  $\mu\text{g kg}^{-1}$ ) than  
194 conifer needles (Se mean: 23  $\mu\text{g kg}^{-1}$ , I mean: 116  $\mu\text{g kg}^{-1}$ ). The lowest concentrations were  
195 found in spruce and pine needles, while concentrations found in Douglas fir needles were not  
196 statistically different from those in oak leaves (Table 2; Figures 1.1-1.2; Se, I: Kruskal-  
197 Wallis, p-value < 0.001). In litterfall-leaves, Se concentrations (range of 60-90  $\mu\text{g kg}^{-1}$ ) were  
198 higher under pine compared to spruce and oak (Figures 1.3; ANOVA, p-value < 0.001). Pine,  
199 spruce and oak had lower I concentration compared to Douglas fir (Figures 1.4; ANOVA p-  
200 value < 0.001). Unlike leaves, litterfall-branches of deciduous trees contained lower  
201 concentrations of elements (Se mean: 51  $\mu\text{g kg}^{-1}$ , I mean: 367  $\mu\text{g kg}^{-1}$ ) than that of  
202 coniferous trees (Se mean: 245  $\mu\text{g kg}^{-1}$ , I mean: 2346  $\mu\text{g kg}^{-1}$ ). Tree species effect was only  
203 noticeable for selenium with lower concentrations found in litterfall-branches under pine  
204 compared with spruce (Figures 1.5-1.6, Kruskal-Wallis, Se: p-value < 0.01, I: p-value <  
205 0.001).

### 206 **3.2 Humus**

207 Both element concentrations increased considerably from *oln* to *oh* horizon in the forest floor  
208 of each stand (Figure 2.1-2.2 and Table 2). On average, Se content increased from 74 to 751  
209  $\mu\text{g kg}^{-1}$  and I content from 0.27 to 5.5  $\text{mg kg}^{-1}$  resulting in average enrichment factors from  
210 upper to deeper humus layers of 10 and 20 for Se and I, respectively. Resulting selenium  
211 concentrations in bulk humus varied from 484  $\mu\text{g kg}^{-1}$  to 742  $\mu\text{g kg}^{-1}$  (Table 1), which is  
212 within the ranges of values of 57 to 1022  $\mu\text{g kg}^{-1}$  reported for deciduous forests and 108 to  
213 1608  $\mu\text{g kg}^{-1}$  for conifers (Di Tullo, 2015; Pisarek et al., 2021; Tyler, 2005). The range of  
214 iodine concentrations varied from 3.1 to 5.5  $\text{mg kg}^{-1}$  that was also in agreement with ranges  
215 of 1.0 - 5.5  $\text{mg kg}^{-1}$  for deciduous forests and 1.2 - 14  $\text{mg kg}^{-1}$  for coniferous forests reported

216 by Roulier et al. (2019), Takeda et al. (2015), and Bostock (2004). Oak and pine bulk humus  
217 showed the lowest Se concentrations ( $484 \pm 61 \mu\text{g kg}^{-1}$  for oak and  $500 \pm 98 \mu\text{g kg}^{-1}$  for  
218 pine), while Douglas fir, spruce and beech humus were characterized by similar Se levels  
219 ( $682 \pm 137 \mu\text{g kg}^{-1}$ ,  $684 \pm 134$ ,  $742 \pm 174 \mu\text{g kg}^{-1}$ ) (Figure 2.3, Kruskal-Wallis, p-value <  
220 0.001). Oak bulk humus had the lowest I concentration varying from 2.2 to 3.9 mg kg<sup>-1</sup>  
221 (Figure 2.4, ANOVA, p-value < 0.001).

### 222 3.3 Soil

223 Selenium and iodine soil profiles under the five tree stands are presented in Figures 3.1-3.2.  
224 Selenium concentrations in all profiles ranged from 331 to 690  $\mu\text{g kg}^{-1}$  (Table 2) with mean  
225 of weighted concentrations for the entire profile of 461  $\mu\text{g kg}^{-1}$ , which is similar to the global  
226 average of 400  $\mu\text{g kg}^{-1}$  (Shahid et al., 2018) and in agreement with the range of  
227 concentrations (25 - 1222  $\mu\text{g kg}^{-1}$ ) for French forest soils (Tolu et al., 2014). Typical low Se  
228 concentration of  $46 \pm 5 \mu\text{g}\cdot\text{kg}^{-1}$  was found in 2 - 5 mm soil fraction (depth 60 – 75 cm)  
229 derived from granite bedrock material that corresponds well to average levels of 25-50  $\mu\text{g}$   
230 (Se)  $\text{kg}^{-1}$  reported for granites (Shahid et al., 2018; Sharma et al., 2015).

231 Iodine concentration in soils varied with depth from 4.5 to 14.5 mg kg<sup>-1</sup>. The average iodine  
232 concentration was 9.5 mg kg<sup>-1</sup> that is slightly higher than the European mean of 5.6 mg kg<sup>-1</sup>  
233 (Fuge and Johnson, 2015). These results were within the same range as iodine levels of 2-10  
234 mg kg<sup>-1</sup> reported in Germany (Epp et al., 2020), 0.39 - 35.65 mg kg<sup>-1</sup> in French forests  
235 (Roulier et al., 2019) and 2.89 - 32.0 mg kg<sup>-1</sup> in Ireland (Bowley et al., 2019). Iodine  
236 concentrations showed maximum values at a depth of 15 to 30 cm, and then decreased to 70  
237 to 75 cm depth to values similar to those measured at the soil surface (Figure 3.2). Low  
238 iodine concentration of  $0.30 \pm 0.03 \text{ mg kg}^{-1}$  found in soil fraction > 2 mm confirmed that the  
239 granite material is generally poor in iodine (mean of 0.25 mg kg<sup>-1</sup>, Fuge and Johnson, 2015).

240 Selenium concentrations showed strong positive correlation (Pearson test,  $r_p > 0.6$ ) with  $C_{org}$   
241 and exchangeable Al and Fe (Table SI-4). Moreover, moderate positive correlations (Pearson  
242 test,  $r_p > 0.4$ ) were observed with oxalate extractable Fe and the cationic exchange capacity  
243 (CEC). Iodine concentrations were positively correlated with oxalate extractable Al and  
244 negatively correlated with  $C_{org}$ , CEC, and exchangeable Fe (Pearson test,  $|r_p| > 0.4$ ).  
245 Significant statistical differences between tree species were visible for Se and I  
246 concentrations only in topsoil layers (Table 2; t-test,  $p < 0.05$ ; 0 to 7.5 cm: Se: [Douglas fir,  
247 pine] < [spruce]; I: [pine, oak] < [spruce, Douglas fir]; 7.5 to 15 cm: Se: [Douglas fir, pine] <  
248 [oak]; I: [oak] < [spruce]). For the whole soil column (0 to 70 cm), differences between mean  
249 concentrations were observed for Se: [Douglas fir, pine] < [spruce, beech, oak], and for I:  
250 [pine] < [beech]. Differences with oak stand should be viewed with caution as soil profile  
251 was obtained from single sampling point.

