

# What deep-soil profiles can teach us on deep-time pH dynamics after land use change?

Boris Brasseur, Fabien Spicher, Jonathan Roger Michel Henri Lenoir, Emilie Gallet-Moron, Jérome Buridant, Hélène Horen

### ► To cite this version:

Boris Brasseur, Fabien Spicher, Jonathan Roger Michel Henri Lenoir, Emilie Gallet-Moron, Jérome Buridant, et al.. What deep-soil profiles can teach us on deep-time pH dynamics after land use change?. Land Degradation and Development, 2018, 29 (9), pp.2951-2961. 10.1002/ldr.3065 . hal-02356959

## HAL Id: hal-02356959 https://hal.science/hal-02356959

Submitted on 18 Nov 2019

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

#### 1 What deep-soil profiles can teach us on deep-time pH dynamics after land use change?

2 B. Brasseur, F. Spicher, J. Lenoir, E. Gallet-Moron, J. Buridant, H. Horen

#### 3 Short title: Deep-soil profiles teach us deep-time pH dynamics

- Affiliation of all authors: UMR7058 CRNS EDYSAN Ecologie et Dynamiques des Systèmes
   Anthropisés Université de Picardie Jules Verne Amiens FRANCE
- 6 **Principal author address:** Université de Picardie Jules Verne 1, rue des Louvels 80 000
- 7 Amiens Cedex FRANCE
- 8 Email: boris.brasseur@u-picardie.fr
- 9 Tel: +33 (0)3 22 82 77 57
- 10 Fax: +33 (0)3 22 82 78 29

11 How to cite this article: Brasseur B, Spicher F, Lenoir J, Gallet-Moron E, Buridant J, Horen H. What deep-soil profiles can

14

#### 15 Abstract

Soil profiles keep records of the legacies of historical land uses on soil physicochemical 16 properties with deep-soil (3m depth) profiles providing information on centuries-old 17 dynamics. By combining geohistorical archives on past land-uses and management practices 18 together with soil pH data from 19 plots scattered across five study areas in North-France, 19 we analyzed the effect of two contrasting historical land-use change trajectories 20 21 (afforestation of former arable land vs. cultivation after deforestation) on the acidification 22 and alkalinisation dynamics along 3m depth loessic luvisols profiles. An analysis of covariance (ANCOVA) and a generalized additive model (GAM) were used to test the 23 24 interaction effect between depth and the two studied chronosequences levels on soil pH. Results show two contrasting alkalinization dynamics for formerly acidic forest soils 25 26 converted to arable land depending on past management practices. One century of liming 27 practices based on hydrated lime is enough for a complete deprotonation whereas alkalinization dynamic was much slower (2-4 centuries are necessary to neutralize the 28 relictual acidity below 2m) under liming based on chalk-blocks supply. Per contra, 29

<sup>12</sup> teach us on deep-time pH dynamics after land use change?. Land Degrad Dev. 2018;1–11.

<sup>13</sup> https://doi.org/10.1002/ldr.3065

afforestation of former arable land cultivated and limed during at least 2-4 centuries keep an alkaline "memory" effect over more than a millennium. The acidification process is progressive through time from the surface to deeper soil horizons. This suggests that soil pH profiles can be used to estimate time since afforestation of former arable land. Linking deepsoil pH profiles with chronosequences is a powerful tool to understand the potential impact of future land-use change trajectories on soil physicochemical properties.

#### 36 Keywords: Afforestation, deforestation, geohistory, liming, loess, luvisol

- 37
- 38
- 39

#### 40 Introduction

41 It is now widely acknowledged that today's soils encapsulate the legacy of not only natural processes but also actual and past human activities (Dambrine et al., 2007; Kluiving et al., 42 2015). Since the Neolithic, farmers and foresters throughout their activities are influencing 43 the pedogenesis and evolution of anthropogenic landscapes (Van Vliet-Lanoe et al., 1992; 44 Langhor, 2001; Emadodin et al., 2009). Imprints of past land uses (e.g., croplands, 45 grasslands, woodlands) are still visible in current biodiversity patterns (Closset-Kopp & 46 47 Decocq, 2015; Dambrine et al., 2007), molecular biomarkers (Lavrieux et al., 2012) and the chemical composition of soils (Koerner et al., 1999; Cornu et al., 2007; Von Oheimb et al., 48 2008). Among soil physico-chemical parameters, potential hydrogen (pH) is a very good 49 indicator of land-use changes (LUCs) from forest to arable land and vice versa (Blake et al., 50 1999; Bossuyt et al., 1999; Verheyen et al., 1999). Soil pH controls pedogenetic processes 51 52 such as acidolysis and acido-complexolysis when soil horizons become acid, it also controls the bioavailability of nutrients for plant growth and influences bacterial, fungal, vegetal and 53 54 animal biodiversity (see for example, Chauchan, 2014; Fichtner et al., 2014), hence soil pH is 55 a key parameter in pedogenetic dynamics through time.

The dynamic of soil alkalinization results from a negative proton balance which can 56 be triggered by calcic mineral amendment (e.g., limestones, lime, marls) in agricultural 57 regions under temperate humid climate. These agronomic practices, first described during 58 the 1<sup>st</sup> century BC by Varron (Ed. 1864: book I - chapter VIII) and much more detailed later 59 on by Pliny the Elder (Pliny the Elder, 1877: book XVII - chapter 4), are very old in north-60 western Europe and varies in nature (Goulding, 2016), amount and frequency of inputs 61 (Tables S1-S2). Liming is required as cereals growth is seriously limited on acid soils of 62 formerly forested lands which were deforested to extend land cultivation. Thus in North 63 64 France, liming was a very systematic practice in the first years following deforestation sometimes accompanied by grazing (Crescentiis, 1548). First writings on soil liming following 65 deforestation are from the 12<sup>th</sup> century (Fossier, 1987) (i.e., the "sartier" contracts). Other 66 67 farming practices than liming such as fertilization, product removal and crop rotations may also affect the proton  $(H^{\dagger})$  and hydroxyl  $(OH^{-})$  budgets (Merry, 2009; Schroder *et al.*, 2011) 68 and thus the dynamic of soil alkalinization. Not only the dynamic of soil alkalinization due to 69 70 agronomic practices is a well-known phenomenon, but also the short-term (few decades) dynamic of soil acidification after afforestation of former cultivated areas has been heavily 71 investigated (de Schrijver et al., 2012). For instance, the impact of LUC on soil pH is known to 72 73 persist over 30-80 years of forest growth on formerly agricultural lands irrespective to the pH measurement method, i.e., pH<sub>water</sub>, pH<sub>KCl</sub> or pH<sub>CaCl2</sub> (Ritter *et al.*, 2003; Falkengren-Grerup 74 75 et al., 2006). Some authors have even demonstrated that the dynamic of soil acidification after afforestation depends on the selected tree species (Hagen-Thorn et al., 2004; de 76 Schrijver et al., 2012), albeit land-use history seems to influence soil properties more than 77 the selected tree species (Ritter et al., 2003). 78

