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1 **Mg isotope composition in beech forest ecosystems and variations induced**
2 **by liming: insights from four experimental sites in Northern France**

3
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16
17 **Abstract**

18 Many forest soils are acidic and have very low plant-available pools of magnesium. Past
19 and present silvicultural, nutritional and/or climatic pressures endured by forest ecosystems
20 can result in net losses of nutrients and ecosystem function losses. Liming with a carbonate
21 product is an alternative to counteract these degradations but the effects of liming on the
22 biogeochemical cycling of nutrients over time and the dynamics of Mg released from liming
23 products are still unclear. We studied the Mg isotopes composition in four paired-treatment
24 experimental beech forest ecosystems in northern France. At the sites where dolomitic lime
25 was applied, the variation in exchangeable and foliar $\delta^{26}\text{Mg}$ demonstrated the direct
26 contribution of dolomite-derived Mg to the replenishment of topsoil exchangeable pools and
27 to tree nutrition improvement: dolomite-derived Mg was incorporated into the biological
28 cycling which allows its retention on the mid to long term in the soil-plant system. At the sites
29 limed with calcium carbonate, the changes in exchangeable and foliar Mg contents and $\delta^{26}\text{Mg}$
30 observed on the long term suggest that the applied product contained a small amount of Mg
31 and/or that Mg cycling changed after liming, to cope in particular with the low Mg
32 availability. Lastly, our results highlight the high $\delta^{26}\text{Mg}$ of the organic layer (humus):

33 fractionation processes occurring within this layer (mineralization/ageing of organic matter,
34 preferential retention of ^{26}Mg) could explain these singular signatures that could greatly
35 influence the topsoil Mg exchangeable pools.

36

37 **Keywords:** forest ecosystem, liming, biogeochemical cycling, Mg isotopes, dolomite lime,
38 calcium carbonate

39

40 **1. Introduction**

41 Magnesium is an essential and major nutrient in forest ecosystems (Marschner 1995) which
42 plays an important role in plants (Bush 1995; Epstein 1956; Kirkby & Mengel 1976;
43 Marschner 1995; Schulze et al. 2005) and soils by contributing to the pH buffer capacity, soil
44 structure and biological activity (Wolters 1991). In forest soils, plant-available pools of
45 magnesium are assumed to be stored as exchangeable cations adsorbed on the cation
46 exchange complex. This exchangeable pool may change over time in relation to ecosystem
47 inputs (atmospheric deposition and mineral weathering) and outputs (leaching and storage in
48 tree biomass), particularly under the influence of external pressure. Indeed, decreasing
49 atmospheric inputs (Likens et al. 1996) or the intensification of biomass harvesting and
50 silvicultural practices (Achat et al. 2015; Pyttel et al. 2015; Ranger et al. 2011; Thiffault et al.
51 2015) may lead to forest soil fertility degradation, most particularly in low soil fertility forest
52 ecosystems which are expected to be the most sensitive (Jonard et al. 2015; Van der Heijden
53 et al. 2011). When the pressure endured is too intense, this degradation may lead to the
54 progressive losses of nutrients and ecosystem functions, forest decline being the final stage of
55 this process (Court et al. 2018).

56 To mitigate such issues, carbonate products (typically calcium carbonate or dolostone) may be
57 applied (liming operation) in order to decrease soil acidity, maintain or restore forest soil
58 fertility and improve tree nutrition (Court et al. 2018). Many studies of forest ecosystem
59 liming operations have been carried out to assess the effects on organic matter decomposition
60 (Baath et al. 1980; Geissen & Brümmer 1999; Marschner & Waldemar Wilczynski 1991;
61 Smolander et al. 1996; Vuorenmaa et al. 2017), soil biological activity and community
62 (Balland-Bolou-Bi & Poszwa 2012; Clivot et al. 2012; Lepleux et al. 2013; Moore 2014;
63 Moore et al. 2013), topsoil fertility and tree nutrition or growth at short (Burke & Raynal
64 1998; Huettl 1989; Meiwes et al. 2002; Moore & Ouimet 2014; Wilmot et al. 1996) and long
65 term time scales (Bakker 1998; Forey et al. 2015; Long et al. 2015; Moore et al. 2012).

66 However, because fewer studies have investigated these effects at the ecosystem scale
67 (considering at the same time the mineral soil, the soil organic layer and the trees), the long-
68 term dynamics and impacts of liming on the biogeochemical cycling of nutrients remains
69 poorly understood.

70 Following liming operations using dolostone or calcium carbonate, the Mg concentrations in
71 the soil are expected to increase on the short-term from i) the dissolution of the dolostone (Mg
72 input) and/or ii) induced shifts of biogeochemical processes in the ecosystem (e.g. change in
73 organic matter decomposition, etc.). However, conventional approaches based on
74 measurements of nutrient contents in the ecosystem over time are not sufficient to i)
75 distinguish precisely the native from the non-native (i.e. liming product) sources of Mg and ii)
76 determine the proportion of the input of Mg from the liming that remains in the ecosystem
77 over time.

78 The study of the natural variations of Mg stable isotope ratios coupled with conventional
79 approaches have be proven to be a powerful tool to trace sources and processes in forest
80 ecosystems (Uhlig et al. 2017). In particular, Mg isotope ratios allowed to specify the
81 dynamics of Mg in soil/plant systems and the study of isotopic fractionations of Mg has made
82 it possible to identify specific biological and/or physicochemical processes (Black et al. 2008;
83 Bolou-Bi et al. 2010; Bolou-Bi et al. 2012; Opfergelt et al. 2014; Schmitt et al. 2012; Tipper
84 et al. 2010). Despite Mg isotopic fractionation processes in the soil/plant systems and the
85 small range of isotopic variations at the scale of the globe (-4.6 to 0.8‰, Fig. 1), Mg isotope
86 ratios can, under certain conditions, be used to trace ecosystem sources (Bolou-Bi *et al.*,
87 2012). A sufficient difference of Mg isotopic composition between the liming product and the
88 Mg pools in the forest ecosystem may allow for tracing the fate and distribution of the Mg
89 originating from the dissolution of the liming product, as demonstrated by Bolou-Bi et al.
90 (2016) on two paired forest catchments in the Vosges Mountains (one control and one limed).

91 In a previous study, Court et al (2018) studied with conventional approaches five permanent
92 paired (limed and control) experimental beech (*Fagus sylvatica* L.) plots in Northern France
93 to define and quantify the ecosystem response to liming. The objective of the present study
94 was to investigate the variations of Mg stable isotopes in four of the five experimental sites
95 studied by Court et al (2018) in order to i) better characterize the processes controlling the Mg
96 cycle in forest ecosystems and ii) gain insight on the dynamics of Mg released from the
97 liming product and the biogeochemical cycling changes induced by liming practices.

98

99 2. Material and Methods

100 2.1. Study sites

101 Four experimental liming sites in Northern France were selected for this study to cover a
102 range of soil and climatic conditions: Potées, Humont, Darney and Fougères. The sites were
103 described in detail in a previous study (Court et al. 2018) and only a summarized description
104 is given here. The Fougères site is located in Brittany (oceanic climate), the Darney and
105 Humont sites in the Vosges Mountains (semi-continental climate) and the Potées site in the
106 Ardennes (semi-continental climate). Precipitation was evenly distributed throughout the year
107 for all sites and average annual precipitation calculated over the 1995–2016 period was close
108 to 1000 mm for all sites except Humont which was *ca* 1600 mm. Site elevation ranges from
109 185 m (Fougères) to 570 m (Humont). All sites are situated within state-owned forests and are
110 managed following the local forest management plan. The stands are composed of pure
111 European beech (*Fagus sylvatica* L.) and originated from the natural regeneration of the forest
112 at all sites except Potées (plantation in 1986). Bedrock at each site is sandstone in the Vosges
113 Mountains, shale in the Ardennes, and granodiorite in Brittany. The soils at the different sites
114 classified as Dystric or Luvic Cambisols (WRB 2006) and are acidic and nutrient poor.

115

116 2.2. Liming experimental design

117 At each liming assay site, a similar experimental design was set up prior to the liming
118 operation. Replicated plots were assigned for each treatment, depending on the sites: 2
119 Ca/MgCO₃ and 2 control plots at Potées and Humont; 1 CaCO₃ and 1 control plot at Darney
120 and 4 CaCO₃ and 4 control plots at Fougères. Sampling subplots were then defined within
121 each plot to avoid edge effects and all the measurements and samplings were done within
122 these subplots. More details are available in Court et al (2018).

123 The liming products and the applied amounts were adapted at each site in relation to the soil
124 physical and chemical properties and tree nutrient requirements. The granulometry of the
125 applied liming products was < 2 mm and the products were manually applied (2–3 cross
126 passages to homogenize the product application).

127 At Fougères and Darney, the liming operation consisted of a single application of commercial
128 calcium carbonate (CaCO₃), applied in 1971 at Darney (1070 kg.ha⁻¹ Ca) and in 1973 at
129 Fougères (1070 kg.ha⁻¹ Ca). At Humont, the liming operation consisted of a single
130 application of dolomitic limestone applied in 1991 (541 kg.ha⁻¹ Ca; 228 kg.ha⁻¹ Mg). At

131 Potées, a commercial product (Resilvert®, MEAC) mainly composed of dolomitic limestone
132 with P and K additives was applied in 1995 (543 kg.ha⁻¹ Ca; 120 kg.ha⁻¹ Mg).

133

134 **2.3. Site monitoring: Sampling and elemental analysis**

135 Samples were collected at different dates after the application of the liming product. Pre-
136 treatment samples were only available at the Potées site. The sampled ecosystem
137 compartment (organic layer, mineral soil and green leaves) and the corresponding sampling
138 dates are detailed in Court et al (2018). The physical and chemical analysis of the different
139 samples was performed directly after sampling.

140

141 2.3.1. Organic layer

142 The organic layer samples were collected using a 0.1 m² square frame. Three replicated
143 samples were collected in each plot. The dry weight of the organic layer was measured for all
144 replicates after oven-drying (35°C). Samples were milled and the three replicates were bulked
145 together (one composite sample per plot) for further determinations. C and N contents were
146 measured with a Thermo Quest NCS 2500. P, Mg, Ca and K contents were determined by
147 ICP-AES after sample complete digestion in 50% nitric acid. Results represent the total
148 amounts of nutrients in the organic layer.

149

150 2.3.2. Mineral soil

151 Mineral soil samples were collected at different depths with a soil auger (15 cm long helical
152 auger for soil samples collected before 2000 and 15 cm long, 8 cm diameter cylindrical auger
153 for samples collected after 2000). Three replicated soil profiles were sampled in each plot and
154 a composite sample was generated for each sampled depth. Soil samples were air-dried and
155 sieved (2 mm). Soil physical and chemical properties were analyzed by the INRA-ARRAS
156 laboratory: humidity (NF ISO 11465), soil granulometry distribution (NF X 31-107), pH_{water}
157 (soil water pH – 1:5 soil to water volume ratio) and pH_{KCl} (soil KCl pH – 1:5 soil to 0.1
158 mol.L⁻¹ KCl reagent volume ratio) (NF ISO 10390), C and N content, cationic exchange
159 capacity and exchangeable pools (Mg, Ca, K, Na, Al, H, and Mn) by cobaltihexamine (2.5 g
160 of soil in a 50 mL of a 50 mmol_c.L⁻¹ Co(NH₃)₆³⁺ solution) (Standard method NF X 31-130)
161 and soil total pools by hydrofluoric acid digestion (Mg, Ca, K, Na, Si, Al, Fe, Mn and P)
162 (Standard method NF X 31-147).