### 252 **3.4 Stocks in forest ecosystem**

253 Selenium and iodine stocks are presented in Table 3. The average total pools of elements in  
254 monospecific stands were  $3.2 \pm 0.5$  kg Se  $ha^{-1}$  and  $67 \pm 11$  kg I  $ha^{-1}$ , lowest values being  
255 observed in the pine forest. The average total element partitioning between the forest  
256 compartments decreased as follow: soil ( $\geq 98.8\%$ ), humus ( $\leq 1.2\%$ ), tree compartments  
257 ( $\leq 0.09\%$ ). The higher stocks of elements for conifers compared to deciduous trees (Table 3)  
258 can be explained by the higher biomass of conifers (Table SI-2). Selenium stock in bole-  
259 wood represented generally the largest reservoir (34 – 60%) among the aboveground tree  
260 compartments. The very low iodine concentrations (< LoD) was compensated by a large  
261 biomass ( $\approx 70\%$  of the tree biomass), leading to iodine stocks in bole-wood of 8 – 17%.  
262 Except beech, bole-bark and foliage compartments stored less Se than branches. Iodine stocks  
263 were homogeneously distributed between bole-bark, branches and foliage in oak and Douglas  
264 fir, while main reservoirs were branches for spruce and pine, and bole-bark for beech. For

265 both elements, their total stock in aboveground tree parts increased in the order: oak < beech,  
266 pine  $\leq$  spruce, Douglas fir (t-test,  $p < 0.05$ ). This is in agreement with the increase in biomass  
267 pools from oak to Douglas fir stands, while beech, pine and spruce have similar intermediary  
268 biomass pools.

## 269 **4. Discussion**

### 270 **4.1 Biological distribution and cycling in tree compartments**

271 Calculated requirement fluxes of Se and I were significantly lower for pine (Table 4,  $p <$   
272  $0.05$ ), as expected due to its lower aboveground biomass productivity compared to the other  
273 stands. For both elements, foliage compartment accounted for the pool for deciduous stands  
274 (57-85%) and a minor part for conifer plots (8-26%), in agreement with the low foliage  
275 productivity of conifers (Table SI-3). Mean annual uptake fluxes (Se:  $0.29 - 0.58 \text{ g ha}^{-1} \text{ y}^{-1}$ ;  
276 I:  $1.52 - 3.52 \text{ g ha}^{-1} \text{ y}^{-1}$ ) increased as follows: beech  $\leq$  oak, spruce, pine < Douglas fir. The  
277 proportion of element uptake annually restituted through litterfall was lower for Se (65-86%)  
278 compared to I (84-92%). Immobilization in ligneous organs was thus a minor part of the  
279 uptake, accounting for about 25% for Se and 14% for I. Uptake exceeded requirement flux,  
280 indicating that the amount of absorbed elements surpassed that involved in new biomass  
281 production. These gaps were higher for conifers, especially pine, compared to deciduous  
282 stands (uptake/requirement ratios; Table 4). A large fraction of the absorbed elements was  
283 accumulated in the senescing foliage, as shown by negative annual fluxes of internal  
284 translocation for all tree species. Translocated fractions were similar for both elements with  
285 the highest values for pine (63% of uptake fluxes) compared to other trees (20-39%). The  
286 returns by litterfall contributed  $0.19-0.38 \text{ g ha}^{-1} \text{ yr}^{-1}$  of Se and  $1.40-2.94 \text{ g ha}^{-1} \text{ yr}^{-1}$  of I to the  
287 forest floor. For comparison, average atmospheric inputs by wet deposition in continental  
288 climate were reported to be  $0.35 \text{ g ha}^{-1} \text{ yr}^{-1}$  of Se and  $8.8 \text{ g ha}^{-1} \text{ yr}^{-1}$  of I (Roulier et al.,  
289 2020a).

290 Elements occurring in tree compartments can originate from both the soil solution and the  
291 atmosphere. The very low quantities of Se and I in bole-wood (Table 2) in addition to  
292 relatively low apparent root uptake is indicative of a low biological recycling of these  
293 elements from the soil. Elements originating from atmospheric deposition can accumulate on  
294 tree surface or/and be absorbed by the leaves (Roulier et al., 2020b). Selenium and iodine  
295 have been shown to be taken up by aerial parts of plants (Cakmak et al., 2017; Hurtevent et  
296 al., 2013; Kikkert and Berkelaar, 2013). Only 13% of total I content was washable from  
297 beech leaf surface, which would indicate that most of iodine was absorbed into leaf (Roulier  
298 et al., 2018). Deciduous trees showed higher concentrations of elements in leaves compared  
299 to conifers, which, combined with higher annual foliage production (Table SI-3), resulted in  
300 an apparent higher requirement flux for foliage production (Table 4). Leaves and needles in  
301 litterfall showed an average increase in Se and I concentrations by a factor of 2.5 to 4  
302 compared to living foliage, which can be explained by the translocation of elements to  
303 senescing leaves, longer exposure to atmospheric deposition, and loss of litterfall mass. The  
304 former process is a detoxification mechanism, in which excessive or redundant elements (e.g.  
305 Se, I, Ca, Mn, Zn, Fe, U) are translocated to the older organs to be eliminated by defoliation  
306 (Di Tullo, 2015; Roulier et al., 2018; Thiry et al., 2005; Ukonmaanaho et al., 2008), but its  
307 contribution to the net litterfall enrichment remains uncertain. Moreover, much higher  
308 concentrations of Se and I were measured in litterfall-branches compared to branches,  
309 especially for conifers (Figure 1). The increase was even higher than previously discussed for  
310 leaves and, almost twice for iodine compared to selenium for all tree species. The observed  
311 differences between tree species may be caused by longer living period of conifer branches  
312 compared to deciduous trees (as reflected in higher ratios of biomass branches productivity to  
313 litterfall-branches for conifers), and thus longer exposure to atmospheric deposits. These

314 results indicated that sorption of elements from atmospheric deposits might lead to  
315 overestimation of the apparent uptake fluxes and biological recycling capacities of forest.

#### 316 **4.2 The influence of tree species on Se and I contents in humus and soil**

317 The gradual enrichment in Se and I of humus horizons was observed for all tree species  
318 (Figures 2.1-2.2). A part of element enrichment in humus layers is merely associated to the  
319 loss of biomass during the ageing of organic matter and the mineralization/stabilization  
320 processes (Laganière et al., 2010; Prescott et al., 2000). Another part may originate from  
321 atmospheric deposition via throughfall, and further sorptive retention by decaying organic  
322 matter as demonstrated for other elements (van der Heijden et al., 2013). Faster  
323 decomposition under deciduous trees was reflected by lower calculated average residence  
324 times for humus from oak and beech (10 and 19 years) than for Douglas fir, spruce and pine  
325 (48, 34 and 71 years, Table 5). Even so, the net accumulation rates for all stands, which  
326 varied between 0.2 and 1.2 g ha<sup>-1</sup> yr<sup>-1</sup> for Se, and between 2.4 and 7.2 g ha<sup>-1</sup> yr<sup>-1</sup> for I (Table  
327 5). In general, annual accumulation rates were higher than litterfall fluxes, corroborating the  
328 atmospheric origin of a significant fraction of both elements retained in humus. Element  
329 accumulation rates were within the ranges reported for French forests (Se: -0.02-1.6 g ha<sup>-1</sup>  
330 yr<sup>-1</sup>; I: 0.02-15.8 g ha<sup>-1</sup> yr<sup>-1</sup>; Pisarek et al., 2021; Roulier et al., 2019). Despite similar  
331 element restitution to the forest soil (Table 4), rapid organic matter degradation in humus of  
332 oak and beech plots led to low humus biomass pools and the lowest storage of Se (next to  
333 pine) and I. Additionally, oak had the lowest concentration of Se (next to pine) and I in  
334 humus bulk. The homogenous element concentrations observed among other tree species  
335 might be due to the relatively long residence times of humus compared to the age of the  
336 stands (29 years at the time of sampling). Considering the mean concentration in soil column  
337 (0 to 70 cm), pine and Douglas fir had generally lower concentration of Se and I, probably  
338 due to lower input from humus caused by its long residence time (71 and 48 years; Table 5).