Over longer timescale, the imprint of former agricultural practices on soil properties, including soil pH, is also documented. However, the few studies documenting it focused on the soil surface solely (from 5 and up to 20cm) (Dambrine *et al.*, 2007; Closset-Kopp &

Decocq, 2015), which precludes a wide part of the available soil archives. Wigley et al. (2013) 82 nicely emphasized this issue by asking: "What do ecologists miss by not digging deep 83 enough?". Drouet et al. (2015) provided some answers by showing that deep nutrients could 84 significantly contribute to tree growth. Only a handful of studies, focusing on loessic luvisols 85 exclusively, examined the long-term acidification or alkalinisation dynamics through deep 86 87 soil profiles. The experimental site of Rothamsted farm (UK) is a good example where soil 88 profiles were sampled up to 90cm depth during a period of 110 years after the afforestation of cultivated chromic luvisols. The pH<sub>CaCl2</sub> profiles of this experimental site revealed a net 89 90 surface acidification from 6.2 to 3.8, whereas the deepest horizons of the investigated soil 91 profiles showed a drop of 0.25 pH unit only (Blake et al., 1999). In parallel, Bossuyt et al (1999) used a chronosequence (i.e., a synchronic approach) of afforested loessic luvisols in 92 93 Belgium and found a very marked decline in pH<sub>KCI</sub> during the first 30 years after afforestation followed by a slower decline in the next 20 years after which  $pH_{KCI}$  was similar to the level 94 observed in an ancient forest. Regarding the long term dynamic of soil alkalinisation, Pécrot 95 96 (1956) focused on cultivated soils after deforestation in Belgium and concluded that the 97 alkalinization process of the Ap horizon was performed after 50 years, with the Bt horizon being neutral to alkaline after 100 years. After 700 years of agricultural practices, the entire 98 99 soil profile was neutral to alkaline. More recently, Colinet (2003) measured, within the same region, a slower alkalinisation dynamic with the Bt horizon remaining acid ( $pH_{water} = 5.3$ ) 100 101 after 100 years of cultivation while a neutral (around 6.5) soil profile was observed up to 102 165cm depth after 250 years of cultivation. Hence, deep-time (>100 years) acidification and 103 alkalinization dynamics following LUCs still remain unknown and yet could provide important 104 insights for ecosystem functioning such as productivity.

Here we use two chronosequences spanning several centuries (up to 1500 years) of land use legacies and supported by robust and ascertainable geohistorical data (Tables S1-S2 & S3) to describe deep-time alkalinization and acidification dynamics. A total of 102 soil 108 pH<sub>water</sub> profiles on luvisols in North France were examined to answer the following 109 questions: (i) How long are the impacts of past LUCs (e.g., cultivation after deforestation and 110 afforestation after cultivation) still noticeable on soil pH? (ii) What are the vertical and 111 temporal dynamics of soil alkalinization and acidification? (iii) Is soil pH a good proxy to date 112 past LUCs?

113

#### 114 Materials and methods

115

116 Study area

117 The study area is located on loessic luvisols in North France (Thiérache: 49°48'34"N, 4°01'11"E). We sampled 19 plots within a geographical area (Figure 1) comprising 5 ancient 118 119 forests sites: (1) the Haye of Aubenton (AU; 4 plots); (2) the Val-Saint-Pierre forest (VSP; 4 120 plots); (3) the Andigny forest (AN; 4 plots); (4) the Plomion wood (PL; 6 plots); and (5) the Metz-en-Couture/Havrincourt wood (MC; 1 plot). The local plateau is about 100-150m above 121 122 sea level and the climate is humid temperate: mean annual air temperature is 8.75°C; 123 precipitations are 750-800mm/year; and ETP Turc is 570mm (SILVAE portal, AgroParisTech-INRA & LERFoB). Forest stands are dominated by common beech (Fagus sylvatica), oak 124 125 (Quercus robur) and hornbeam (Carpinus betulus), all older than 80 years excepting the recently afforested plots. Every plots are located on the plateau (i.e., very gentle slopes <5%) 126 to avoid soil erosion and spatial heterogeneity in acid neutralizing capacity due to 127 topography (Brahy et al., 2000). 128

129 The studied soils are developed on periglacial thick loamy deposits of the Western-130 European loess belt, which is a relatively uniform parent material in North France (Antoine 131 *et al.*, 2003; Jamagne, 1972). This loess belt is lying on Turonian-Santonian clayey marls and 132 white chalks.

134 Geohistorical reconstruction of land uses across the study area

Various kinds of historical data sources were used to reconstruct past land uses, such as: the
regional archaeological database (*Patriarche*); local archives; the Napoleonic cadastral maps
and the associated register of status of the land (1820); historical Etat-Major maps (1830);
aerial photographs from 1930 to 2013 (IGN); and oral interviews.

We prepared two chronosequences spanning several centuries of LUCs each. The first one (cf. the afforestation chronosequence) focused on afforestation after cultivation and the second one (cf. the deforestation chronosequence) focused on cultivation after deforestation. For each of the 19 studied plots, LUCs from arable land to forest and *vice versa* were controlled by robust and ascertainable geohistorical data (see Figure 2 for example), all summarized in the supporting information (Table S3).