163

164 2.3.3. Green tree leaves

165 Green leaves were sampled using a rifle at the end of the vegetation season (late august) from
166 3 branches in the top of the canopy collected from 3 dominant trees. A composite sample was
167 generated for each plot. The nutrient content in green leaves was measured after oven-drying
168 (65°C) and sample milling: C and N content was measured with a Thermo Quest NCS 2500.
169 P, K, Ca, Mg, S, Mn content was determined by ICP-AES after sample digestion with 50%
170 nitric acid. Results represent the total amounts of nutrients in foliage.

171 The majority of samples and liming products were then archived at the “Biogéochimie des
172 Ecosystèmes Forestiers (INRAE)” laboratory.

173

174 2.4. Magnesium isotopic composition analysis

175 2.4.1. Sample preparation

176 The labware used during the treatment of the samples (*e.g.* pipette tips, tubes) was cleaned in
177 an acid bath (2% HNO₃) before use for at least 24 hours and rinsed with ultrapure water (18.2
178 mΩ, PURELAB® Flex). The acids used were bidistilled (DST-1000 Savillex® distiller) to
179 remove any impurities that may contaminate the samples. For sample acid digestion, Teflon®
180 reactors were previously cleaned with concentrated acids in a scrubber (Analab®, ETC EVO
181 II-ST). Sample processing and isotopic analysis were performed in a clean lab to avoid
182 sample contamination.

183 The isotopic composition analysis were performed on a subset of the available samples. The
184 selection of samples was based on the following criteria:

- 185 - Sites and years for which samples from the three studied compartments (soil, humus
186 and tree foliage) were available.
- 187 - The most extreme years (beginning and end of monitoring) and, when possible, an
188 intermediate year were favored.
- 189 - If the three compartments were not sampled within the same year, sampling dates for
190 each compartment were selected as close as possible.

191

192 The selected samples were prepared differently for the isotopic analysis according to their
193 type:

- 194 • For organic samples (green leaves and humus), 200 mg of sample was digested in 6 ml
195 of 69% bidistilled HNO₃ and heated in a microwave oven (Anton Paar® Multiwave
196 3000). The solution was evaporated to dryness and 2 ml of 69% bidistilled HNO₃ were
197 added.
- 198 • For total soil mineralization, the samples were digested in 1 ml of 40% HF for 24
199 hours at room temperature and then 4 ml of 69% bidistilled HNO₃ was added and
200 samples were heated at 120°C for 48 h. The solution was evaporated to dryness and 2
201 ml of 37% bidistilled HCl were added and heated for 4 hours at 120°C. Then, the
202 solution was again evaporated to dryness and 2 ml of 69% bidistilled HNO₃ were
203 added.
- 204 • For soil exchangeable pool extractions, 4 g of sample were placed in a 50 mL Falcon
205 tube with 40 ml of 1M ammonium acetate solution (Fractopur® NH₄OAc, Merck).
206 The solution was continuously shaken during 1 hour and then centrifuged during 20
207 minutes at 3000 rpm. The supernatant was recovered and the extraction was repeated
208 by adding 40 ml of 1M acetate ammonium solution, continuously shaking during 24h
209 and centrifuging during 20 min at 3000 rpm. The exchangeable Mg extraction was
210 done three times for each sample and the three replicates were bulked together in order
211 to obtain a sufficient amount for isotope analysis, filtered (0.45 µm, nylon,
212 Millipore®) and evaporated to dryness. The ammonium acetate was then digested by
213 adding 5 mL of H₂O₂ (30% Suprapur®, Merck), evaporated to dryness and then taken
214 up in 2 mL of 69% bi-distilled HNO₃.

215

216 2.4.2. Sample purification

217 Prior to Mg isotope composition measurements, each sample (soil and organic material
218 digests and soil extracts) was purified according to a procedure adapted from Bolou-Bi et al.
219 (2009). Purified Mg (~30µg) samples were obtained by ion-chromatography using a
220 combination of AG®-MP-1M and AG®-50W-X12 resins:

- 221 • First the sample was eluted through an anion exchange resin (2mL AG®-MP-1M)
222 using 7M HCl to elute cations with a high affinity for the anion exchange resin (e.g.
223 Fe, Cu, Zn)

- 224 • Then the sample was sequentially purified through two cation exchange resin columns
225 (2mL and then 1mL AG®-50W-X12). The samples were eluted with HNO₃ 1M in the
226 first column and with HCl 4M in the second.

227 The purified Mg solutions were evaporated to dryness.

228

229 2.4.3. Isotope ratio analysis

230 Mg stable isotope ratios (²⁶Mg/²⁴Mg and ²⁵Mg/²⁴Mg) were analysed with a multi-collector,
231 inductively couple plasma mass spectrometer (MC-ICPMS) instrument (Neptune Plus,
232 ThermoScientific), at the PARI analytical platform of the “Institut de Physique du Globe de
233 Paris” (IPGP, Paris, France). The samples were diluted to 300 µg.L⁻¹ of Mg in 0.5M HNO₃
234 and introduced into the mass spectrometer through an Apex HF (ESI) equipped with a PFA
235 concentric nebulizer (ESI, 50 µl.min⁻¹). The measurements were carried out with a medium
236 resolution and by block of 25 cycles of 8s. Instrumental mass bias was monitored and
237 corrected with the standard-sample bracketing technique using the DSM3 standard (Galy et al.
238 2003). Isotopic compositions are expressed in δ notation as the part per thousand (‰)
239 deviation from the DSM3 standard (Equation 1):

$$240 \quad (1) \quad \delta^x Mg = \left(\frac{\frac{x_{Mg}}{24_{Mg}}(sample)}{\frac{x_{Mg}}{24_{Mg}}(DSM3)} - 1 \right) \times 10^3$$

241 where x is either mass 26 or 25.

242 Each sample was measured in triplicate (Std-Smpl-Std-Smpl-Std-Smpl-Std). Uncertainties
243 from replicates are reported as two standard deviations (2SD).

244 For Potées (1995 and 2004) and Darney (1995), 0-5 cm and 5-15 cm soil layers were analyzed
245 separately and the isotopic composition of the 0-15cm was calculated (supplementary material
246 Fig. S1). For the other sites, the topsoil layers (0-5cm and 5-15cm) and the deep soil layers
247 (30-45cm and 45-60cm) were bulked together prior to analysis (0-15cm and 30-60cm
248 respectively) to reduce the number of samples to analyze.

249

250 The difference of Mg isotopic composition between control and limed plots is expressed in Δ
251 notation (Equation 2):

$$252 \quad (2) \quad \Delta^{26}Mg_{Limed-ctl} = \delta^{26}Mg_{Limed} - \delta^{26}Mg_{Control} (\text{‰})$$

253 In the same way, the difference of Mg isotopic composition between two compartments A and
254 B is expressed in Δ notation (Equation 3):

$$255 \quad (3) \quad \Delta^{26}Mg_{A-B} = \delta^{26}Mg_A - \delta^{26}Mg_B (\text{‰})$$

256 where A and B represent two different ecosystem compartments (e.g. Foliage (Fol), Organic
257 layer (Org) or Exchangeable soil (Exch)).

258

259 2.4.4. Isotope ratio data validation

260 To assess measurement reproducibility, each sample was analysed several times in different
261 sample sequences, *i.e.* at different dates. To assess measurement accuracy, standard solutions
262 and reference materials were analysed in each sample sequence. The pure Mg ‘Cambridge’
263 standard was measured at the beginning and end of each sample sequence and was used as a
264 reference to validate the results of the sample sequence. When the Cambridge standard
265 measurements differed by more than 0.15‰ from the average value reported in the literature
266 (-2.60‰), the $\delta^{26}\text{Mg}$ and $\delta^{25}\text{Mg}$ values of the samples measured during the same sequence
267 were rejected. For the approved sample sequences, the Cambridge standard measurements
268 were on average $-2.61 \pm 0.09 \text{‰}$ (2SD, n=314). The reference materials that were analysed
269 together with the samples were in good agreement with the accepted values: silicate rock
270 reference material GA (Granite, $\delta^{26}\text{Mg} = -0.43 \pm 0.08 \text{‰}$, n=6), soil reference material Till1
271 (forest soil, $\delta^{26}\text{Mg} = -0.62 \pm 0.38 \text{‰}$, n=7), vegetation reference material BCR 281 (Rye Grass,
272 $\delta^{26}\text{Mg} = -1.06 \pm 0.08 \text{‰}$, n=13) and sea water (BCR 403, $\delta^{26}\text{Mg} = -0.88 \pm 0.11 \text{‰}$, n=29), which
273 validates both the Mg extraction protocol and the MC-ICP-MS analysis.

274 In addition, two indicators were used to check individual sample measurements:

275 1. To check the linearity of instrumental mass bias drift during a sequence, the $\delta^{26}\text{Mg}$
276 variation between two successive measurements of the DSM3 standard solution
277 ("Dead Sea Magnesium") was estimated. Individual sample measurements were
278 rejected if at least one of the two DSM3 bracketing showed a variation exceeding
279 0.15‰.

280 2. To check for mass-dependent fractionation (and thus for absence of isobaric
281 interferences), each individual measurement was represented in a plot of $\Delta^{25}\text{Mg}'$ vs.
282 $\delta^{26}\text{Mg}'$ (Young & Galy 2004), where (Equation 4):

$$283 \quad (4) \quad \delta^x Mg' = 1000 \times \log \left(\frac{\delta^x Mg + 1000}{1000} \right)$$

284 and

$$285 \quad \Delta^{25}\text{Mg}' = \delta^{25}\text{Mg}' - 0.521 \times \delta^{26}\text{Mg}'$$

286 An individual measurement was rejected if, in this plot, the distance between the
287 measurement and the kinetic (slope = 0.511) or equilibrium (slope = 0.521) theoretical
288 mass fractionation line was greater than 0.01.

289

290 **2.5. Modeling Mg isotopic variations**

291 In order to investigate the reasons underlying the isotopic differences between the limed and
292 control plots, a simple pool and flux model taking into account isotopic fractionation
293 processes was developed (supplementary material Fig. S2). The numerical model simulates
294 the annual change of the isotopic composition of the exchangeable pool of Mg in the 0-15 cm
295 soil layer as a function of 1) the input fluxes of Mg to this layer (atmospheric, mineral
296 weathering and organic layer sources) and their respective isotopic composition and 2) the
297 isotopic fractionation processes that may occur through the output fluxes (root absorption, Mg
298 leaching). In the limed plots, the model takes into account the input of Mg from the liming
299 product and its isotopic composition and assumes that all lime-derived Mg is released within
300 the first year.