339 Although the effect of tree species on composition of the soil solution (e.g. anions content)  
340 was noticeable (Legout et al., 2016), Se and I contents in soil layers were statistically different  
341 only in topsoil, and these differences disappeared below 15 cm depth. It is supposed, that  
342 recently introduced tree species did not yet affect the content and distribution of Se and I in  
343 deeper soil layers and that their storage depends rather on longer term influence of  
344 atmospheric deposition and soil functional parameters.

### 345 **4.3 Selenium and iodine distributions in soil profiles**

346 The Se- and I-poor status of bedrock material ( $46 \mu\text{g Se kg}^{-1}$  and  $0.30 \text{ mg I kg}^{-1}$ ; Table 2)  
347 combined with the observed enrichment of both elements in bulk humus indicated that rock  
348 weathering was a minor source of Se and I in soil reserves compared to atmospheric  
349 deposition. Contrasting vertical profiles of concentrations suggest that external inputs of each  
350 element at the surface were redistributed by different processes during downward transports.

351 In general, Se concentrations slightly decreased with soil depth, by 30% on average from  
352 topsoil layer to the deepest mineral layer. Reimann et al. (2015) similarly observed a decrease  
353 from  $400\text{-}6300 \mu\text{g Se kg}^{-1}$  in O-horizon to  $100 - 2700 \mu\text{g Se kg}^{-1}$  in the deeper mineral C-  
354 horizon in Norwegian forest soils. High correlation between Se and C concentrations ( $r_p =$   
355  $0.64$ ,  $p < 0.01$ , Table SI-4) underlined the pivotal role of OM in Se redistribution and possible  
356 retention in soil (Sharma et al., 2015). Selenium concentrations were positively correlated  
357 with the mineral soil constituents: Al exchangeable, Fe exchangeable, and Fe oxalate.  
358 However, co-correlations with  $C_{\text{tot}}$  and Fe concentrations prevented the discrimination of a  
359 dominant carrier phase for Se.

360 In contrast to selenium, iodine concentration increased from the forest floor to a maximum at  
361 a soil depth of 15 to 35 cm, and then decreased until 70 to 75 cm to values similar to those  
362 measured at the soil surface (Figure 3.1). Different soil iodine profiles have been reported in

363 the literature. Bostock (2004) and Shetaya et al. (2012) reported a gradual decrease of I  
364 content with soil depth (until 110 cm) and decreasing OM content. Like in our study, increase  
365 of I with soil depth and anti-correlation with OM content were observed in some studies (Epp  
366 et al., 2020; Roulier et al., 2018; Xu et al., 2016). Roulier et al. (2018) measured a modest  
367 increase in I concentration at 15 to 45 cm depth in beech forest. Xu et al. (2016) reported also  
368 an increase of I concentration until a 15 cm depth in a coniferous forest but no depth-related  
369 variation in a deciduous forest.

370 In surface forest soil, iodine is rapidly transformed into organic forms (Takeda et al., 2015),  
371 that determines its mobility in the soil profile. The behavior of dissolved organo-iodine (DOI)  
372 in forest soil is strongly affected by dynamic of dissolved organic carbon DOC (Takeda et al.,  
373 2019). At our forest site, it is postulated that iodine that is derived from the surface horizon as  
374 DOI may accumulate on Al oxides (Takeda et al., 2018; Cortizas et al. 2016; Whitehead,  
375 1978). That hypothesis is supported by the co-existence of a maximum of oxalate extracted  
376 Al and total I at the depth of 15 to 35 cm (Figure SI-2,  $r^2=0.63$ ). An enrichment-depletion  
377 profile is common for Al in acid soils, where conditions in the topsoil promote the dissolution  
378 of Al and the formation of secondary Al precipitates in deeper profile (Brantley et al., 2007).  
379 The sorption capacity of Al oxides for DOC (Kalbitz et al., 2000), and thus including DOI, is  
380 well documented (Söderlund et al., 2017). Similarly, an iodine retention front was noticed by  
381 Unno et al. (2017) at 20 to 30 cm in a forest soil, in close agreement with the partitioning of  
382 OC between solid and liquid soil phases. That means that leaching and further immobilization  
383 of both Al and OC may have a strong influence on iodine migration in acid forest soils.

384 The possible influence of volatilization and root uptake on iodine depletion in surface horizon  
385 is less clear. Our estimation of iodine absorption by roots, based on calculated uptake flux,  
386 would represent no more than 0.02% of iodine stored in the 0 to 15 cm soil. Given this  
387 meager annual output, root uptake would not radically influence the distribution of I in soil.

388 Even though volatilization from terrestrial environment contributes to the global atmospheric  
389 budget (Feinberg A. et al., 2020; Kadowaki et al., 2020), iodine loss by volatilization is  
390 probably a minor process to explain iodine depletion in the upper layer of these soil profiles.  
391 In experiments with spiked forest soils, the loss of  $^{125}\text{I}$  through volatilization accounted for  
392 0.011-0.07% of total iodine for reaction times  $\leq 66$  days (Bostock et al., 2003; Sheppard, M.  
393 et al., 1994). According to the poor knowledge on the magnitude of I volatilization and its  
394 environmental controls in different site conditions, further studies are needed to investigate  
395 the balance between retention and volatilization depending on the physicochemical properties  
396 of the soil.

## 397 **5. Conclusions**

398 Monitoring of monospecific stands located under identical climatic and geogenic conditions  
399 enabled specifying the impact of tree species on inventories of Se and I. For both elements,  
400 stocks in aboveground tree parts were marginal compared to forest soil ( $\leq 0.09\%$  for Se,  $\leq$   
401  $0.013\%$  for I). Tree species affected element stocks in aboveground tree parts (oak < beech,  
402 pine  $\leq$  spruce, Douglas fir) and uptake fluxes (beech  $\leq$  oak, pine, spruce < Douglas fir).  
403 Returns to forest floor through litterfall were lower at beech stand for Se and higher at  
404 Douglas fir stand for I. Vegetation type influenced the organic material and element turnover  
405 in humus. The stands with high humus biomass (i.e Douglas fir and spruce) stored more Se  
406 and I, while low biomass and short residence time of OM resulted in reduced pools of  
407 elements in deciduous humus. Under our edaphic conditions and stand ages, tree species did  
408 not influence soil Se and I reserves. In the soil profile, selenium concentrations slightly  
409 decreased with soil depth while iodine showed an “enrichment-depletion profile” that could  
410 be explained by downward leaching of DOI and enrichment with depth by sorption onto Al  
411 oxides.