The afforestation chronosequence contains six different levels along an ordinal scale 145 ranging from "recently afforested" to "afforested since a very long time": a medieval strip 146 field afforested 15 years ago (C $\rightarrow$ F<sub>1999</sub>); a field afforested 80 years ago (C $\rightarrow$ F<sub>1935</sub>); two 147 148 medieval strip fields afforested 150 years ago ( $C \rightarrow F_{1865ck} \& C \rightarrow F_{1865}$ ; with and without Ck 149 loessic lower horizon, respectively); one medieval strip field afforested 500 to 1000 years ago (C $\rightarrow$ F<sub>1000-1500</sub>); and four ancient forests (C $\rightarrow$ F<sub>500AU+VSP+PL+AN</sub>). These four ancient forests 150 were already forested during the 12<sup>th</sup> or 13<sup>th</sup> century (Table S3) but could have been 151 cultivated during bronze-iron ages and/or antiquity as many west European ancient forests 152 153 (Dubois, 1990). Gallo-Roman sites such as farms (AU, PL), roads (AU, AN, VSP), buildings (AU, PL, VSP), and cremation graves (AU, AN, PL, VSP) occur within these ancient forests. No 154 Medieval or Modern sites were found suggesting that these areas could have been 155 cultivated during antiquity and afforested as early as the troubled period of the end of the 156 Western Roman Empire during the 5<sup>th</sup> century AD. The absence of the upper part of 157 158 albeluvic tonguing on these luvisols (ISSS Working Group RB, 1998) as well as the brownish 159 colour of the E horizon (7.5YR4/4) due to an increased burrowing activity from earthworms

and moles enhanced by liming and manuring (Bossuyt *et al.*, 1999; Langhor, 2001) support this hypothesis. Hence, a putative age of 1500 years BP has been attributed to  $C \rightarrow F_{500}$  plots.

The deforestation chronosequence contains six different levels along an ordinal scale 162 ranging from "recently deforested" to "deforestation since a very long time": several 163 164 formerly ancient forests deforested about 5 ( $F \rightarrow C_{2009}$ ); 30 ( $F \rightarrow C_{1985}$ ); 140 ( $F \rightarrow C_{1875}$ ); 150  $(F \rightarrow C_{1865})$ ; and 170  $(F \rightarrow P_{1845} \rightarrow C_{1920} \& F \rightarrow C_{1845}$ ; with  $F \rightarrow P_{1845} \rightarrow C_{1920}$  being a pasture without 165 liming practices, but manured, during the first 85 years following deforestation and then 166 being limed & cultivated during the next 85 years until now) years ago. In addition to those 167 168 six levels, four cultivated ancient fields ( $C \rightarrow C_{AU+VSP+PL+AN}$ ) whose great age was proven by medieval strip shapes (Sivery, 1990) were used as control plots. All plots from the 169 deforestation chronosequence excepting the control plots were located in sites previously 170 171 described as medieval forests ( $F \rightarrow C_{2009}$ ,  $F \rightarrow C_{1985}$ ,  $F \rightarrow C_{1875}$ ,  $F \rightarrow C_{1865}$ ,  $F \rightarrow P_{1845} \rightarrow C_{1920}$  and  $F \rightarrow C_{1845}$ ). 172

Following Pliny the Elder (Pline l'ancien, 1877), liming practices were already used by 173 Gallic, Germanic and Britannic farmers for cereals culture during Antiquity. In North France, 174 the use of white chalk (sometimes miscalled marl) is confirmed from the beginning of the 175 high medieval period and marling contracts were recorded for lands after deforestation 176 (Derville, 1999; Fossier, 1987). Note that liming practices (amount and periodicity of inputs) 177 were likely variable over time (Table S1). Actual practices from the farmers who currently 178 179 own the arable fields where our studied plots are located are usually around 15T/Ha each 10-20 years (Table S2). Bolton (1977) observed, on the sandy-clay loam fields of the 180 Rothamsted farm, that CaCO<sub>3</sub> mean loss rate were ranging from 368kg/Ha/yr (for 5T/Ha 181 182 added initially) to 823 kg/Ha/yr (for 20T/Ha added initially) over 12 years. Thus adding 5T/Ha each 12 years was sufficient to maintain soil pH values over long time periods. Liming in 183 greater amount once and/or more frequently over time can help to increase soil pH values 184 185 from acidic to neutral conditions for farming. Thus, local liming practices reported across our

study sites and from the lower medieval to the modern period were sufficient to maintainand even raise soil pH values (Tables S1 and S2).

188

#### 189 Soil sampling and measurement methods

Between 2013 and 2016, a total of 6 soil profiles were sampled for each of the 19 studied 190 plots (C $\rightarrow$ C<sub>AN</sub>, C $\rightarrow$ C<sub>VSP</sub>, C $\rightarrow$ C<sub>AU</sub>, C $\rightarrow$ C<sub>BP</sub>, C $\rightarrow$ F<sub>500AN</sub>, C $\rightarrow$ F<sub>500VSP</sub>, C $\rightarrow$ F<sub>500AU</sub>, C $\rightarrow$ F<sub>500BP</sub>, F $\rightarrow$ C<sub>2009</sub>, 191  $\mathsf{F} \rightarrow \mathsf{C}_{1985}, \, \mathsf{F} \rightarrow \mathsf{C}_{1875}, \, \mathsf{F} \rightarrow \mathsf{C}_{1865}, \, \mathsf{F} \rightarrow \mathsf{P}_{1845} \rightarrow \mathsf{C}_{1920}, \, \mathsf{F} \rightarrow \mathsf{C}_{1845}, \, \mathsf{C} \rightarrow \mathsf{F}_{1999}, \, \mathsf{C} \rightarrow \mathsf{F}_{1935}, \, \mathsf{C} \rightarrow \mathsf{F}_{1865ck}, \, \mathsf{C} \rightarrow \mathsf{F}_{1865}, \, \mathsf{C} \rightarrow \mathsf{F}_{1865ck}, \, \mathsf{C} \rightarrow \mathsf{C}_{1865ck}, \, \mathsf{C} \rightarrow$ 192  $C \rightarrow F_{1000-1500}$ ) excepting  $C \rightarrow C_{AN}$ ,  $C \rightarrow C_{VSP}$ ,  $C \rightarrow C_{AU}$  and  $F \rightarrow C_{1875}$ , where only 3 soil profiles were 193 194 sampled, thus totalizing 102 soil profiles across the entire study area (Table S4). By using an 195 Edelman hand auger, we retrieved a deep-soil profile (3m depth) for each of these 102 soil profiles. The soil profiles were distant from each other of about 10 to 100m within a given 196 plot. The soil augering descriptions were carried out following the FAO standard guidelines. 197 Soil horizon depths and textures were noted and Munsell colors were determined on moist 198 samples. For each of the 102 sampled soil profiles, we characterized the humus (Brethes et 199 200 al., 1995) and noted the humus index (Ponge & Chevalier, 2006) as well as the colors of the 201 soil horizons (Table S5). None of the 102 profiles had any archaeological remains, topographic anomalies, natural (windthrow) or anthropogenic disturbances, anomaly of 202 humus thickness (Bossuyt et al., 1999) or a tree trunk that was closer than 2m (Beniamino et 203 204 *al.,* 1991).