301 The model was then used to test different possible hypotheses for the Darney and Potées sites
302 that are listed in supplementary material Table S3. Depending on the hypotheses tested, the
303 model was sequentially run for a range of input variables (e.g. tree uptake and leaching Mg
304 flux, isotopic fractionation factors). For each set of input parameters, the model calculates
305 either the possible values for the isotopic fractionation factors or the possible values for the
306 input sources in order to reproduce the isotopic composition of the Mg exchangeable pool in
307 both plots. This modelling approach could not be carried out for the Fougères and Humont
308 sites because there were too few isotope data at these sites.

309 At the Potées site, where the pre-treatment data was available for the limed plot and for more
310 pools, and at which flux and isotope data were available for both treatment plots, the model
311 simulated the isotopic composition of the soil exchangeable pool of Mg in the lime plot over
312 the 1995 to 2004 period following the measured changes in the exchangeable Mg pool of the
313 0-15cm soil layer.

314 At the Darney site, less data was available. In order to overcome the lack of data on the Mg
315 fluxes, their isotopic composition, and the associated isotopic fractionation processes, the

316 model was run to simulate both the isotopic composition of the soil exchangeable Mg in the
317 control and limed plots over the 1971 to 1995 period by 1) assuming that the exchangeable
318 Mg pool in the control plot and its isotopic composition remained constant over the period
319 (steady state), 2) assuming that model parameters related to Mg isotopic fractionation were
320 the same for both plots, and finally 3) calculating the differences of Mg fluxes between plots
321 in order to respect the measured change in Mg exchangeable pool in the limed plot.

322

323 **3. Results**

324 **3.1. Comparison with previous published data on terrestrial ecosystems**

325 The $\delta^{26}\text{Mg}$ values measured in the control and limed plots at each of the four forest
326 ecosystems were within the previously reported range of $\delta^{26}\text{Mg}$ variation (Fig. 1) for bulk
327 soil, soil exchangeable pool (-0.9 to -0.3‰ for the control plots; -1.4 to -0.7‰ for the limed
328 plots), organic layer and tree foliage (-1.4 to -0.4‰ for the control plots; -1.8 to -0.9‰ for the
329 limed plots) (Table 1). The $\delta^{26}\text{Mg}$ values of bulk soils (-0.8 to -0.6‰) were only measured at
330 Potées and were close to the lowest values observed in terrestrial ecosystems. The range of
331 $\delta^{26}\text{Mg}$ for organic layer samples measured in this study (-0.7 to -0.3‰ for the control plots; -
332 0.7 to -0.0‰ for the limed plots) was narrower than that of soil exchangeable pools and leaves
333 were close to previously published values (-0.54 to -0.20‰). Unfortunately, not all the
334 applied liming products were archived and Mg isotope analyses was possible only for the
335 product applied at Potées ($\delta^{26}\text{Mg} = -1.60\text{‰}$) and Humont ($\delta^{26}\text{Mg} = -1.56\text{‰}$). This isotopic
336 composition was within the reported range of variation of dolomite (-2.2‰ to -1.1‰).

337

338 **3.2. Mg isotope compositions of green leaves, soils and organic layer in the control** 339 **plots**

340 In the control plots, differences between $\delta^{26}\text{Mg}$ values of different compartments were, on
341 average, noticeable between sites and may be ranked as follow (Fig. 2):

- 342 - Potées > Humont > Fougères and Darney for green leaves
- 343 - Humont > Darney > Potées and Fougères for soil exchangeable pools
- 344 - Darney > Humont and Potées > Fougères for organic layer

345 The foliar $\delta^{26}\text{Mg}$ was lower than that of the [0-15cm] exchangeable pool of the soil at all sites
346 and years, except at Potées in 1995 (Ctl pre-treatment plot, $\Delta^{26}\text{Mg}_{\text{Fol-Exch}} = 0.05\text{‰}$; Ca/MgCO_3
347 pre-treatment plot, $\Delta^{26}\text{Mg}_{\text{Fol-Exch}} = 0.06\text{‰}$) and 2004 ($\Delta^{26}\text{Mg}_{\text{Fol-Exch}} = 0.12\text{‰}$) (Table 2).

348 The organic layer was enriched in ^{26}Mg isotope compared to the foliage at all sites and years,
349 except at Potées in 2004 ($\Delta^{26}\text{Mg}_{\text{Fol-Org}} = 0.08\text{‰}$). At the four sites, the $\delta^{26}\text{Mg}$ of the organic
350 layer was slightly higher than the $\delta^{26}\text{Mg}$ of the [0-15cm] exchangeable pool of the soil, or
351 very close ($\Delta^{26}\text{Mg}_{\text{Org-Exch}}$ ranging from -0.11 to 0.18‰).

352

353 **3.3. Relationship between Mg concentration and Mg isotope composition in the** 354 **control plots**

355 In the control plots, differences in the foliar Mg concentrations were, on average, noticeable
356 between sites and may be ranked as follow: Fougères > Potées > Humont > Darney (Table 1).

357 Across the sampling sites, a positive relationship was observed between Mg concentrations
358 and $\delta^{26}\text{Mg}$ in the foliage in the control plots (Fig. 3), with Potées having the highest foliar Mg
359 concentration and $\delta^{26}\text{Mg}$ values, and Darney the lowest foliar Mg concentrations and $\delta^{26}\text{Mg}$
360 values. At Fougères, the intercept of the relation between foliar Mg concentrations and $\delta^{26}\text{Mg}$
361 was different (with higher foliar Mg concentrations compared to the other sites) but the slope
362 of the trend was very similar. The lowest Mg concentration and $\delta^{26}\text{Mg}$ in leaves at Fougères
363 were observed in 2005 whereas the highest were recorded in 2014.

364 In the control plots, the differences of $\delta^{26}\text{Mg}$ values between the organic layer and foliage
365 ($\Delta^{26}\text{Mg}_{\text{Fol-Org}}$) and between the [0-15cm] exchangeable pool of the soil and foliage ($\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$)
366 were larger when the foliar Mg concentration was low (Table 2 and supplementary
367 material Fig. S4). For the Mg isotope differences between compartments ($\Delta^{26}\text{Mg}_{\text{Fol-Org}}$ and
368 $\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$), the sites were ranked as follow: Fougères and Potées < Humont < Darney. No
369 trend was observed between Mg concentrations and $\delta^{26}\text{Mg}$ or the Mg isotope differences
370 between compartments ($\Delta^{26}\text{Mg}$) for the organic pool and the [0-15cm] exchangeable pool of
371 the soil.

372

373 **3.4. Mg isotope variations between limed and control plots**

374 **3.4.1. Foliage**

375 For all sites foliage $\delta^{26}\text{Mg}$ of the limed plots were generally lower than those of the control
376 plots. The foliar $\delta^{26}\text{Mg}$ of the limed plots were ranked as follow: Potées > Fougères >
377 Humont > Darney (Fig. 2). Similar to the control plots, a high spatial and/or temporal
378 variability of the foliar $\delta^{26}\text{Mg}$ values were observed throughout the study period (Table 1). As
379 for the control plots, a positive relationship was observed between foliar Mg concentrations

380 and $\delta^{26}\text{Mg}$ values (Fig. 3). The foliar $\Delta^{26}\text{Mg}_{\text{Limed-Ctl}}$ was negative for all sites except Fougères
381 in 2005 ($\Delta^{26}\text{Mg}_{\text{Limed-Ctl}} = 0.08\text{‰}$) (Fig. 4). It increased gradually over time at Potées and
382 Humont whereas it was stable over time at Fougères and Darney. As for the control plots, the
383 $\delta^{26}\text{Mg}$ of foliage was lower than that of the [0-15cm] exchangeable pool of the soil in the
384 limed plots at all sites (Fig. 2 and Table 2), except at Potées in 2004 ($\Delta^{26}\text{Mg}_{\text{Fol-Exch}} = 0.26\text{‰}$).
385 The trend between the $\Delta^{26}\text{Mg}_{\text{Fol-Org}}$ and foliar Mg concentration was not observed in the limed
386 plots (Table 2 and supplementary material Fig. S4). As for the control plots, the $\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$
387 was larger when the foliar Mg concentration was low in the limed plots, except at Potées in
388 2004. In terms of $\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$, the sites were ranked as follow: Darney > Fougères > Potées
389 and Humont.

390

391 3.4.2. Organic layer

392 In the limed plots, the $\delta^{26}\text{Mg}$ values of the organic layer were ranked as follow: Humont >
393 Darney > Potées and Fougères (Fig. 2). The $\Delta^{26}\text{Mg}_{\text{Limed-Ctl}}$ of the organic layer was close to
394 zero at Potées in 2004 and in 2016 ($\Delta^{26}\text{Mg}_{\text{Limed-Ctl}} = -0.06\text{‰}$) and at Darney in 2016
395 ($\Delta^{26}\text{Mg}_{\text{Limed-Ctl}} = 0.01\text{‰}$), slightly positive at Fougères in 2014 ($\Delta^{26}\text{Mg}_{\text{Limed-Ctl}} = 0.14\text{‰}$), and
396 widely positive at Humont in 2016 ($\Delta^{26}\text{Mg}_{\text{Limed-Ctl}} = 0.40\text{‰}$) (Fig. 4). As for the control plots,
397 the organic layer was enriched in ^{26}Mg compared to the foliage in limed plots but this
398 enrichment in ^{26}Mg was larger in the limed plots than in the control plots (Fig. 2 and Table 2).
399 No trend was observed between Mg concentration and $\delta^{26}\text{Mg}$ nor the Mg isotope differences
400 between compartments ($\Delta^{26}\text{Mg}$) for the organic pool in limed plots.

401

402 3.4.3. Soils

403 In the limed plots, the $\delta^{26}\text{Mg}$ values of the [0-15cm] exchangeable pool of the soil were
404 ranked as follow: Fougères and Darney > Potées > Humont (Fig. 2). Similar to the control
405 plots, high spatial and/or temporal variability of the $\delta^{26}\text{Mg}$ was also observed for the [0-15cm]
406 exchangeable pool of the soil at Potées throughout the study period (Table 1). The $\Delta^{26}\text{Mg}_{\text{Limed-}}$
407 Ctl of the [0-15cm] exchangeable pool of the soil were negative for all sites except Fougères
408 ($\Delta^{26}\text{Mg}_{\text{Limed-Ctl}} = 0.07\text{‰}$ in 1994) (Fig. 4). Humont had the largest $\Delta^{26}\text{Mg}_{\text{Limed-Ctl}}$ (-1.01‰, 25
409 years after liming). At Potées, the $\Delta^{26}\text{Mg}_{\text{Limed-Ctl}}$ was highest in 2004 (-0.68‰) and decreased
410 to -0.34‰ in 2016. The $\Delta^{26}\text{Mg}_{\text{Limed-Ctl}}$ were smaller at Darney (mean $\Delta^{26}\text{Mg}_{\text{Limed-Ctl}} = -0.3\text{‰}$),
411 and did not vary over time. The $\delta^{26}\text{Mg}$ of the organic layer was higher than that of the [0-

412 15cm] exchangeable pool of the soil at all sites for the limed plots, and at Potées (2004) and
413 Darney for the control plot (Fig. 2 and Table 2). Like the control plots, no trend was observed
414 between Mg concentration and $\delta^{26}\text{Mg}$ or the Mg isotope differences between compartments
415 ($\Delta^{26}\text{Mg}$) and the [0-15cm] exchangeable pool of the soil in limed plots.