412 *Supplementary information*

413 Additional information as noted in the text.

414 **Declaration of interests**

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

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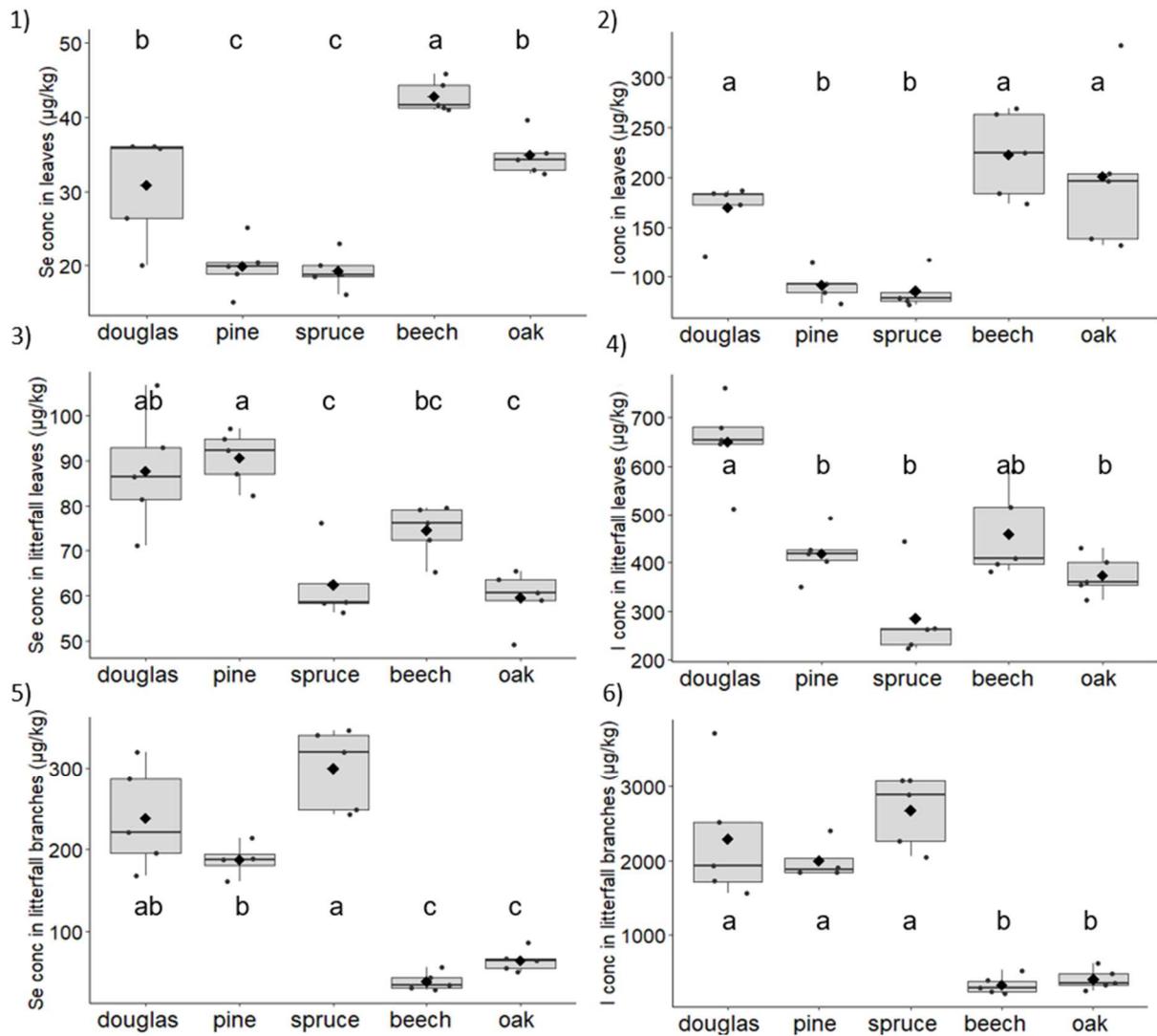


Figure 1 Selenium and iodine concentrations (dots) in (1), (2) leaves; (3), (4) litterfall-leaves and (5), (6) litterfall-branches in forest stands (n=5/stand). Box plots show the first quartile (Q<sub>1</sub>), third quartile (Q<sub>3</sub>) and median of the data. Black diamond symbols indicate average element concentrations in groups. Upper and lower whiskers extend from the hinges by  $Q_1 - 1.5 \times IQR$  ( $IQR$  - interquartile range) and  $Q_3 + 1.5 \times IQR$ , respectively. Letters above the boxes correspond to significant differences between the groups according to Wilcoxon or Tukey tests.