Soil samples for soil pH analyses were collected at the following depths: 5; 25; 50; 75; 100; 125; 150; 175; 200; 250; and 300cm. For each of these 11 depths, 500g of soil material was extracted and sealed in labeled plastic bags and transported to the lab where the soil was air-dried and sieved to 2mm. Soil pH was measured with two glass electrodes in distilled water (ISO 10390:2004) on two sub-samples, before being averaged. Among the 1122 (n = 102x11) samples of 500g of soil material, soil pH measurements could be obtained and used for subsequent analyses for a total of 1074 samples (Table S4). For subsequent analyses, soil pH measurements for  $C \rightarrow C_{AN}$ ,  $C \rightarrow C_{VSP}$ ,  $C \rightarrow C_{AU}$  and  $C \rightarrow C_{BP}$  as well as for  $C \rightarrow F_{500AN}$ ,  $C \rightarrow F_{500VSP}$ ,  $C \rightarrow F_{500AU}$  and  $C \rightarrow F_{500BP}$  were pooled in the  $C \rightarrow C$  and  $C \rightarrow F_{500}$  levels, respectively.

214

#### 215 Data analyses

To detect an imprint of past LUCs on soil pH profiles and represent the inter- and intra-plot 216 variability in the data (n = 1044), we ran several statistical models testing the impact of 217 218 different LUC trajectories (the main explanatory variable) on soil pH (the response variable) 219 while controlling for the potential confounding effect of soil depth (a covariate) and its 220 potential interaction with past LUCs. Soil pH and depth were treated as continuous variables 221 while the variable representing the different LUC trajectories of the afforestation and 222 deforestation chronosequences tested here was treated as a factor variable with 12 levels 223  $(C \rightarrow C, C \rightarrow F_{500}, F \rightarrow C_{2009}, F \rightarrow C_{1985}, F \rightarrow C_{1865}, F \rightarrow P_{1845} \rightarrow C_{1920}, F \rightarrow C_{1845}, C \rightarrow F_{1999}, C \rightarrow F_{1935}, F \rightarrow C_{1865}, F \rightarrow C_{1845}, F \rightarrow C_{1845}, F \rightarrow C_{1845}, F \rightarrow C_{1845}, F \rightarrow C_{1985}, F \rightarrow C_{198$  $C \rightarrow F_{1865ck}$ ,  $C \rightarrow F_{1865}$ ,  $C \rightarrow F_{1000-1500}$ ). Note here that the  $F \rightarrow C_{1875}$  level of LUC trajectory was not 224 225 considered in any of the statistical models due to low sample size (n = 3 replicates per soil depth for  $F \rightarrow C_{1875}$ ). Using ordinary least square (OLS) regression and generalized additive 226 model (GAM), we tested four candidate models: a null model (or intercept model) without 227 any explanatory variable as a control or baseline (m0: pH~1); a model that tests the effect of 228 soil depth on soil pH (m1: pH~Depth); a model that tests the effect of the different LUC 229 230 trajectories while controlling for soil depth (m2: pH~Depth+LUC); and a model that tests the interaction effect between soil depth and LUC trajectories (m3: pH~Depth\*LUC). The OLS 231 regression approach assumes a linear relationship between soil pH and depth while the 232 233 GAM approach can allow non-linear relationships. As an intermediate type of model, we also ran OLS regression models with a second-order polynomial relationship between soil pH and 234 depth. In total, we ran 3x4 different models (Table 2) and we used the Akaike information 235 236 criterion (AIC) to select the best model (Burnham & Anderson, 2002), which is the one with

the lowest AIC value (AIC<sub>min</sub>). We also computed the relative support for each model, using the differences in AIC values,  $\Delta AIC_i = AIC_i - AIC_{min}$ , where AIC<sub>i</sub> is the observed AIC value of the focal model *i* and AIC<sub>min</sub> is the lowest AIC value from the best model in the set of candidate models (Burnham & Anderson, 2002). Based on the outputs from the best model with the lowest AIC value, we subsequently ran Wilcoxon signed-rank tests to compare the impact of different LUC trajectories on soil pH at a given depth.

All statistical analyses were performed within the R software environment (R Development Core Team, 2016), using the "bbmle" and "mgcv" packages. All our models' predictions were associated to a 95% confidence interval (CI = 1.96xSEM; where SEM means standard error of the mean) around the mean soil pH profile. Residuals from the best model were checked for normality and homoscedasticity (Figure S1).

248

- 249 Results
- 250
- 251 Soils' description

The 102 soils exhibited a contrasted development of eluvial (E) and illuvial (Bt) horizons. The 252 253 diffuse transition horizon (BC) and C decarbonated loess material mostly constitute the 254 lower half part of the soil profile whereas calcareous loess material was completely absent except for  $C \rightarrow F_{1865ck}$ , which exhibited calcareous loess at 220-250cm depth. For afforested 255 256 soils, the humus index score increased from 1 (i.e., eumull) to 3.6 (i.e., oligomull-dysmull) 257 with increasing afforestation age (Table S5) and the A/E transition appeared between 12cm  $(C \rightarrow F_{500})$  and 25cm  $(C \rightarrow F_{1999})$  depth. For tilled cultivated soils, the Ap/E transition showed a 258 259 lesser variation with a range between 26cm ( $F \rightarrow C_{1845}$ ) and 35cm ( $F \rightarrow C_{2009}$ ).

260 The E horizons were bioturbated with some soils sampled in the Aubenton site 261  $(C \rightarrow F_{500AU}, C \rightarrow F_{1865}$  and  $C \rightarrow F_{1865ck}$ ) having gleyic colour patterns evidenced by high values

and low chroma in the Munsell soil color chart. The E/Bt transition across the study area was 262 found between 33cm ( $F \rightarrow C_{1865}$ ) and 60cm (AF) depth. Importantly, the upper parts of the 263 transitional albeluvic tonguing between the E and Bt horizons were systematically truncated 264 and leveled by bioturbation, even on  $C \rightarrow F_{500}$  luvisols which suggests cultures as past land 265 uses. The Bt horizon of most plots showed light grey polygonal crack networks typical of a 266 fragipan. Hence, the soils are classified (IUSS Working Group WRB, 2006) in luvisols but still 267 bear relict patterns of albeluvisols. All soils' horizons textures are very homogeneous, from 268 silt loam in the surface to silty clay loam for the deep part (Figure S2), albeit the sand 269 270 content of  $C \rightarrow F_{1999}$  raised from 100cm towards the deepest part.