416

417 **4. Discussion**

418 **4.1. Variations of the Mg isotope composition in forest ecosystems and possible** 419 **controlling factors**

420 4.1.1. Inter-site variability of weathering and atmospheric Mg input fluxes

421 The Mg isotopic composition of the 0-15cm exchangeable pool varied quite widely between
422 the different studied sites. This variability could be partly due to the inter-site variability of
423 the soil mineral weathering flux and isotopic composition. We do not have the $\delta^{26}\text{Mg}$ values
424 of the bedrocks underlying the four sites studied here. However, Darney and Humont are
425 developed on Vosgian sandstone and Bolou-Bi et al. (2012) measured the $\delta^{26}\text{Mg}$ values of
426 this bedrock (+0.13‰). In the same study, Bolou-Bi et al. (2012) also reported $\delta^{26}\text{Mg}$ values
427 of the bulk soil that varied from -0.48‰ in the topsoil to +0.25‰ in the E horizon (30 cm
428 depth) and +0.00‰ in the C horizon (110 cm depth). No data in this study or in the literature
429 was available to characterize the isotopic signature of the loessic sedimentary deposits at
430 Fougères and Potées but fine-grained sedimentary rocks are generally enriched in the light
431 isotope ^{24}Mg compared to silicate rocks (Bolou-Bi et al. 2010). Under this assumption, the
432 Mg mineral weathering flux in the 0-15cm soil layer would be more enriched in ^{24}Mg at
433 Fougères and Potées compared to Darney and Humont which would contribute to explaining
434 the observed inter-site $\delta^{26}\text{Mg}$ differences.

435 Though the Mg isotopic composition of the ocean and of the seawater-derived component of
436 atmospheric inputs is relatively constant (between -0.7 and -0.9 ‰), the Mg isotopic
437 composition of atmospheric inputs may vary from -1.05 to -0.51‰ (Bedel 2015; Bolou-Bi et
438 al. 2012; Pogge von Strandmann et al. 2008; Tipper et al. 2010). No data is available to
439 conclude on this issue, but Mg atmospheric inputs and their isotopic composition may also
440 contribute to explain the observed inter-site $\delta^{26}\text{Mg}$ differences.

441

442 4.1.2. Relation between Mg plant-availability and Mg isotope variation

443 Similar to the soil, notable differences in foliar $\delta^{26}\text{Mg}$ values were observed between the
444 control plots of the different studied sites. The range of variation of foliar $\delta^{26}\text{Mg}$ values was
445 broader than that of the soil exchangeable pool and the foliage compartment was
446 systematically enriched in ^{24}Mg compared to the soil exchangeable pool (except at Potées in
447 1995 and 2004). Previous studies on low-chemical fertility forest ecosystems have shown that
448 Mg exchangeable pools in the top layers of the soil profile represent the main source of Mg
449 for tree uptake (van der Heijden et al. 2015). Under this assumption, the $\delta^{26}\text{Mg}$ values of the
450 [0-15cm] exchangeable pool alone could not explain the foliar $\delta^{26}\text{Mg}$ values. Therefore, it is
451 most likely that isotopic fractionation processes occurring in the plant and/or between the soil
452 and the foliage explain the $\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$ observed in our data set, as suggested previously by
453 several authors (Black et al. 2008; Bolou-Bi et al. 2010; Schmitt et al. 2012).

454 The $\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$ varied between sites and the greatest values were observed at Humont and
455 Darney where exchangeable Mg contents in the topsoil were amongst the lowest of the four
456 studied sites. The $\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$ was positively related to the foliage Mg concentration
457 (supplementary material Fig. S4), which is commonly assumed to be a good indicator of the
458 availability of nutrients in the ecosystem (Bonneau 1995). When the Mg availability in the
459 soil decreases, the plant-internal recycling of Mg through the re-translocation processes
460 occurring prior to litterfall contributes increasingly to sustaining Mg nutrition. Re
461 translocation is known to fractionate Mg isotopes (Bedel 2015; Black et al. 2007; Bolou-Bi et
462 al. 2016; Bolou-Bi et al. 2012; Kimmig et al. 2018; Kraj 2015; Matile 2000; Moynier & Fujii
463 2017). However, it is unlikely that foliar re-translocation dynamics explain the relation
464 between $\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$ and Mg concentration in foliage because it is ^{26}Mg that is preferentially
465 transferred from the leaf to the younger tissues and/or to storage and reproductive organs of
466 the tree (Bolou Bi et al, 2010). This suggests that low Mg availability in the soil may
467 influence the isotopic fractionation processes that occur along the Mg uptake and allocation
468 paths from the soil pools to the canopy.

469

470 4.1.3. Evidence of specific Mg isotope fractionation processes occurring in the 471 organic layer and inheritance to the soil

472 Our results showed that the organic layer at each of the four sites was almost systematically
473 the most ^{26}Mg -enriched compartment in the ecosystem. To our knowledge, few studies have
474 investigated the Mg isotope composition of organic layers in forest ecosystems (Bolou Bi
475 2009; Mavromatis et al. 2014) and no mechanisms have yet been proposed to explain how the

476 organic layer becomes enriched in ^{26}Mg compared to the other compartments. In forest
477 ecosystems, the Mg pool in the organic layer originates mostly from leaf litter inputs and
478 atmospheric inputs that are retained on the cationic exchange capacity of the organic layer
479 (van der Heijden et al. 2014). The $\delta^{26}\text{Mg}$ values of atmospheric inputs ranges from -1.05 to -
480 0.51‰ (Bedel 2015; Bolou-Bi et al. 2012; Pogge von Strandmann et al. 2008; Tipper et al.
481 2010) and was lower than the $\delta^{26}\text{Mg}$ values of organic layers at all sites (except at Fougères).
482 Furthermore, previous study in forest ecosystems highlight that leaf litter is slightly enriched
483 in ^{24}Mg compared to green leaves due to re-translocation of the foliar Mg during the growing
484 season (Bedel 2015; Black et al. 2007; Bolou-Bi et al. 2016; Bolou-Bi et al. 2012; Kimmig et
485 al. 2018; Kraj 2015; Matile 2000; Moynier & Fujii 2017), which would imply that the $\delta^{26}\text{Mg}$
486 values of leaf litter at our four sites (not measured) was equal or below the measured foliar
487 $\delta^{26}\text{Mg}$ values. It would thus not be possible to obtain the $\delta^{26}\text{Mg}$ values measured in the
488 organic layer from the simple isotopic mixing of these two sources (*i.e.* leaf litter and
489 atmospheric inputs).

490 Therefore, to explain the observed ^{26}Mg enrichment in the organic layer, we hypothesized that
491 one or several processes fractionating Mg isotopes occur in this layer during the
492 mineralization and ageing of organic matter (*i.e.* leading to the preferential release of ^{24}Mg ,
493 for example due the capacity of fungi to fractionate stable isotopes of magnesium when
494 mobilizing base cations from an organic substrate; Fahad *et al.*, 2016; Pokharel *et al.*, 2017)
495 and/or the transfer of atmospheric Mg inputs through the layer with a preferential
496 adsorption/binding of ^{26}Mg on the cationic exchange capacity developed by the organic layer
497 (Bolou-Bi et al, 2010; Chang, 2004). Under this hypothesis, these isotopic fractionation
498 processes would most likely vary depending on the organic matter turnover rate and thus on
499 the climatic and edaphic conditions.

500 Interestingly, the sites with the highest organic layer $\delta^{26}\text{Mg}$ values (Darney and Humont) also
501 have the highest $\delta^{26}\text{Mg}$ values of [0-15cm] exchangeable pools. In forest ecosystems, the
502 annual Mg flux released from the decomposition of the organic layer may represent a large
503 input flux to the topsoil compared to atmospheric inputs and mineral weathering. The Mg
504 isotopic composition of the topsoil Mg exchangeable pool is thus most probably greatly
505 influenced by the Mg isotopic signature of the organic layer.

506

507 **4.2. Effect of liming on Mg cycling and isotope composition**

508 Calcium carbonate was applied at Fougères and Darney while dolomitic limestone
509 (Ca/MgCO_3) was applied at Potées and Humont. We may consider that these products were
510 probably completely dissolved in the first years following the application, due to rainfall and
511 the high acidity of the organic and topsoil layers ($\text{pH}<5$). The isotopic composition of the
512 dolomitic limestone was close to that measured by Bolou-Bi et al. (2016) (-1.75‰) and within
513 the reported range of variation of dolomite 2.5‰ to -1‰ (Brenot et al. 2008; Jacobson et al.
514 2010). In contrast to dolomite lime, we were unable to perform analysis on the applied
515 calcium carbonate, but the Mg content of such products is generally low ($\text{MgCO}_3 < 5\%$) with
516 $\delta^{26}\text{Mg}$ values ranging from -5‰ to -4‰ (Galy et al. 2002). The $\delta^{26}\text{Mg}$ values of the [0-15cm]
517 exchangeable pool and organic layer of the control plots (- 1‰ to -0.3‰) were always higher
518 than that of applied products, while the range of variation for foliage $\delta^{26}\text{Mg}$ values sometimes
519 overlapped it slightly (- 1.4‰ to -0.4‰), without compromising the tracing of Mg originating
520 from the dissolution of the products.

521

522 4.2.1. Effect of dolomitic limestone (Ca/MgCO_3)

523 At the sites limed with dolomitic limestone, we observed lower $\delta^{26}\text{Mg}$ values and higher Mg
524 concentrations in the [0-15cm] exchangeable pool and foliage in the limed plots compared to
525 the control plots. The Mg isotopic approach provides here additional and useful information
526 to distinguish the native from the non-native sources of Mg and our results strongly suggest
527 that a significant part of foliar Mg originated from the dissolved dolomite and that the
528 dolomite-derived Mg was quite rapidly incorporated in the biological cycle. The biological
529 cycling of Mg then contributed to preserving the dolomitic Mg in the soil-plant system, and
530 thus improving tree nutrition on the mid to the long-term. Bolou-Bi et al. (2016) showed
531 similar patterns over a 10-years period after a dolomitic lime application in a limed forested
532 catchment located in the Vosges Mountain. However, the efficiency of the biological cycle in
533 maintaining the dolomite-derived Mg in the topsoil seemed to be limited at Potées compared
534 to Humont: the $\delta^{26}\text{Mg}$ values of the topsoil exchangeable pools was significantly influenced
535 by dolomite-derived Mg at Humont more than 15 years after liming and to a much lesser
536 extent at Potées. Interestingly, the input-output budgets computed by Court et al. (2018) at
537 Potées and the Mg concentrations in the different ecosystem compartments showed a small
538 recovery of Mg released from the liming products (39% of Mg applied recovered in the
539 ecosystem in 2004 and 12% in 2016), with a null recovery for soil in 2016. Court et al. (2018)
540 hypothesized that a large fraction of the liming product was leached out of the ecosystem, but

541 the $\delta^{26}\text{Mg}$ values of the [0-15cm] exchangeable pool was still lower in the limed plot than in
542 the control plot 19 years after the product application. The modelling of the isotopic variations
543 (supplementary material Table S3) corroborates the hypothesis of elevated leaching losses
544 either by a leaching flush occurring in the first years after the application or by a leaching flux
545 associated with an isotopic fractionation (-0.2 to -0.6 ‰: preferential leaching of ^{24}Mg).