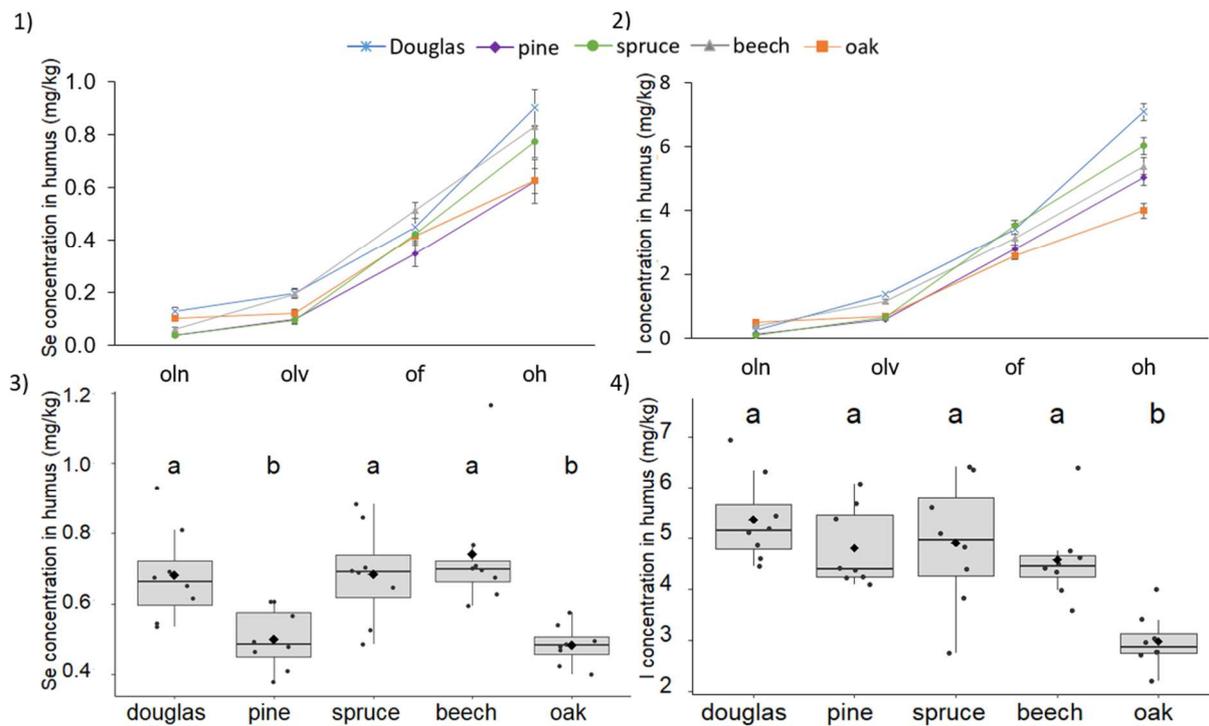


Figure 2 Selenium and iodine concentrations in (1), (2) humus horizons: oln (organic, litter, new), olv (organic, litter, old), of (organic, fragmented), oh (organic, humified) and (3), (4) bulk humus (n=8/stand) of Douglas, pine, spruce, beech and oak monospecific stands. Box plots show the first quartile (Q<sub>1</sub>), third quartile (Q<sub>3</sub>) and median of the data. Black diamond symbols indicate average element concentrations in groups. Upper and lower whiskers extend from the hinges by Q<sub>1</sub> - 1.5 × IQR (IQR - interquartile range) and Q<sub>3</sub> + 1.5 × IQR, respectively. Letters above the boxes correspond to significant differences between the groups according to Wilcoxon or Tukey tests.

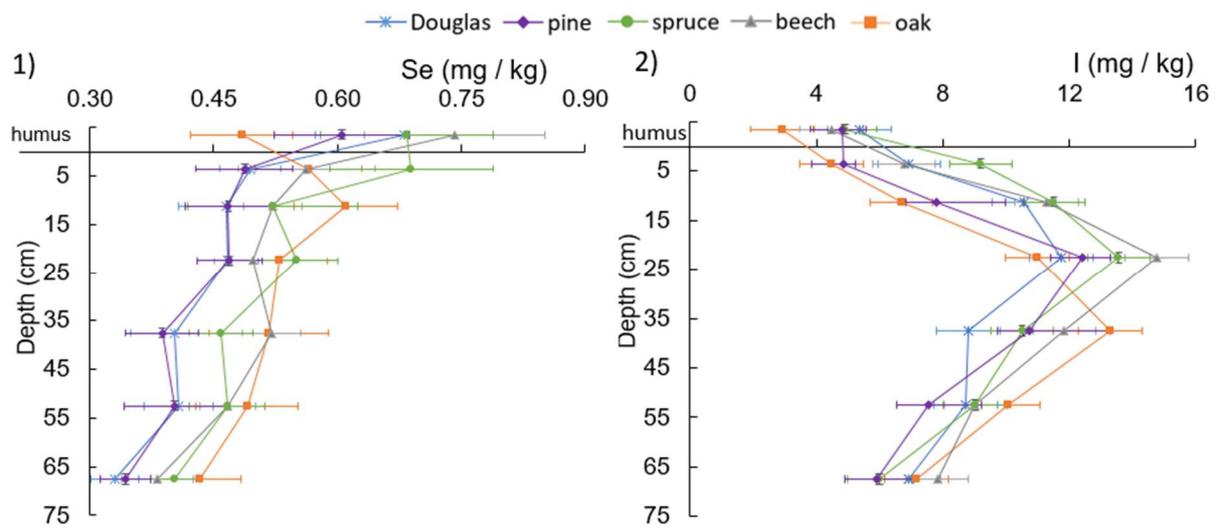


Figure 3. Soil profiles of selenium (1) and iodine (2) in forest stands (mean value  $\pm$  standard deviation).

Table 1 Equations to calculate main annual fluxes of Se and I in forest ecosystem.

<b>Annual flux</b>	<b>Equation</b>	<b>Variables</b>
<b>Requirement (R)</b>	$R = \sum_i (\text{annual biomass production})_i \times [X]_i$	i = leaves, branches, bark, wood
<b>Uptake (U)</b>	$U = (U_1) + (U_2)$	
(U <sub>1</sub> ) Immobilization	$U_1 = \sum_i (\text{annual biomass production})_i \times [X]_i$	i = branches, wood, bark
(U <sub>2</sub> ) Returns	$U_2 = \sum_i (\text{annual biomass production})_i \times [X]_i$	i = litterfall leaves , branches
<b>Translocations (T)</b>	$T = (\text{annual biomass production})_i \times ([X]_j - [X]_i)$	i = litterfall leaves; j = leaves

Where: [X] is Se or I concentration

Table 2 Selenium and iodine concentrations in Breuil forest stands (mean value  $\pm$  standard deviation). Concentrations with various letters are significantly different among tree species at the 95% level (2tailed t-test).