271

#### 272 Soil pH profiles of ancient forested ( $C \rightarrow F_{500}$ ) and ancient cultivated ( $C \rightarrow C$ ) soils

We found a strong variation in soil pH values with depth in both the afforestation and deforestation chronosequences (Table 1 and Figure S3). The topsoil of ancient forests (AF) showed the lowest pH values (4.61) while the highest pH values (8.18) were found at 3m depth in the carbonated plot ( $C \rightarrow F_{1865ck}$ ). Soil pH in  $C \rightarrow F_{500}$  increased with depth and reached 5.17, 5.54 and 5.71 at 1m, 2m and 3m depth, respectively, whereas the ancient cultivated soil reference ( $C \rightarrow C$ ) exhibited a homogeneous soil pH profile around neutral values independently of soil depth.

Independently of the modelling approach used, the best candidate models of soil pH included an interaction effect between soil depth and LUC trajectories (Table 2) with soil pH profiles being specific to each of the 12 studied LUC trajectories (Figure 3). The lowest AIC value was obtained with a generalized additive model (GAM) (cf. model "m3g" in Table 2), which explained 91% of the total variation in soil pH.

285

286 Chronosequences

For the youngest afforested plot ( $C \rightarrow F_{1999}$ ), neutro-alkaline pH values were still found below 287 75cm depth but decreased significantly to 6.66 and 6.03 at 50cm and 5cm depth, 288 respectively. Soil pH values in the  $C \rightarrow F_{1999}$  LUC trajectory significantly differed from the 289 290 ancient cultivated soils (C $\rightarrow$ C) reference above 100cm depth and up to the surface (Figure 3). Eighty years after afforestation ( $C \rightarrow F_{1935}$ ), soil pH values became significantly more acidic 291 292 than the C $\rightarrow$ C reference all along the soil profile (Figure 3), with pH dropping to 6.10 at 5cm 293 depth. About 150 years after afforestation, the mean of topsoil pH was 5.2 and 4.8 for the  $C \rightarrow F_{1865}$  and  $C \rightarrow F_{1865ck}$  LUC trajectories, respectively, with topsoil pH being significantly 294 295 greater than the C $\rightarrow$ F<sub>500</sub> reference only for the C $\rightarrow$ F<sub>1865</sub> LUC trajectory (Wilcoxon signed-rank 296 test: W = 131, p = 0.0012). At 175cm depth, soil pH increased up to 7.07 for the C $\rightarrow$ F<sub>1865ck</sub> LUC trajectory, thus being not significantly different from the C $\rightarrow$ C reference. The oldest 297 298 afforested plot (C $\rightarrow$ F<sub>1000-1500</sub>) exhibited greater mean pH values than the C $\rightarrow$ F<sub>500</sub> reference 299 for all depths, with the difference being significant for the topsoil (Wilcoxon signed-rank test: W = 105, p = 0.0435) but not significant in the deeper part of the soil profile: from 300 301 250cm depth (Wilcoxon signed-rank test: W = 91, p = 0.0551).

302 Shortly after deforestation and cultivation ( $F \rightarrow C_{2009}$ ,  $F \rightarrow C_{1985}$ ), soil pH values were near neutral in the topsoil but remained very acid (<5) under ploughed horizons (i.e., 50 to 303 300cm depth) (Table 1 and Figure 3). On  $F \rightarrow C_{1875}$  and  $F \rightarrow C_{1865}$  plots, about 150 years after 304 deforestation and cultivation, soil pH values were neutro-alkaline (≥7) from topsoil down to 305 300cm depth. For the  $F \rightarrow P_{1845} \rightarrow C_{1920}$  LUC trajectory, the neutro-alkaline pH values observed 306 between the surface and 100cm depth sharply declined to acid values (5.51 at 250cm depth) 307 more or less similar to the C $\rightarrow$ F<sub>500</sub> reference. The F $\rightarrow$ C<sub>1845</sub> LUC trajectory showed weakly acid 308 to neutral pH values from topsoil (pH = 6.09 at 5cm depth) to the deep C horizon (pH = 6.33 309 at 250cm depth) and through the Bt horizon (pH = 7.02 at 75cm depth). 310

311

312 Discussion

314 Alkalinization and acidification dynamics

Based on deep-soil profiles, the strong interaction we found between the different land-use changes (LUCs) we tested here and soil depth explained 91% of the observed variation in soil pH across the study area. This clearly suggests different acidification and alkalinization dynamics with soil depth depending on the two LUC trajectories. Regarding the potential drivers for the unexplained variation see Appendix S1.

On the deforestation and culture chronosequence, time since deforestation clearly 320 321 matters to explain the alkalinization kinetic in the soil profile. For recent deforestation 322 events that happened 5-30 years ago (i.e.,  $F \rightarrow C_{2009}$  and  $F \rightarrow C_{1985}$ ), our results suggest that the alkalinization process is confined within the ploughed horizon (Ap) with a sharp decline in pH 323 values towards initial acidic conditions, reflecting former land use as forest. Following the 324 Rothamsted's model ( $\Delta pH = 0.109L-0.690$ ; L is lime added as T/Ha CaCO<sub>3</sub>) (Goulding *et al.*, 325 1986; Goulding, 2016), we found that for a topsoil pH value of 4.6 (equivalent to ancient 326 327 forest) if we add 30T/Ha of chalk, pH should rise up by 2.6 points in the next years. This 328 situation is likely similar to the one observed for  $F \rightarrow C_{2009}$ , showing an increase in pH values reaching 7.1 in the Ap horizon (Table 1). 329

330 Coherently, for older deforestation events that happened 140 to 170 years ago (e.g.,  $F \rightarrow C_{1865}$ ), the alkalinization process migrates (more or less rapidly) towards the deepest soil 331 layers. Interestingly and for LUC trajectories being more or less equivalent in terms of time 332 since deforestation and culture (i.e.,  $F \rightarrow C_{1875}$ ,  $F \rightarrow C_{1865}$ ,  $F \rightarrow P_{1845} \rightarrow C_{1920}$  and  $F \rightarrow C_{1845}$ ), the 333 migration rate of this alkalinization front likely depends on the former management 334 practices used by farmers. For cultivated soils limed with defecation lime ( $F \rightarrow C_{1875}$  and 335  $F \rightarrow C_{1865}$ ), the alkalinisation dynamic operated all along the soil profile and soil pH values are 336 337 now neutro-alkaline from the surface until 3m depth. However, for cultivated soils limed 338 with chalk  $(F \rightarrow P_{1845} \rightarrow C_{1920}$  and  $F \rightarrow C_{1845})$ , the alkalinisation process was much less

pronounced and pH values gently decline towards acid values in the deep soil layers. For the 339  $F \rightarrow P_{1845} \rightarrow C_{1920}$  LUC trajectory, for which successive liming practices were restricted to the 340 last 85 years, the migration rate of the alkalinization front is more or less intermediate with 341 342 pH values declining towards initial acidic conditions at 3m depth. These results suggest that 343 the high solubility of Ca(OH)<sub>2</sub> in defecation lime (Table S6) has triggered faster and deeper soil alkalinization rates through winter infiltrations of waters (Girard et al., 2005) highly 344 concentrated in OH<sup>-</sup> ions (strong base). In contrast, the low solubility of coarse chalk 345 fragments limits the basicity of infiltration waters, which bring few  $CO_3^{2-}$  (weak base). 346