546

547 4.2.2. Effect of calcium carbonate (CaCO_3)

548 At Darney, the $\delta^{26}\text{Mg}$ values and the Mg content of the [0-15cm] exchangeable pool were
549 lower in the limed plot than in the control plot over the study period. Although these
550 differences could be due to spatial variability between the limed and control plots (the
551 available data was insufficient to fully dismiss such an effect), the lower $\delta^{26}\text{Mg}$ values in the
552 [0-15cm] exchangeable pool of the limed plot may be related to a shift in the biogeochemical
553 cycling of Mg occurring after the lime application. Several related hypotheses were tested
554 with the numerical modelling approach (supplementary material Table S3) and the results
555 show that the lower $\delta^{26}\text{Mg}$ values in the topsoil in the limed plot is most likely explained by a
556 small amount of Mg contained in the applied calcium carbonate, with an isotope signature of
557 $\delta^{26}\text{Mg} \sim 4.5\text{‰}$. The lower foliar $\delta^{26}\text{Mg}$ values observed in the limed plot compared to the
558 control plot supports this hypothesis and may be partly associated to the $\delta^{26}\text{Mg}$ values
559 variations of the exchangeable pool at this site.

560 At Fougères, no difference in terms of exchangeable $\delta^{26}\text{Mg}$ values was observed between the
561 limed and the control plots, most likely because the calcium carbonate applied at the soil
562 surface did not contain any significant amount of Mg. It is unlikely that the change in foliar
563 $\delta^{26}\text{Mg}$ values observed in the limed plot at this site may be explained by an input of Mg
564 originating from the liming product. Court et al. (2018) showed that liming with calcium
565 carbonate induced on the long term (> 40 years) at Fougères and Darney a decrease in
566 exchangeable Mg content in the topsoil and foliar Mg concentrations. We may hypothesize
567 that a change in tree nutrition strategy occurred after liming (uptake distribution within the
568 soil profile) and/or, as discussed previously (see section 4.1.2), that the decreasing availability
569 of Mg in the soil changed the isotope fractionation processes in the plant, resulting in a
570 decrease of foliar $\delta^{26}\text{Mg}$ values at both sites (supplementary material Fig. S4).

571

572 4.2.3. Organic layer Mg isotope variations induced by liming

573 The only noticeable difference of $\delta^{26}\text{Mg}$ values between limed and control plots for the
574 organic layer was observed at the Humont site, with larger $\delta^{26}\text{Mg}$ values in the limed plot 25
575 years after liming ($\Delta_{\text{Limed-Ctrl}}^{26}\text{Mg} = 0.39\text{‰}$ in 2016). We previously hypothesized that isotope
576 fractionation processes may occur within the organic layer (see section 4.1.3), leading to an
577 enrichment of this layer in ^{26}Mg . The enhancement of the organic matter mineralization
578 induced by liming (Court et al. 2018; Forey et al. 2015) may have resulted in an enrichment of
579 the organic layer in ^{26}Mg (through the preferential release of ^{24}Mg). This hypothesis is
580 supported by the strong decrease of the organic layer dry weight at Humont (strongest
581 decrease among all four sites, Court et al. (2018)) in the limed plot (2.09 kg.m^{-2}) compared to
582 the control plot (5.55 kg.m^{-2}) which suggests that liming strongly impacted the dynamics of
583 organic matter decomposition at this site.

584 For the other sites, given the lower foliar $\delta^{26}\text{Mg}$ values and exchangeable $\delta^{26}\text{Mg}$ values
585 (Potées and Darney) and the lower organic layer dry weight (Fougères and Darney) in the
586 limed plot compared to the control plot, we may have expected to observe a difference in the
587 $\delta^{26}\text{Mg}$ values of the organic layer between limed and control plots. The changes in
588 biogeochemical cycling induced by liming (increase of the mineralization rate of organic
589 matter and of soil pH, Mg retranslocation before leaf abscission...) could partly explain the
590 stability of the organic layer $\delta^{26}\text{Mg}$ values in the limed plot at these sites over time.

591

592 **5. Conclusion**

593 Biological cycling has been shown to have a great impact on stable Mg isotope ratio
594 variations in terrestrial ecosystems and it has been argued that the range of Mg isotopic
595 variations related to biotic processes is wider than the range related to abiotic processes. The
596 $\delta^{26}\text{Mg}$ data from the present study also suggests that observed Mg isotopic variations in forest
597 ecosystems are strongly controlled/influenced by biotic processes along the biological cycle
598 (plant uptake and plant internal cycling, litterfall and organic matter decomposition). Our
599 results also highlight the high $\delta^{26}\text{Mg}$ values of the organic layer, most likely explained by Mg
600 isotope fractionation processes occurring within this layer (mineralization/ageing of organic
601 matter, preferential retention/binding of ^{26}Mg). The results of this study show that future
602 research should focus on understanding Mg isotope dynamics and their controlling factors in
603 two key components of the biogeochemical cycle of Mg: the uptake and transport of Mg from
604 the roots to the foliage and the supply, the storage and the release of Mg in the organic layer.

605 At plots where dolomitic lime was applied, the Mg content in the soil exchangeable pool,
606 leaves foliage and organic layer increased in the 10 to 15 years after liming and then
607 decreased slowly. The results from this isotopic tracing approach concur with the temporal
608 pattern of response to liming observed through conventional elemental approaches: dolomite-
609 derived Mg contributed to increase foliar concentrations and plant-available pools in the
610 topsoil during the 10 to 15 years after the liming which then gradually decreased. However,
611 magnesium isotopic variations also enable to demonstrate the incorporation of dolomite-
612 derived Mg in the biological cycling and its retention on the mid to long term in the soil-plant
613 system (at least 15 to 25 years after liming). At the sites limed with calcium carbonate, the
614 conventional approaches showed a decrease in exchangeable Mg content in the topsoil and
615 foliar Mg concentrations on the long term (30 to 45 years after liming). In the same time, a
616 decrease of exchangeable and foliar $\delta^{26}\text{Mg}$ values was observed in the limed plots, suggesting
617 that the applied product contained a small amount of Mg and/or that Mg cycling changed after
618 liming, to cope in particular with the low Mg availability through shifts in soil organic matter
619 mineralisation rates or in tree nutrition strategy.

620 The results of this study also highlight that, because carbonate liming has multiple effects on
621 soil processes, reservoirs and nutrient cycling, conventional approaches (based on
622 measurements of the content of Mg and of other nutrients such as Ca in the ecosystem over
623 time) are most often insufficient to identify the direct effects of liming on the Mg cycle (i.e.
624 the release of Mg contained in the liming product) from the indirect effects (i.e. the release
625 and/or displacement of native Mg induced by liming). The isotopic approach coupled with
626 numerical modelling tools provides additional information on the fate of the Mg released by
627 liming products in the ecosystem. Because the difference in Mg isotopic signature between
628 carbonate rocks and Mg pools in forest ecosystems is most often sufficiently big, the Mg
629 released from the liming product can be traced in the ecosystem and its contribution to the
630 different pools of Mg in the ecosystem can be quantified.

631

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642

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795

796

797 **Figure captions**

798

799 **Fig.1.** Range of $\delta^{26}\text{Mg}$ variations in compartments of terrestrial ecosystems. Literature data*
800 are presented for atmospheric deposition (dark blue), vegetation (green), soils (red), waters
801 (light blue) and rocks (brown); data of this study are presented in white (control plots) and
802 black (limed plots).

803 *(*Bedel 2015; Bizzarro et al. 2005; Bolou-Bi et al. 2016; Bolou-Bi et al. 2009; Bolou-Bi et al. 2012;*
804 *Brenot et al. 2008; De Villiers et al. 2005; Kimmig et al. 2018; Opfergelt et al. 2014; Teng et al.*
805 *2007; Wiechert & Halliday 2007; Young & Galy 2004*).

806

807 **Fig.2.** $\delta^{26}\text{Mg}$ variations in the different ecosystem compartments of the control (blue) and
808 limed (dark red) plots of the studied sites: Potées (a), Humont (b), Fougères (c) and Darney
809 (d).