	Douglas	Pine	Spruce	Beech	Oak
<b>Selenium concentrations</b>					
<b>Tree parts (<math>\mu\text{g kg}^{-1}</math>)</b>					
Bole-wood	9 $\pm$ 3	13 $\pm$ 4	11 $\pm$ 3	11 $\pm$ 4	8 $\pm$ 2
Branches	32 $\pm$ 4 <sup>a</sup>	23 $\pm$ 4 <sup>b</sup>	36 $\pm$ 5 <sup>a</sup>	18 $\pm$ 3 <sup>b</sup>	19 $\pm$ 3 <sup>b</sup>
Bole-bark	33 $\pm$ 5 <sup>b</sup>	20 $\pm$ 4 <sup>d</sup>	24 $\pm$ 3 <sup>c</sup>	47 $\pm$ 5 <sup>a</sup>	16 $\pm$ 2 <sup>d</sup>
Foliage	31 $\pm$ 4 <sup>b</sup>	20 $\pm$ 3 <sup>c</sup>	19 $\pm$ 3 <sup>c</sup>	43 $\pm$ 4 <sup>a</sup>	35 $\pm$ 4 <sup>b</sup>
<b>Forest floor (<math>\mu\text{g kg}^{-1}</math>)</b>					
LF leaves	89 $\pm$ 8 <sup>ab</sup>	90 $\pm$ 7 <sup>a</sup>	62 $\pm$ 6 <sup>c</sup>	74 $\pm$ 6 <sup>bc</sup>	60 $\pm$ 6 <sup>c</sup>
LF branches	222 $\pm$ 26 <sup>ab</sup>	187 $\pm$ 14 <sup>b</sup>	291 $\pm$ 24 <sup>a</sup>	40 $\pm$ 11 <sup>c</sup>	65 $\pm$ 8 <sup>c</sup>
LF total	108 $\pm$ 26	92 $\pm$ 53	85 $\pm$ 33	66 $\pm$ 27	62 $\pm$ 18
Humus bulk	682 $\pm$ 137 <sup>a</sup>	500 $\pm$ 98 <sup>b</sup>	684 $\pm$ 134 <sup>a</sup>	742 $\pm$ 174 <sup>a</sup>	484 $\pm$ 61 <sup>b</sup>
oln	129 $\pm$ 14	37 $\pm$ 6	40 $\pm$ 7	60 $\pm$ 10	103 $\pm$ 12
olv	197 $\pm$ 18	98 $\pm$ 14	94 $\pm$ 14	194 $\pm$ 17	122 $\pm$ 13
of	449 $\pm$ 35	347 $\pm$ 48	425 $\pm$ 34	516 $\pm$ 30	414 $\pm$ 33
oh	902 $\pm$ 70	624 $\pm$ 83	774 $\pm$ 59	830 $\pm$ 61	626 $\pm$ 47
<b>Soil (<math>\mu\text{g kg}^{-1}</math>)</b>					
0-7.5 cm	494 $\pm$ 37 <sup>b</sup>	488 $\pm$ 59 <sup>b</sup>	690 $\pm$ 99 <sup>a</sup>	564 $\pm$ 82 <sup>ab</sup>	566 $\pm$ 64 <sup>ab</sup>
7.5-15 cm	466 $\pm$ 59 <sup>b</sup>	468 $\pm$ 52 <sup>b</sup>	522 $\pm$ 103 <sup>ab</sup>	522 $\pm$ 36 <sup>ab</sup>	610 $\pm$ 63 <sup>a</sup>
15-30 cm	467 $\pm$ 58	469 $\pm$ 39	550 $\pm$ 50	499 $\pm$ 47	530 $\pm$ 58
30-45 cm	408 $\pm$ 54	388 $\pm$ 74	459 $\pm$ 38	520 $\pm$ 36	517 $\pm$ 72
45-60 cm	404 $\pm$ 42	403 $\pm$ 62	467 $\pm$ 45	468 $\pm$ 34	491 $\pm$ 62
60-75(70) cm	331 $\pm$ 59	343 $\pm$ 38	405 $\pm$ 54	382 $\pm$ 43	433 $\pm$ 51
0-70 cm	417 $\pm$ 47	415 $\pm$ 55	493 $\pm$ 55	482 $\pm$ 42	510 $\pm$ 64
<b>Bedrock material</b>	46 $\pm$ 5				
<b>Iodine concentrations</b>					
<b>Tree parts (<math>\mu\text{g kg}^{-1}</math>)</b>					
Bole-wood	<16.5	<16.5	<16.5	<16.5	<16.5
Branches	118 $\pm$ 5 <sup>b</sup>	104 $\pm$ 5 <sup>c</sup>	158 $\pm$ 5 <sup>a</sup>	39 $\pm$ 2 <sup>c</sup>	57 $\pm$ 2 <sup>d</sup>
Bole-Bark	144 $\pm$ 6 <sup>b</sup>	89 $\pm$ 6 <sup>c</sup>	100 $\pm$ 5 <sup>c</sup>	226 $\pm$ 20 <sup>a</sup>	49 $\pm$ 5 <sup>d</sup>
Foliage	163 $\pm$ 14 <sup>a</sup>	89 $\pm$ 8 <sup>b</sup>	82 $\pm$ 9 <sup>b</sup>	208 $\pm$ 21 <sup>a</sup>	182 $\pm$ 32 <sup>a</sup>
<b>Forest floor (mg kg<sup>-1</sup>)</b>					
LF leaves	0.63 $\pm$ 0.04 <sup>a</sup>	0.40 $\pm$ 0.02 <sup>b</sup>	0.28 $\pm$ 0.04 <sup>b</sup>	0.44 $\pm$ 0.04 <sup>ab</sup>	0.36 $\pm$ 0.02 <sup>b</sup>
LF branches	2.1 $\pm$ 0.3 <sup>a</sup>	1.9 $\pm$ 0.1 <sup>a</sup>	2.5 $\pm$ 0.2 <sup>a</sup>	0.34 $\pm$ 0.05 <sup>b</sup>	0.40 $\pm$ 0.06 <sup>b</sup>
LF total	0.83 $\pm$ 0.20	0.40 $\pm$ 0.20	0.51 $\pm$ 0.20	0.41 $\pm$ 0.16	0.37 $\pm$ 0.11
Humus bulk	5.4 $\pm$ 0.8 <sup>b</sup>	4.8 $\pm$ 0.7 <sup>b</sup>	4.9 $\pm$ 1.2 <sup>b</sup>	4.5 $\pm$ 0.8 <sup>b</sup>	2.9 $\pm$ 0.5 <sup>a</sup>
oln	0.24 $\pm$ 0.02	0.12 $\pm$ 0.01	0.09 $\pm$ 0.01	0.39 $\pm$ 0.03	0.51 $\pm$ 0.03
olv	1.4 $\pm$ 0.1	0.58 $\pm$ 0.03	0.64 $\pm$ 0.05	1.1 $\pm$ 0.1	0.68 $\pm$ 0.05
of	3.4 $\pm$ 0.2	2.8 $\pm$ 0.1	3.5 $\pm$ 0.2	3.1 $\pm$ 0.2	2.6 $\pm$ 0.1
oh	7.1 $\pm$ 0.3	5.0 $\pm$ 0.2	6.0 $\pm$ 0.3	5.4 $\pm$ 0.3	4.0 $\pm$ 0.2
<b>Soil (mg kg<sup>-1</sup>)</b>					
0-7.5 cm	6.7 $\pm$ 0.5 <sup>a</sup>	4.8 $\pm$ 0.4 <sup>b</sup>	9.0 $\pm$ 1.7 <sup>a</sup>	6.6 $\pm$ 1.9 <sup>ab</sup>	4.5 $\pm$ 0.3 <sup>b</sup>
7.5-15 cm	10.3 $\pm$ 1.9 <sup>ab</sup>	7.7 $\pm$ 2.1 <sup>ab</sup>	11.3 $\pm$ 2.5 <sup>a</sup>	11.1 $\pm$ 2.8 <sup>ab</sup>	6.7 $\pm$ 0.2 <sup>b</sup>
15-30 cm	11.4 $\pm$ 4.3	12.2 $\pm$ 0.8	13.3 $\pm$ 1.4	14.5 $\pm$ 1.4	11.0 $\pm$ 0.4
30-45 cm	8.6 $\pm$ 1.3	10.5 $\pm$ 2.5	10.3 $\pm$ 1.6	11.6 $\pm$ 1.2	13.3 $\pm$ 0.5
45-60 cm	8.5 $\pm$ 0.7	7.4 $\pm$ 1.6	8.8 $\pm$ 1.2	8.9 $\pm$ 0.6	10.1 $\pm$ 0.4
60-75(70) cm	6.8 $\pm$ 2.4	5.8 $\pm$ 1.2	5.9 $\pm$ 1.1	7.7 $\pm$ 1.3	7.2 $\pm$ 0.2
0-70 cm	8.8 $\pm$ 1.9	8.7 $\pm$ 1.5	9.8 $\pm$ 1.5	10.5 $\pm$ 1.3	9.9 $\pm$ 0.4
<b>Bedrock material</b>	0.30 $\pm$ 0.03				

Table 3 Selenium and iodine stocks in monospecific forest stands (mean value  $\pm$  standard deviation). Tree and humus stocks with various letters are significantly different among tree species at the 95% level (t-test).