347 As expected for the afforestation chronosequence, we found a progressive acidification process over time, from topsoil to deeper soil horizons, with the oldest 348 afforestation events ( $C \rightarrow F_{1000-1500}$ ) showing the largest decrease in soil pH values relative to 349 350 the ancient culture (AC) reference. The magnitude and rate of this acidification dynamic, relatively to the C $\rightarrow$ C reference, is more important towards the soil surface where the main 351 sources of acidity are concentrated: litter decomposition (Markewitz et al., 1998; Reich et 352 al., 2005); root exudation and respiration (Hinsinger et al., 2003); and atmospheric 353 precipitations (Van Breemen et al., 1983). Below pH = 5.5, soil Al solubilization trigger acidity 354 buffering, which decrease acidification kinetic on the acid forest plots. Hence, depending on 355 the time since afforestation, the imprint of the former cultivation on soil pH can still be 356 visible in the deepest part of the soil profile. For instance, 150 years after afforestation 357 358  $(C \rightarrow F_{1865})$ , soil pH values below 200cm depth are more or less similar to the C $\rightarrow$ C reference. Noteworthy, our results suggest that the alkalinisation process after deforestation and 359 culture is faster (F $\rightarrow$ C<sub>1845</sub>; chalk) to much faster (F $\rightarrow$ C<sub>1865</sub>; defecation lime) than the 360 361 acidification process after afforestation.

362

363 Towards a use of pH for dating of LUCs?

Our results of alkalinisation dynamics after deforestation and culture are very consistent with former findings from the East Hesbaye area in Belgium (Pécrot, 1956), where the alkalinisation front propagated until 40cm (B<sub>21</sub> horizon) and 125cm (B<sub>3</sub>) depths after 50 and 100 years of cultivation, respectively. However, pH data from the West Hesbaye area (Colinet, 2003) suggested slower alkalinisation dynamics with slightly acid pH values in the Ap horizon after 100 years of cultivation and such acidity preserved below 150cm depth even after 200 years of cultivation.

Owing to the diversity of liming practices (chalk vs. hydrated lime, frequency, amount 371 372 by Ha, granulometry) there is a high variability of alkalinisation dynamics after deforestation 373 and culture. If farmers use fine-grained hydrated lime, this could trigger a very rapid and deep alkalinisation process whereas if they use coarse chalk blocks this process will be much 374 375 slower. Because of this variability, it appears difficult to date a deforestation event just from 376 soil pH profiles except if precise historical liming practices are available to infer the most likely alkalinisation dynamics. Therefore, historical archives should always be preferred over 377 378 inferences from soil pH profiles only to date a deforestation event.

For the afforestation chronosequence, we found an age dependent evolution of the acidification dynamic from topsoil to the deeper horizons. In the upper soil layers, pH values rapidly decreased with time and became hardly discriminable from the  $C \rightarrow F_{500}$  reference 150 years after afforestation. In contrast, beyond one meter depth, the more recently afforested soils (i.e., 15 years ago) were indistinguishable from the cultivated soils. Similarly and beyond 100cm depth, soil pH values from the  $C \rightarrow F_{1935}$  &  $C \rightarrow F_{1865}$  LUC trajectories are indistinguishable.

Considering soil pH at 50cm depth, it rapidly decreased from 7.33 to 5.61 during the first 150 years and then slowly decreased to 4.74 after 1500 years of forest land use (Figure 4). Our 5 plots from the last 150 years were precisely dated and we compared our results with the pH<sub>CaCl2</sub> values, converted in pH<sub>water</sub> (Libohova *et al.*, 2014), from the Rothamsted

experimental station (Figure 4, Blake et al., 1999) where the climatic conditions are close to 390 our study area. Both datasets show high consistency and suggest that soil pH at 50cm depth 391 is negatively correlated with afforestation age during the first 150 years (see Figure 4). The 392 393 slower decay of soil pH values at 50cm depth after 150 years likely reflects the increase in the soil buffering capacity caused by the alteration of aluminosilicates when pH drops below 394 395 5.5. The basal part of the studied soil profiles (2m to 3m depth) best preserves the initial soil properties before LUCs, like the  $C \rightarrow F_{1865ck}$  LUC trajectory which still show high pH values at 396 3m depth due to loess initial calcium carbonate. As 2-4 centuries of chalk liming practices 397 398 are necessary to complete deep soil deprotonation and re-saturate exchange complex with Ca<sup>2+</sup> (Colinet, 2003), only the afforestation plots on ancient fields display deep neutro-399 alkaline profiles when acidification began. 400

401

#### 402 Conclusion

Our findings suggest that one century of liming practices with fine-grained hydrated lime is 403 404 enough to reach a complete deprotonation of formerly acid forest soils. A much slower alkalinization is observed when coarse chalk (or limestone) blocks are used during 170 years 405 406 of cultivation and liming, with pH values remaining slightly acid (6-6.5) below 2m depth. We assume that two to four centuries of chalk liming practices over loessic luvisols should be 407 408 necessary to completely neutralize the relictual acidity below 2m depth. Hence, the occurrence of imprints of former forest occupation in the deepest soil layers will mainly 409 depend upon the wide diversity of liming practices through time rather than time elapsed 410 since deforestation and culture. Per contra, imprints of former agricultural lands limed 411 during at least two to four centuries will still be visible in the deepest soil layers more than 412 413 millennia after afforestation. The acidification dynamic is more regular and progressive from 414 the surface to the deepest soil horizons. Finally, we argue that soil pH at 50cm depth could

be used as a proxy to track afforestation events that happened few decades to a millennium ago. Below 2m depth, heterogeneities between soils initial alkalinity levels may still influence the level of acidity of afforested sites. Human exploitation of the landscape deeply transform the pedosphere chemistry through history and studying deep soil pH profiles may help, together with complementary sources, to reconstruct the history of past land uses.