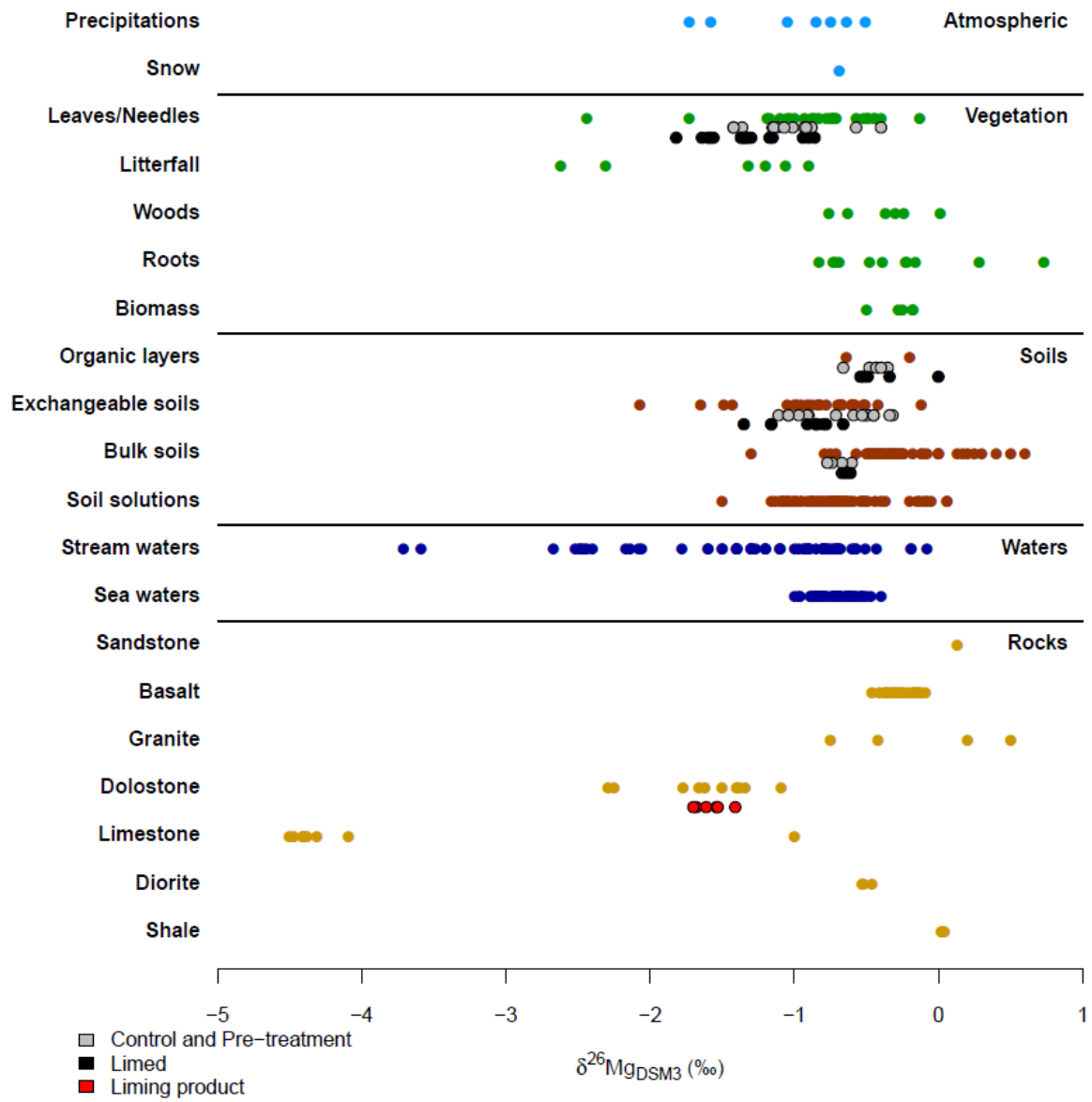
810

811 **Fig.3.** Relationship between $\delta^{26}\text{Mg}$ and Mg foliar concentration in the control (blue) and
812 limed (red) plots of the four study sites: Regression lines are presented in dashed lines for
813 control plots (blue), limed plots (red) and control + limed plots (black).

814

815 **Fig.4.** Differences in Mg concentration (empty markers) and $\delta^{26}\text{Mg}$ (filled markers) between
816 limed and control plots ($\Delta_{\text{Limed-Control}}^{26}\text{Mg}$) as a function of time after liming for the different
817 ecosystem compartments of the four sites: [0-15cm] exchangeable soils (a), organic layers (b)
818 and green leaves (c).

819

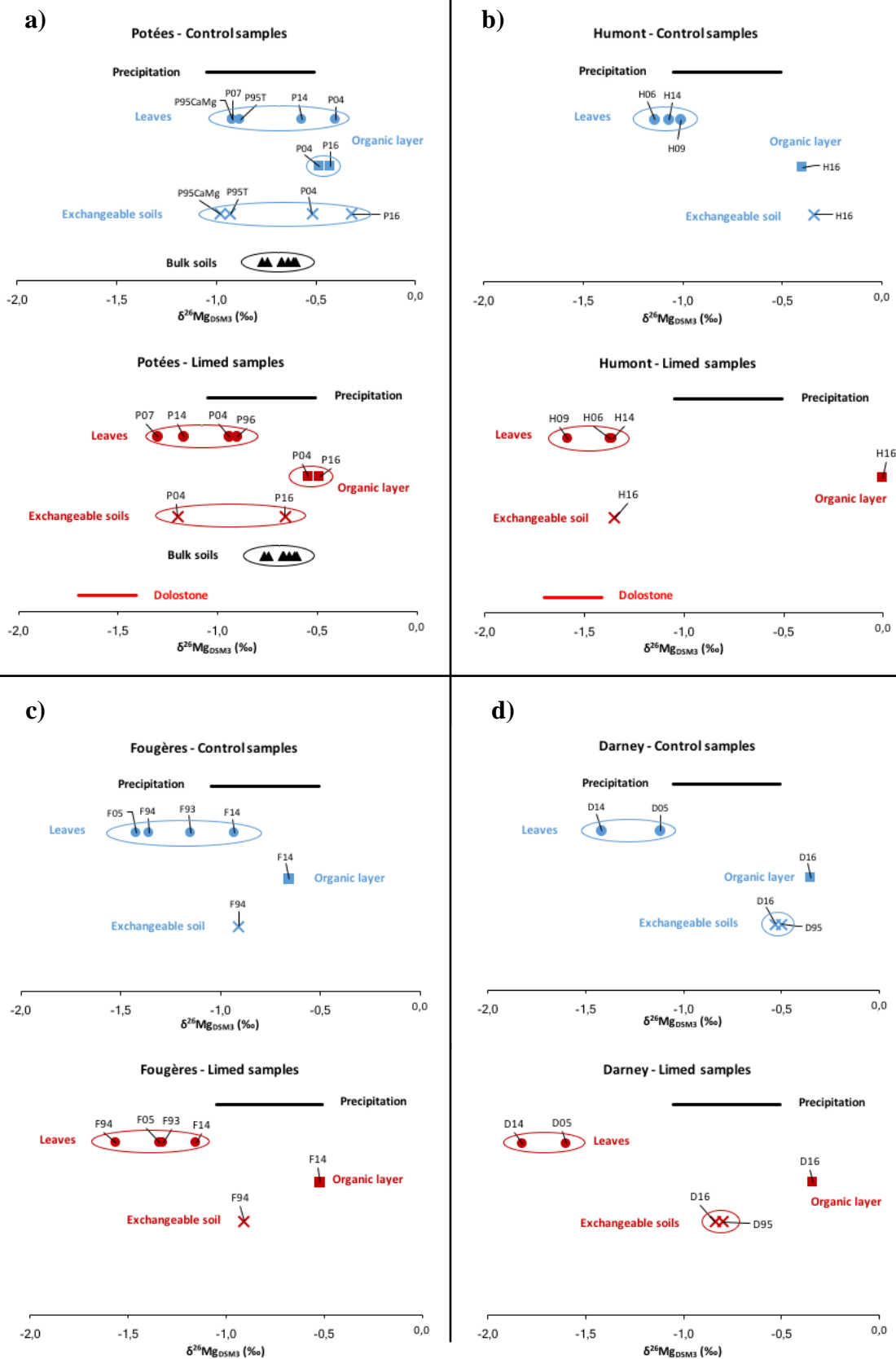


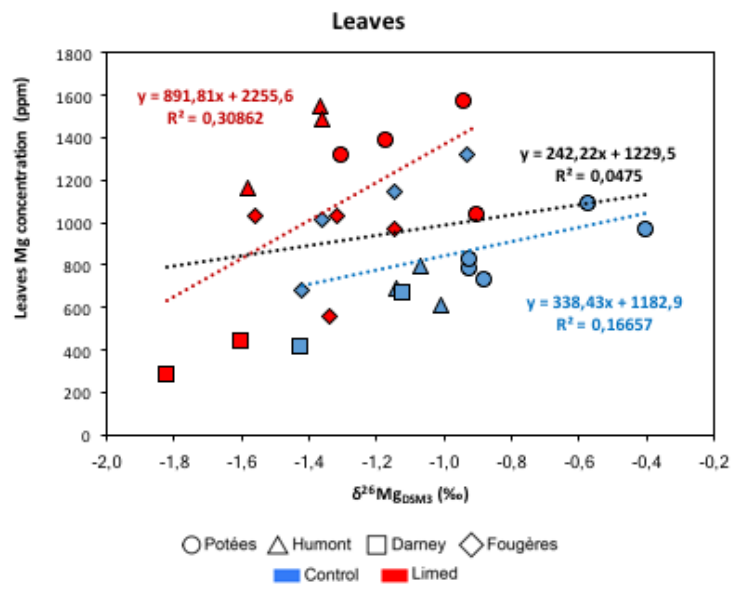
822 Fig.2

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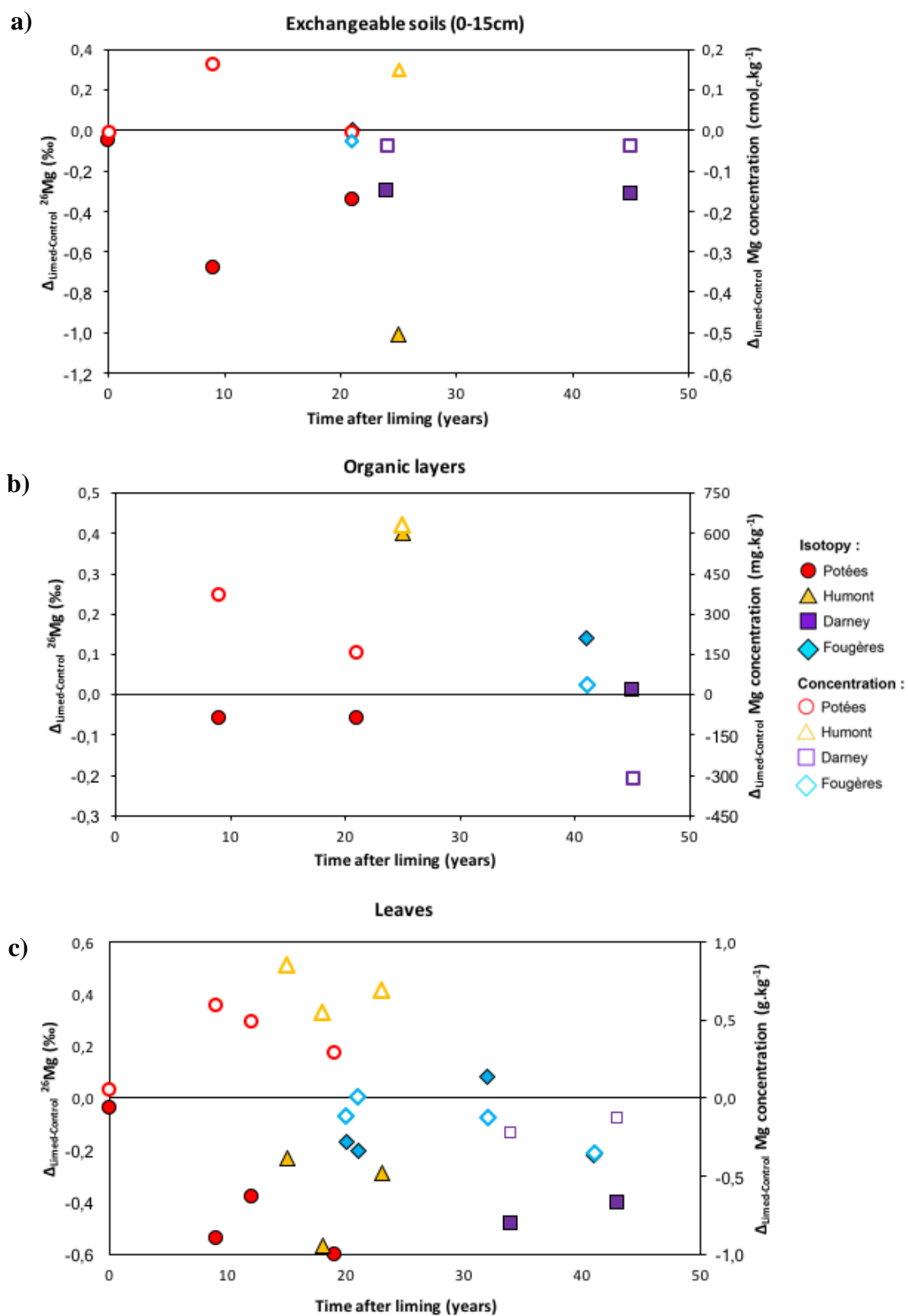
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831 **Table captions**

832 **Table 1.** K, Ca and Mg concentrations and $\delta^{26}\text{Mg}$ of the leaves (a), organic layer (b) and [0-
833 15cm] exchangeable soil pools (c) for each studied site. Slope, N and n values refer to the
834 $\delta^{26}\text{Mg}$ analysis: N = number of sample sequences, n = number of validated replicates, n.a =
835 not analysed, * = calculated values (see supplementary Fig. S1).