	<b>Douglas</b>	<b>Pine</b>	<b>Spruce</b>	<b>Beech</b>	<b>Oak</b>
<b>Selenium stocks (g ha<sup>-1</sup>)</b>					
<b>Tree</b>	<b>2.60 <math>\pm</math> 0.34<sup>a</sup></b> (0.09%)	<b>2.00 <math>\pm</math> 0.38<sup>a,b</sup></b> (0.07%)	<b>2.57 <math>\pm</math> 0.30<sup>a</sup></b> (0.07%)	<b>1.82 <math>\pm</math> 0.33<sup>b</sup></b> (0.06%)	<b>0.91 <math>\pm</math> 0.12<sup>c</sup></b> (0.03%)
Bole-wood	0.96 $\pm$ 0.29	1.21 $\pm$ 0.37	0.86 $\pm$ 0.25	0.92 $\pm$ 0.32	0.38 $\pm$ 0.10
Branches	0.65 $\pm$ 0.08	0.36 $\pm$ 0.07	0.93 $\pm$ 0.13	0.38 $\pm$ 0.07	0.25 $\pm$ 0.04
Bole-bark	0.53 $\pm$ 0.08	0.28 $\pm$ 0.05	0.22 $\pm$ 0.03	0.38 $\pm$ 0.04	0.14 $\pm$ 0.02
Foliage	0.45 $\pm$ 0.13	0.15 $\pm$ 0.04	0.56 $\pm$ 0.10	0.14 $\pm$ 0.05	0.14 $\pm$ 0.04
<b>Forest soil</b>	<b>2988 <math>\pm</math> 335</b>	<b>2824 <math>\pm</math> 370</b>	<b>3535 <math>\pm</math> 389</b>	<b>3071 <math>\pm</math> 264</b>	<b>3553 <math>\pm</math> 441</b>
Humus	34.5 $\pm$ 7.3 <sup>a</sup> (1.2%)	19.3 $\pm$ 4.6 <sup>b</sup> (0.7%)	35.6 $\pm$ 8.0 <sup>a</sup> (1%)	16.4 $\pm$ 6.9 <sup>b,c</sup> (0.5%)	13.3 $\pm$ 1.7 <sup>c</sup> (0.4%)
Soil (0-70 cm)	2954 $\pm$ 335 (98.8%)	2805 $\pm$ 370 (99.2%)	3499 $\pm$ 389 (98.9%)	3054 $\pm$ 264 (99.4%)	3540 $\pm$ 441 (99.6%)
<b>Total stock</b>	<b>2991 <math>\pm</math> 335</b>	<b>2826 <math>\pm</math> 370</b>	<b>3538 <math>\pm</math> 389</b>	<b>3073 <math>\pm</math> 264</b>	<b>3554 <math>\pm</math> 441</b>
<b>Iodine stocks (g ha<sup>-1</sup>)</b>					
<b>Tree</b>	<b>8.09 <math>\pm</math> 0.99<sup>a</sup></b> (0.013%)	<b>4.49 <math>\pm</math> 0.81<sup>b</sup></b> (0.008%)	<b>7.99 <math>\pm</math> 0.77<sup>a</sup></b> (0.011%)	<b>4.07 <math>\pm</math> 0.74<sup>b</sup></b> (0.006%)	<b>2.40 <math>\pm</math> 0.45<sup>c</sup></b> (0.003%)
Bole-wood	0.86 $\pm$ 0.86	0.78 $\pm$ 0.78	0.66 $\pm$ 0.66	0.70 $\pm$ 0.70	0.39 $\pm$ 0.39
Branches	2.47 $\pm$ 0.09	1.76 $\pm$ 0.08	3.99 $\pm$ 0.14	0.84 $\pm$ 0.03	0.76 $\pm$ 0.03
Bole-bark	2.39 $\pm$ 0.17	1.28 $\pm$ 0.13	0.97 $\pm$ 0.12	1.85 $\pm$ 0.16	0.50 $\pm$ 0.12
Foliage	2.37 $\pm$ 0.44	0.67 $\pm$ 0.13	2.37 $\pm$ 0.34	0.68 $\pm$ 0.18	0.75 $\pm$ 0.18
<b>Iodine stocks (kg ha<sup>-1</sup>)</b>					
<b>Forest soil</b>	<b>63 <math>\pm</math> 14</b>	<b>59 <math>\pm</math> 10</b>	<b>70 <math>\pm</math> 10</b>	<b>67 <math>\pm</math> 8</b>	<b>68 <math>\pm</math> 3</b>
Humus	0.27 $\pm$ 0.05 <sup>a</sup> (0.4%)	0.19 $\pm$ 0.04 <sup>b</sup> (0.3%)	0.26 $\pm$ 0.07 <sup>a</sup> (0.4%)	0.10 $\pm$ 0.04 <sup>c</sup> (0.15%)	0.08 $\pm$ 0.01 <sup>c</sup> (0.12%)
Soil (0-70 cm)	63 $\pm$ 14 (99.6%)	59 $\pm$ 10 (99.7%)	70 $\pm$ 10 (99.6%)	66 $\pm$ 8 (99.8%)	68 $\pm$ 3 (99.9%)
<b>Total stock</b>	<b>63 <math>\pm</math> 14</b>	<b>59 <math>\pm</math> 10</b>	<b>70 <math>\pm</math> 10</b>	<b>67 <math>\pm</math> 8</b>	<b>68 <math>\pm</math> 3</b>

Table 4 Selenium and iodine annual fluxes in monospecific forest stands (mean value  $\pm$  standard deviation). Fluxes with various letters are significantly different among tree species at the 95% level (t-test).