420

#### 421 Acknowledgements

422 We would like to thanks colleagues and students who helped us in the field or during 423 laboratory analysis: V. Avedikian; A. Fauconnier; C. Gaella Kotokpo; M. Leborgne; S. Lobjois;

424 R. Saguez; L. Storr; M. Tessié; and M. Vergne.

425

#### 426 Supporting information

- 427 Figure S1: Histogram of residuals on the best AIC score model (m3g)
- 428 Figure S2: Soil textures (Pipette method NF X31-107) ratio values for Clay-Silt-Sand
- 429 Figure S3: Mean pH of the plots studied in the two chronosequences: a-afforestation on
- 430 culture and b-deforestation and culture
- 431 Table S1: Local liming practices from the lower medieval period to the modern period and
- 432 antique liming practices over Gaul and Britain territories.
- 433 Table S2: Liming practices on the recent field plots ( $F \rightarrow C_{year}$ ) following oral communications
- and archives.
- Table S3: Geohistory and geohistorical arguments of the studied plots over the a)
- 436 afforestation and b) deforestation chronosequences
- 437 Table S4: Number of soil samples at each soil depth and for each of the 13 levels
- 438 characterizing the factor variable of LUC trajectories

- 439 Table S5: Summary statistics of the average soil profile, in terms of humus type, horizon
- 440 colors and depth limits, for each of the 13 levels
- 441 Table S6: Alkaline reagent used on our F→C plots and their principal characteristics
- 442 Appendix S1: Comments on intra- and inter-plot variability
- 443
- 444 References
- Antoine P, Catt J, Lautridou J-P, Sommé J. 2003. The loess and coversands of northern France and
  southern England. *Journal of Quaternary Science* 18: 309–318. DOI: 10.1002/jqs.750
- Beniamino F, Ponge JF, Arpin P. 1991. Soil acidification under the crown of oak trees I. Spatial
  distribution. *Forest Ecology and Management* 40: 221–232. DOI: 10.1016/03781127(91)90041-S
- Blake L, Goulding KWT, Mott CJB, Johnston AE. 1999. Changes in soil chemistry accompanying
  acidification over more than 100 years under woodland and grass at Rothamsted
  Experimental Station, UK. *European Journal of Soil Science* 50: 401–412. DOI:
  doi/10.1046/j.1365-2389.1999.00253.x
- Bolton J. 1977. Changes in soil pH and exchangeable calcium in two liming experiments on
  contrasting soils over 12 years. *The Journal of Agricultural Science* 89: 81–86. DOI:
  10.1017/S0021859600027210
- Bossuyt B, Deckers J, Hermy M. 1999. A field methodology for assessing man-made disturbance in
  forest soils developed in loess. *Soil Use and Management* 15: 14–20. DOI: 10.1111/j.14752743.1999.tb00056.x
- 460 Brahy V, Deckers J, Delvaux B. 2000. Estimation of soil weathering stage and acid neutralizing
   461 capacity in a toposequence Luvisol–Cambisol on loess under deciduous forest in Belgium.
   462 *European Journal of Soil Science* 51: 1–13. DOI: 10.1046/j.1365-2389.2000.00285.x
- Brethes A, Brun J, Jabiol B, Ponge J, Toutain F. 1995. Classification of forest humus forms: a French
   proposal. *Annales des Sciences Forestières* 52: 535–546. DOI: 10.1051/forest:19950602
- 465 Burnham K, Anderson D. 2002. *Model Selection and Multimodel Inference*. New-York: Springer-Verlag
- 466 Caton l'ancien, Varron, Columelle, Palladius. 1864. *Les agronomes latins : Caton, Varron, Columelle,* 467 *Palladius*. Paris: Firmin Didot frères fils et cie
- Chauchan RP. 2014. Role of Earthworms in Soil Fertility and Factors Affecting Their Population
   Dynamics: A review. *International Journal of Research* 1: 642–649
- 470 Closset-Kopp D, Decocq G. 2015. Remnant Artificial Habitats as Biodiversity Islets into Forest Oceans.
   471 *Ecosystems* 18: 507–519. DOI: 10.1007/s10021-015-9843-3
- 472 Colinet G. 2003. Eléments traces métalliques dans les sols. Contribution à la connaissance des
  473 déterminants de leur distribution spatiale en région limoneuse Belge. Gembloux: Faculté
  474 Universitaire des Sciences Agronomiques de Gembloux

475 Cornu S, Montagne D, Maguin F, Lay CL, Chevallier P, Cousin I. 2007. Influence of human impacts on 476 Albeluvisol analysed by X-ray microfluorescence: Relative evolution of the transforming front 477 at the tongue scale. Science of The Total Environment 377: 244–254. DOI: 478 10.1016/j.scitotenv.2007.01.083 479 Crescentiis P "de." 1548. De omnibus agriculturae partibus et de plantarum animaliumque natura et 480 utilitate lib. XII: non minus Philosophiae & medicinae, quàm oeconomiae, agricolationis, 481 pastionum[que] studiosis utiles. Basileae: Henrichus Petri 482 Dambrine E, Dupouey J-L, Laüt L, Humbert L, Thinon M, Beaufils T, Richard H. 2007. Present forest 483 biodiversity patterns in France related to former Roman agriculture. *Ecology* 88: 1430–1439. 484 DOI: 10.1890/05-1314 485 de Schrijver A, de Frenne P, Staelens J, Verstraeten G, Muys B, Vesterdal L, Wuyts K, van Nevel L, 486 Schelfhout S, de Neve S, Verheyen K. 2012. Tree species traits cause divergence in soil 487 acidification during four decades of postagricultural forest development. Global Change 488 Biology 18: 1127–1140. DOI: 10.1111/j.1365-2486.2011.02572.x 489 Derville A. 1999. L'agriculture du nord au Moyen Age: Artois, Cambresis, Flandre Wallonne. 490 Villeneuve-d'Ascg: Presses Univ. Septentrion 491 Drouet T, Herbauts J, Demaiffe D. 2015. Influence of deep soil horizons on Ca nutrition of forest 492 stands along a loessic soil sequence. Plant and Soil 394: 373–389. DOI: 10.1007/s11104-015-493 2540-3 494 Dubois JJ. 1990. Espaces et milieux forestiers dans le nord de la France: étude de biogéographie 495 historique. Lille: Université de Lille II 496 Emadodin I, Reiss S, Mitusov AV, Bork HR. 2009. Interdisciplinary and multidisciplinary approaches to 497 the study of long-term soil degradation: A case study from Schleswig-Holstein, Germany. 498 Land Degradation & Development 20: 551-561. DOI: 10.1002/ldr.941 499 Falkengren-Grerup U, Brink D-J ten, Brunet J. 2006. Land use effects on soil N, P, C and pH persist 500 over 40–80 years of forest growth on agricultural soils. *Forest Ecology and Management* 225: 501 74-81. DOI: 10.1016/j.foreco.2005.12.027 502 Fichtner A, von Oheimb G, Härdtle W, Wilken C, Gutknecht JLM. 2014. Effects of anthropogenic 503 disturbances on soil microbial communities in oak forests persist for more than 100 years. 504 Soil Biology and Biochemistry 70: 79–87. DOI: 10.1016/j.soilbio.2013.12.015 505 Fossier R. 1987. La terre et les hommes en Picardie jusqu'à la fin du XIIIe siècle. Amiens: CRDP 506 Girard MC, Walter C, Rémy JC, Berthelin J, Morel JL. 2005. Sols et environnement. Paris: Dunod 507 Goulding KWT. 2016. Soil acidification and the importance of liming agricultural soils with particular 508 reference to the United Kingdom. Soil Use and Management 32: 390-399. DOI: 509 10.1111/sum.12270 510 Goulding KWT, Poulton PR, Thomas VH, Williams RJB. 1986. Atmospheric deposition at Rothamsted, 511 Saxmundham, and Woburn experimental stations, England, 1969–1984. Water, Air, and Soil 512 Pollution 29: 27-49. DOI: 10.1007/BF00149327 Hagen-Thorn A, Callesen I, Armolaitis K, Nihlgård B. 2004. The impact of six European tree species on 513 514 the chemistry of mineral topsoil in forest plantations on former agricultural land. Forest 515 Ecology and Management 195: 373-384. DOI: 10.1016/j.foreco.2004.02.036