836

837 **Table 2.** Isotopic composition difference ($\Delta^{26}\text{Mg}$) between the different ecosystem
838 compartments (leaves, organic layer and [0-15cm] exchangeable soils) for the control and
839 limed plots of the studied sites.

840

841 **Table 1 (a)**

Sites	Treatments	Leaves										
		Years	K (ppm)	Ca (ppm)	Mg (ppm)	$\delta^{26}\text{Mg}_{\text{DSM3}}$ (‰)	$\delta^{25}\text{Mg}_{\text{DSM3}}$ (‰)	Slope	N	n		
Potées	Control	1995	6273	4378	728	-0.88	0.13	-0.45	0.06	0.509	4	11
		1996	6482	4490	852	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2004	8001	3523	966	-0.40	0.11	-0.20	0.05	0.508	4	9
		2007	6705	4832	820	-0.92	0.13	-0.46	0.06	0.504	2	5
	Pre-treatment	2014	5437	4646	1083	-0.57	0.16	-0.29	0.09	0.511	4	9
		1995	6218	4910	782	-0.92	0.04	-0.46	0.02	0.499	3	7
		1996	5978	5996	1033	-0.90	0.07	-0.45	0.04	0.501	3	8
		2004	7943	5541	1569	-0.94	0.12	-0.48	0.06	0.511	6	15
Ca/MgCO ₃	2007	7894	7453	1310	-1.30	0.14	-0.67	0.05	0.517	3	7	
	2014	7398	7986	1380	-1.17	0.04	-0.60	0.02	0.515	3	7	
	2006	8165	1834	695	-1.14	0.04	-0.59	0.05	0.514	3	9	
	2009	10366	1622	612	-1.01	0.06	-0.52	0.03	0.519	2	5	
Humont	Control	2014	8337	2115	792	-1.07	0.06	-0.55	0.03	0.514	2	4
		2006	5728	5973	1551	-1.37	0.05	-0.72	0.04	0.518	2	6
	Ca/MgCO ₃	2009	9084	6170	1162	-1.58	0.05	-0.82	0.03	0.517	3	6
		2014	7316	5371	1489	-1.36	0.06	-0.70	0.02	0.513	2	5
Fougères	Control	1993	8627	3315	1147	-1.15	0.09	-0.59	0.04	0.514	4	11
		1994	8537	3826	1018	-1.36	0.07	-0.69	0.04	0.513	3	9
		2005	8796	2404	683	-1.42	0.10	-0.73	0.05	0.513	2	6
		2014	7025	4930	1322	-0.93	0.18	-0.47	0.08	0.509	3	8
	CaCO ₃	1993	7519	6980	1032	-1.32	0.03	-0.68	0.02	0.517	3	9
		1994	8342	7348	1030	-1.56	0.09	-0.80	0.05	0.515	3	9
		2005	6348	6271	557	-1.34	0.07	-0.68	0.04	0.512	2	5
		2014	7254	7023	973	-1.15	0.05	-0.60	0.02	0.515	2	4
Darney	Control	1994	8850	3500	600	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2005	9789	4589	661	-1.12	0.10	-0.57	0.06	0.512	3	8
	CaCO ₃	2014	11044	3074	410	-1.42	0.09	-0.72	0.05	0.504	2	6
		1994	8900	7500	300	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
Coat-an-Hay	Control	2005	10375	7067	433	-1.60	0.09	-0.80	0.05	0.508	2	6
		2014	10295	4867	277	-1.82	0.06	-0.93	0.04	0.511	2	6
	Ca/MgCO ₃	1995	7243	4100	914	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2014	6837	4610	1278	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>

842

843 **Table 1 (b)**

Sites	Treatments	Organic layer										
		Years	K (ppm)	Ca (ppm)	Mg (ppm)	$\delta^{26}\text{Mg}_{\text{DSM3}}$ (‰)	$\delta^{25}\text{Mg}_{\text{DSM3}}$ (‰)	Slope	N	n		
Potées	Control	1995	4035	2861	1649	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2004	2216	5088	732	-0.48	0.08	-0.25	0.03	0.517	2	5
		2007	2646	6112	910	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2016	1689	7630	1031	-0.43	0.01	-0.22	0.01	0.509	1	2
	Pre-treatment	1995	3179	3737	1483	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2004	2022	7815	1103	-0.54	0.09	-0.28	0.05	0.518	5	10
		2007	2880	9027	1264	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2016	1920	10023	1188	-0.66	0.11	-0.34	0.05	0.510	2	5
Humont	Control	2016	2104	3802	1065	-0.40	0.02	-0.20	0.01	0.502	3	8
	Ca/MgCO ₃	2016	3264	4512	1699	0.00	0.13	0.01	0.06	0.893	3	9
Fougères	Control	1994	1242	4291	1058	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2014	2441	5325	1422	-0.66	0.11	-0.34	0.05	0.510	2	5
	CaCO ₃	1994	1368	8211	933	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
Darney	Control	2014	1970	8973	1462	-0.52	0.13	-0.28	0.06	0.538	2	4
	CaCO ₃	2016	3356	4613	944	-0.35	0.12	-0.19	0.04	0.521	3	6
Coat-an-Hay	Control	2016	1790	4049	636	-0.34	0.14	-0.17	0.08	0.517	4	8
	Ca/MgCO ₃	2014	1583	7632	2042	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>

844

Sites	Treatments	Years	Exchangeable soils (0-15cm)						Slope	N	n	
			K (ppm)	Ca (ppm)	Mg (ppm)	$\delta^{26}\text{Mg}_{\text{DSM3}}$ (‰)	$\delta^{25}\text{Mg}_{\text{DSM3}}$ (‰)					
Potées	Control	1995	100	57	20	-0,93	0,05	-0,48	0,03	0,516	2+2*	6+5*
		2004	66	30	10	-0,52	0,01	-0,27	0,01	0,519	1+1*	2+3*
		2007	71	88	21	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
	Pre-treatment	2016	53	39	12	-0,32	0,03	-0,16	0,02	0,513	2	5
		1995	99	59	19	-0,98	0,03	-0,50	0,02	0,510	1+2*	2+3*
		2004	64	210	30	-1,20	0,09	-0,62	0,05	0,517	3+2*	4+6*
		2007	109	557	59	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
Ca/MgCO ₃	2016	43	62	11	-0,66	0,00	-0,34	0,00	0,515	3	3	
	2007	118	29	23	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	
Humont	Control	2016	87	18	16	-0,34	0,11	-0,18	0,07	0,540	4	10
		2007	119	426	73	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
	Ca/MgCO ₃	2016	83	130	34	-1,35	0,04	-0,68	0,03	0,510	2	5
Fougères	Control	1994	103	75	37	-0,91	0,10	-0,47	0,06	0,511	3	8
		2014	72	75	34	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
	CaCO ₃	1994	88	278	33	-0,91	0,01	-0,48	0,00	0,528	2	4
		2014	34	24	13	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
Darney	Control	1995	103	32	17	-0,50	0,05	-0,26	0,02	0,520	2+2*	8+3*
		2016	78	20	12	-0,53	0,04	-0,27	0,04	0,506	3	6
	CaCO ₃	1995	104	92	12	-0,80	0,07	-0,41	0,04	0,512	1+1*	3+4*
		2016	65	26	8	-0,84	0,02	-0,44	0,01	0,521	2	4
Coat-an-Hay	Control	1995	92	76	40	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2014	59	50	17	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
	Ca/MgCO ₃	1995	81	61	28	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>
		2014	54	153	35	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>

Compartments	Sites	Treatment	Years	$\Delta_{\text{compartments}}^{26}\text{Mg}$ (‰)	$\Delta_{\text{compartments}}^{25}\text{Mg}$ (‰)
Leaves - Organic layer ($\Delta^{26}\text{Mg}_{\text{Fol-Org}}$)	Potées	Control	2004	0.08	0.05
			2014	-0.14	-0.07
		Ca/MgCO ₃	2004	-0.40	-0.20
			2014	-0.68	-0.34
	Humont	Control	2014	-0.67	-0.35
		Ca/MgCO ₃	2014	-1.36	-0.71
	Fougères	Control	2014	-0.27	-0.13
		CaCO ₃	2014	-0.63	-0.32
	Darney	Control	2014	-1.07	-0.53
		CaCO ₃	2014	-1.48	-0.76
Leaves - Exchangeable soil (0-15cm) ($\Delta^{26}\text{Mg}_{\text{Fol-Exch}}$)	Potées	Control	1995	0.05	0.03
			2004	0.12	0.07
			2016	-0.25	-0.13
		Pre-treatment	1995	0.06	0.04
		Ca/MgCO ₃	2004	0.26	0.14
	Humont		2016	-0.51	-0.26
		Control	2016	-0.73	-0.37
		Ca/MgCO ₃	2016	-0.01	-0.02
		Fougères	Control	1994	-0.45
	CaCO ₃		1994	-0.65	-0.32
Darney	Control	2016	-0.89	-0.45	
	CaCO ₃	2016	-0.98	-0.49	
Organic layer - Exchangeable soil (0-15cm) ($\Delta^{26}\text{Mg}_{\text{Org-Exch}}$)	Potées	Control	2004	0.04	0.02
			2016	-0.11	-0.06
		Ca/MgCO ₃	2004	0.66	0.34
			2016	0.17	0.08
	Humont	Control	2016	-0.06	-0.02
		Ca/MgCO ₃	2016	1.35	0.69
	Darney	Control	2016	0.18	0.08
		CaCO ₃	2016	0.50	0.27

850 **Electronic Supplementary material**

851 **Fig. S1.** Description of the methodology to calculate the isotopic composition ($\delta^{26}\text{Mg}$) of the
852 0-15cm soil layer from the measured values of the 0-5 and 5-15 cm soil layers at Potées (1995
853 and 2004) and Darney (1995) sites.