Se annual flux (g ha <sup>-1</sup> y <sup>-1</sup> )					
Annual flux	Douglas	Pine	Spruce	Beech	Oak
<b>Requirement (R)</b>	<b>0.24 <math>\pm</math> 0.04<sup>a</sup></b>	<b>0.06 <math>\pm</math> 0.02<sup>c</sup></b>	<b>0.17 <math>\pm</math> 0.05<sup>a</sup></b>	<b>0.21 <math>\pm</math> 0.02<sup>a</sup></b>	<b>0.25 <math>\pm</math> 0.07<sup>a</sup></b>
Wood	0.079 $\pm$ 0.032	0.032 $\pm$ 0.014	0.055 $\pm$ 0.034	0.033 $\pm$ 0.012	0.044 $\pm$ 0.036
Branches	0.078 $\pm$ 0.024	0.017 $\pm$ 0.006	0.069 $\pm$ 0.038	0.025 $\pm$ 0.005	0.050 $\pm$ 0.041
Bark	0.046 $\pm$ 0.014	0.007 $\pm$ 0.002	0.012 $\pm$ 0.007	0.009 $\pm$ 0.003	0.014 $\pm$ 0.011
Leaves	0.04 $\pm$ 0.01	0.005 $\pm$ 0.002	0.03 $\pm$ 0.02	0.14 $\pm$ 0.01	0.14 $\pm$ 0.05
<b>Uptake (U)</b>	<b>0.58 <math>\pm</math> 0.08<sup>a</sup></b>	<b>0.39 <math>\pm</math> 0.13<sup>b</sup></b>	<b>0.47 <math>\pm</math> 0.10<sup>b</sup></b>	<b>0.29 <math>\pm</math> 0.07<sup>c</sup></b>	<b>0.41 <math>\pm</math> 0.09<sup>b</sup></b>
<b>(U<sub>1</sub>) Immobilization</b>	<b>0.20 <math>\pm</math> 0.04</b>	<b>0.06 <math>\pm</math> 0.02</b>	<b>0.14 <math>\pm</math> 0.05</b>	<b>0.07 <math>\pm</math> 0.01</b>	<b>0.11 <math>\pm</math> 0.06</b>
Wood	0.079 $\pm$ 0.032	0.032 $\pm$ 0.014	0.055 $\pm$ 0.034	0.033 $\pm$ 0.012	0.044 $\pm$ 0.036
Branches	0.078 $\pm$ 0.024	0.017 $\pm$ 0.006	0.069 $\pm$ 0.038	0.025 $\pm$ 0.005	0.050 $\pm$ 0.041
Bark	0.046 $\pm$ 0.014	0.007 $\pm$ 0.002	0.012 $\pm$ 0.007	0.009 $\pm$ 0.003	0.014 $\pm$ 0.011
<b>(U<sub>2</sub>) Returns</b>	<b>0.38 <math>\pm</math> 0.07</b>	<b>0.34 <math>\pm</math> 0.13</b>	<b>0.34 <math>\pm</math> 0.08</b>	<b>0.22 <math>\pm</math> 0.07</b>	<b>0.31 <math>\pm</math> 0.07</b>
Litterfall leaves	0.27 $\pm$ 0.05	0.32 $\pm$ 0.13	0.22 $\pm$ 0.08	0.19 $\pm$ 0.07	0.19 $\pm$ 0.06
Litterfall branches	0.11 $\pm$ 0.04	0.017 $\pm$ 0.004	0.12 $\pm$ 0.03	0.03 $\pm$ 0.01	0.11 $\pm$ 0.02
<b>Translocation (T)</b>	<b>-0.17 <math>\pm</math> 0.04</b>	<b>-0.25 <math>\pm</math> 0.11</b>	<b>-0.15 <math>\pm</math> 0.06</b>	<b>-0.08 <math>\pm</math> 0.03</b>	<b>-0.08 <math>\pm</math> 0.03</b>
<b>Se Uptake/Se Requirement</b>					
Ratio	2.4 $\pm$ 0.6	6.4 $\pm$ 2.7	2.8 $\pm$ 1.1	1.4 $\pm$ 0.4	1.6 $\pm$ 0.6
I annual flux (g ha <sup>-1</sup> y <sup>-1</sup> )					
Annual flux	Douglas	Pine	Spruce	Beech	Oak
<b>Requirement (R)</b>	<b>0.78 <math>\pm</math> 0.14<sup>a</sup></b>	<b>0.16 <math>\pm</math> 0.03<sup>b</sup></b>	<b>0.53 <math>\pm</math> 0.19<sup>a</sup></b>	<b>0.81 <math>\pm</math> 0.07<sup>a</sup></b>	<b>1.00 <math>\pm</math> 0.30<sup>a</sup></b>
Wood	0.07 $\pm$ 0.07	0.02 $\pm$ 0.02	0.04 $\pm$ 0.05	0.03 $\pm$ 0.03	0.05 $\pm$ 0.06
Branches	0.29 $\pm$ 0.08	0.08 $\pm$ 0.02	0.30 $\pm$ 0.16	0.056 $\pm$ 0.004	0.16 $\pm$ 0.12
Bark	0.21 $\pm$ 0.06	0.03 $\pm$ 0.01	0.05 $\pm$ 0.03	0.04 $\pm$ 0.01	0.05 $\pm$ 0.04
Leaves	0.20 $\pm$ 0.06	0.02 $\pm$ 0.01	0.14 $\pm$ 0.08	0.68 $\pm$ 0.07	0.75 $\pm$ 0.26
<b>Uptake (U)</b>	<b>3.52 <math>\pm</math> 0.57<sup>a</sup></b>	<b>1.72 <math>\pm</math> 0.59<sup>b,c</sup></b>	<b>2.39 <math>\pm</math> 0.47<sup>b</sup></b>	<b>1.52 <math>\pm</math> 0.41<sup>c</sup></b>	<b>2.11 <math>\pm</math> 0.41<sup>b,c</sup></b>
<b>(U<sub>1</sub>) Immobilization</b>	<b>0.57 <math>\pm</math> 0.13</b>	<b>0.14 <math>\pm</math> 0.03</b>	<b>0.39 <math>\pm</math> 0.17</b>	<b>0.12 <math>\pm</math> 0.03</b>	<b>0.25 <math>\pm</math> 0.14</b>
Wood	0.07 $\pm$ 0.07	0.02 $\pm$ 0.02	0.04 $\pm$ 0.05	0.03 $\pm$ 0.03	0.05 $\pm$ 0.06
Branches	0.29 $\pm$ 0.08	0.08 $\pm$ 0.02	0.30 $\pm$ 0.16	0.056 $\pm$ 0.004	0.16 $\pm$ 0.12
Bark	0.21 $\pm$ 0.06	0.03 $\pm$ 0.01	0.05 $\pm$ 0.03	0.04 $\pm$ 0.01	0.05 $\pm$ 0.04
<b>(U<sub>2</sub>) Returns</b>	<b>2.94 <math>\pm</math> 0.55</b>	<b>1.58 <math>\pm</math> 0.59</b>	<b>2.00 <math>\pm</math> 0.43</b>	<b>1.40 <math>\pm</math> 0.40</b>	<b>1.86 <math>\pm</math> 0.38</b>
Litterfall leaves	1.90 $\pm$ 0.35	1.41 $\pm$ 0.59	0.98 $\pm$ 0.35	1.13 $\pm$ 0.39	1.16 $\pm$ 0.36
Litterfall branches	1.04 $\pm$ 0.43	0.17 $\pm$ 0.04	1.02 $\pm$ 0.25	0.27 $\pm$ 0.10	0.70 $\pm$ 0.15
<b>Translocation (T)</b>	<b>-1.41 <math>\pm</math> 0.27</b>	<b>-1.10 <math>\pm</math> 0.46</b>	<b>-0.69 <math>\pm</math> 0.27</b>	<b>-0.59 <math>\pm</math> 0.23</b>	<b>-0.57 <math>\pm</math> 0.21</b>
<b>I Uptake/I Requirement</b>					
Ratio	4.5 $\pm$ 1.1	10.8 $\pm$ 4.4	4.5 $\pm$ 1.8	1.9 $\pm$ 0.5	2.1 $\pm$ 0.7

Table 5 Humus residence time and Se and I average accumulation rates in humus of different tree species.

Tree	Humus $t_{resDM}$ (years)	Accumulation rate ( $g\ ha^{-1}yr^{-1}$ )	
		Selenium	Iodine
oak	$10 \pm 2$	$1.2 \pm 0.3$	$7.2 \pm 2.1$
beech	$19 \pm 8$	$0.8 \pm 0.5$	$4.8 \pm 2.8$
Douglas	$48 \pm 8$	$0.6 \pm 0.2$	$4.8 \pm 1.3$
spruce	$34 \pm 11$	$0.9 \pm 0.4$	$6.8 \pm 3.0$
pine	$71 \pm 30$	$0.2 \pm 0.1$	$2.4 \pm 1.2$