854 **Fig. S2.** Schematic description of the Mg pools and fluxes taken into account in the model.
855 The fluxes with no isotopic fractionation are in black while isotopic fractionating fluxes are in
856 red (alpha values represent the respective isotopic fractionation factor).

857 **Table S3.** Description of the different hypotheses tested and results obtained with the
858 numerical modelling approach.

859 **Fig. S4.** Relationship between Mg concentration in leaves and $\Delta_{\text{compartments}}^{26}\text{Mg}$. Studied
860 control plots are in blue, studied limed plots in red and literature data in black*.

861 * (*Bedel 2015; Bolou-Bi et al. 2012*).

862

- 1) Calculation of the ^{26}Mg and ^{24}Mg ratio for DSM3 (mean on all sessions) :

$$\frac{^{26}\text{Mg}}{^{24}\text{Mg}}(\text{DSM3}) = 0.1592887 \quad n = 788$$

- 2) Calculation of the ^{26}Mg and ^{24}Mg ratio for each soil layer sample :

$$\frac{^{26}\text{Mg}}{^{24}\text{Mg}}(\text{sample}) = \left(\left(\frac{\delta^{26}\text{Mg}(\text{sample})}{1000} \right) + 1 \right) \times \frac{^{26}\text{Mg}}{^{24}\text{Mg}}(\text{DSM3})$$

- 3) Calculation of the amount of ^{26}Mg for each soil layer sample :

$$\%^{26}\text{Mg}(\text{sample}) = \frac{\frac{^{26}\text{Mg}}{^{24}\text{Mg}}(\text{sample})}{\left(\left(1 + \frac{^{25}\text{Mg}}{^{24}\text{Mg}}(\text{DSM3}) \right) + \frac{^{26}\text{Mg}}{^{24}\text{Mg}}(\text{sample}) \right)}$$

with $\frac{^{25}\text{Mg}}{^{24}\text{Mg}}(\text{DSM3}) = 0.126742$

- 4) Calculation of the amount of ^{26}Mg for the 0-15 cm soil layer :

$$\%^{26}\text{Mg}(0 - 15\text{cm}) = \frac{\left((\%^{26}\text{Mg}_{(0-5\text{cm})} \times e_{(0-5\text{cm})} \times [\text{Mg}]_{(0-5\text{cm})}) + (\%^{26}\text{Mg}_{(5-15\text{cm})} \times e_{(5-15\text{cm})} \times [\text{Mg}]_{(5-15\text{cm})}) \right)}{\left((e_{(0-5\text{cm})} \times [\text{Mg}]_{(0-5\text{cm})}) + (e_{(5-15\text{cm})} \times [\text{Mg}]_{(5-15\text{cm})}) \right)}$$

with e = thickness of soil layer

$[\text{Mg}]$ = Mg concentration of soil layer

- 5) Calculation of the ^{26}Mg and ^{24}Mg ratio for the 0-15 cm soil layer :

$$\frac{^{26}\text{Mg}}{^{24}\text{Mg}}(0 - 15\text{cm}) = \frac{\left(\%^{26}\text{Mg}_{(0-15\text{cm})} \times \left(1 + \frac{^{25}\text{Mg}}{^{24}\text{Mg}}(\text{DSM3}) \right) \right)}{\left(1 - \%^{26}\text{Mg}_{(0-15\text{cm})} \right)}$$

- 6) Calculation of the $\delta^{26}\text{Mg}$ for the 0-15 cm soil layer :

$$\delta^{26}\text{Mg}_{(0-15\text{cm})} = \left(\left(\frac{\frac{^{26}\text{Mg}}{^{24}\text{Mg}}(0 - 15\text{cm})}{\frac{^{26}\text{Mg}}{^{24}\text{Mg}}(\text{DSM3})} \right) - 1 \right) \times 1000$$

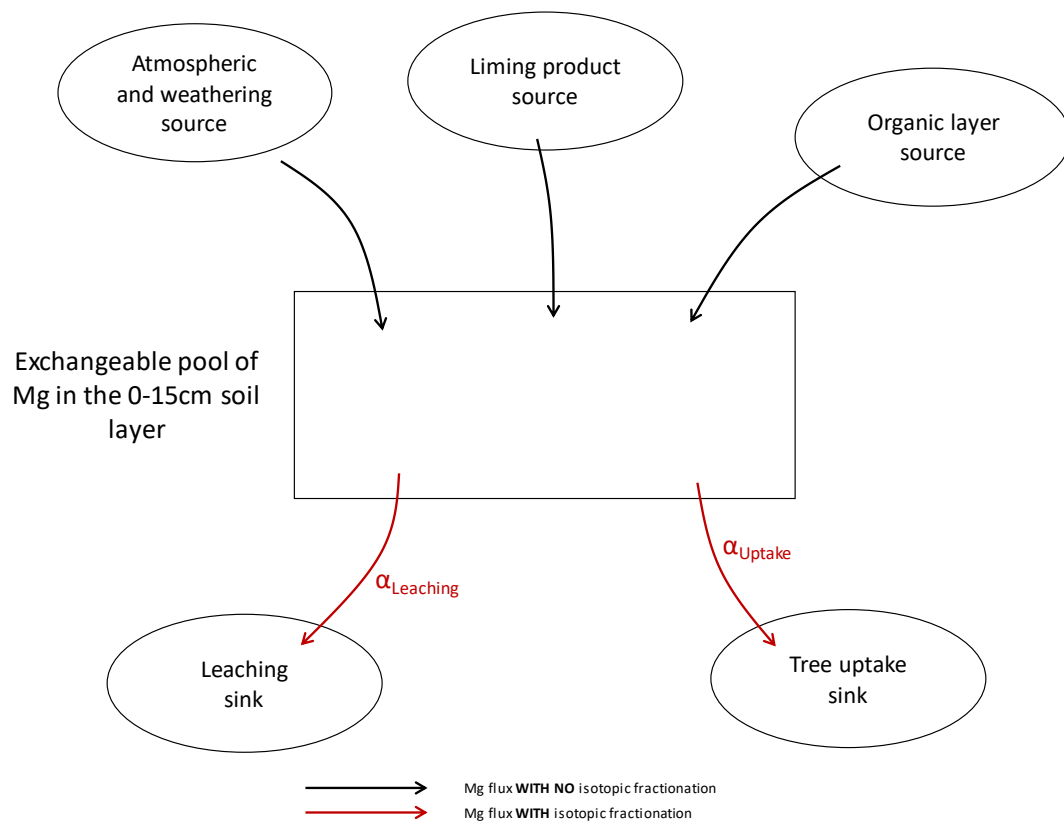


Table S3.

Site	Hypothesis	Description/details	Range of input variables tested	Range of simulated parameters	Hypothesis rejected?	Explanation
Darney	The isotopic fractionation during Mg uptake by roots (preferential uptake of ^{26}Mg) explains the differences between limed and control plots	The Mg uptake flux in the limed plot increased after the liming compared to the control (the exchangeable Mg pool was lower in the limed plot) and caused a $\delta^{26}\text{Mg}$ signature shift in the exchangeable Mg pool.	Mg Uptake: $[0.1 : 2.0] \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	$\alpha_{\text{Uptake}}: [+5 : +10] \text{ ‰}$	YES	The simulated fractionation factors are greater than the Mg isotopic variation at the terrestrial scale and no such fractionation during root adsorption has been reported.
	The isotopic mixing of native exchangeable Mg in the [0-15cm] layer with an input source of Mg with a very low $\delta^{26}\text{Mg}$ signature explains differences between limed and control plots	The input source of Mg originates from the mineralization of the organic layer after liming. Liming operation have been shown to alter mineralization processes and may have induced the preferential release of ^{24}Mg from the organic layer.	Mg organic layer: $5 \text{ kg}\cdot\text{ha}^{-1}$ $\alpha_{\text{Uptake}}: +0.5 \text{ ‰}$ and $+1.0 \text{ ‰}$ Mg Uptake: $0.5 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ $1.0 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	$\delta^{26}\text{Mg}$ organic layer source: [-2.4 : -1.5]‰ [-4.1 : -2.9]‰	YES	The simulated $\delta^{26}\text{Mg}$ of the organic layer source is much lower than the lowest $\delta^{26}\text{Mg}$ reported for organic layers ($\approx -0.5 \text{ ‰}$)
	The input source of Mg originates from the Ca carbonate (reported $\delta^{26}\text{Mg}$ values for Ca carbonate rocks range from -5‰ to -4‰)		$\alpha_{\text{Uptake}}: +0.5 \text{ ‰}$ and $+1.0 \text{ ‰}$ Mg Uptake: $0.5 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ $1.0 \text{ kg}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	Mg input source: [1.9 : 3.0] $\text{kg}\cdot\text{ha}^{-1}$ [3.3 : 5.2] $\text{kg}\cdot\text{ha}^{-1}$	NO	To obtain the simulated contributions of a Mg carbonate source the applied carbonate ($2500 \text{ kg}\cdot\text{ha}^{-1}$) would need to contain between 0.13% and 0.34% MgO which is possible
Les Potées	The large fraction of liming product, which was not found in soil ($111 \text{ kg}\cdot\text{ha}^{-1}$), was immobilized in the biomass.		Not modelled		YES	The input-output budgets only show a slight increase of the Mg stock in the biomass in the limed plots ($+5 \text{ kg}\cdot\text{ha}^{-1}$) over the 1995-2004 period.
	The large fraction of liming product, which was not found in soil, was immobilized in the organic layer.		Not modelled		YES	The input-output budgets only shows a slight increase of the Mg stock in the organic layer ($+3 \text{ kg}\cdot\text{ha}^{-1}$) over the 1995-2004 period.
	A large fraction of the Mg released from the liming product was leached below 15cm depth	A fraction of the liming product was very rapidly (within the 1st year after liming) leached below 15 cm depth and did not isotopically mix with the 0-15cm soil layer	$\alpha_{\text{Uptake}}: +0.5 \text{ ‰}$ and $+1.0 \text{ ‰}$ Lime Mg leaching: $[0 - 120] \text{ kg}\cdot\text{ha}^{-1}$	Preferential leaching of dolomitic Mg: [85% : 89%]	NO	The simulated proportion of dolomitic Mg leached below 15cm in only one year agrees with the calculated recovery of dolomitic Mg in 2004 ($14 \text{ kg}\cdot\text{ha}^{-1}$)
		The Mg leaching was associated to an isotopic fractionation	$\alpha_{\text{Uptake}}: +0.5 \text{ ‰}$ and $+1.0 \text{ ‰}$ Leaching scenarios: - All dolomitic Mg leaching occurs in the first year after application - The leaching of dolomitic Mg is evenly spread over the 10 year period	$\alpha_{\text{Leaching}}:$ - 0.6 ‰ and -0.4 ‰ - 0.35 ‰ and -0.25 ‰	NO	Though isotopic fractionation during Mg leaching is not well characterized in the literature, the simulated isotopic fractionation factors seem realistic

Fig. S4.