- Hinsinger P, Plassard C, Tang C, Jaillard B. 2003. Origins of root-mediated pH changes in the
  rhizosphere and their responses to environmental constraints: A review. *Plant and Soil* 248:
  43–59. DOI: 10.1023/A:1022371130939
- 519 ISSS Working Group RB. 1998. World Reference Base for Soil Resources. Leuven: Acco
- 520 IUSS Working Group WRB. 2006. World Reference Base for Soil Resources 2006. Rome: FAO
- Jamagne M. 1972. Contribution à l'étude pédogénétique des formations loessiques du Nord de la
   France. Gembloux: thèse doctorale en sciences agronomiques, faculté des Sciences
   agronomiques de Gembloux
- Kluiving SJ, Bekkema ME, Roymans NGAM. 2015. Mass migration through soil exhaustion:
   Transformation of habitation patterns in the southern Netherlands (1000 BC–500 AD).
   *Catena* 132: 139–150. DOI: 10.1016/j.catena.2014.12.015
- Koerner W, Dambrine E, Dupouey JL, Benoît M. 1999. δ15N of forest soil and understorey vegetation
   reflect the former agricultural land use. *Oecologia* 121: 421–425. DOI:
   10.1007/s004420050947
- Langhor R. 2001. L'anthropisation du paysage pédologique agricole de la Belgique depuis le
   Néolithique ancien Apport de l'archéopédologie. *Etudes et Gestion des sols* 8: 103–118
- Lavrieux M, Bréheret J-G, Disnar J-R, Jacob J, Le Milbeau C, Zocatelli R. 2012. Preservation of an
   ancient grassland biomarker signature in a forest soil from the French Massif Central.
   Organic Geochemistry 51: 1–10. DOI: 10.1016/j.orggeochem.2012.07.003
- Libohova Z, Wills S, Odgers NP, Ferguson R, Nesser R, Thompson JA, West LT, Hempel JW. 2014.
  Converting pH 1:1 H2O and 1:2CaCl2 to 1:5 H2O to contribute to a harmonized global soil
  database. *Geoderma* 213: 544–550. DOI: 10.1016/j.geoderma.2013.08.019
- Markewitz D, Richter DD, Allen HL, Urrego JB. 1998. Three Decades of Observed Soil Acidification in
   the Calhoun Experimental Forest: Has Acid Rain Made a Difference? *Soil Science Society of America Journal* 62: 1428. DOI: 10.2136/sssaj1998.03615995006200050040x
- 541 Merry R. 2009. Acidity and Alkalinity of Soils. *Environmental and Ecological Chemistry*. Aleksandar
   542 Sabljic: Paris, 115–131
- 543 Pécrot A. 1956. Etude détaillée des sols de la Hesbaye occidentale application à quelques problèmes
  544 d'actualité. Gent: Comité pour l'établissement de la Carte des Sols et de la Végétation de la
  545 Belgique
- 546 Pline l'ancien. 1877. *Histoire naturelle de Pline*. E. Littré Paris: Firmin-Didot et cie
- Ponge J-F, Chevalier R. 2006. Humus Index as an indicator of forest stand and soil properties. *Forest Ecology and Management* 233: 165–175. DOI: 10.1016/j.foreco.2006.06.022
- R Development Core Team. 2016. *R: A language and environment for statistical computing*. Vienna: R
   Foundation for Statistical Computing
- Reich PB, Oleksyn J, Modrzynski J, Mrozinski P, Hobbie SE, Eissenstat DM, Chorover J, Chadwick OA,
  Hale CM, Tjoelker MG. 2005. Linking litter calcium, earthworms and soil properties: a
  common garden test with 14 tree species. *Ecology Letters* 8: 811–818. DOI: 10.1111/j.14610248.2005.00779.x

555 Ritter E, Vesterdal L, Gundersen P. 2003. Changes in soil properties after afforestation of former 556 intensively managed soils with oak and Norway spruce. Plant and Soil 249: 319–330. DOI: 557 10.1023/A:1022808410732 558 Schroder JL, Zhang H, Girma K, Raun WR, Penn CJ, Payton ME. 2011. Soil Acidification from Long-559 Term Use of Nitrogen Fertilizers on Winter Wheat. Soil Science Society of America Journal 75: 560 957-964. DOI: 10.2136/sssaj2010.0187 561 Sivery G. 1990. Terroirs & communautés rurales dans l'Europe Occidentale au Moyen Âge. Villeneuve-562 d'Ascq: Presses Univ. Septentrion Van Breemen N, Mulder J, Driscoll CT. 1983. Acidification and alkalinization of soils. Plant and Soil 75: 563 564 283-308. DOI: 10.1007/BF02369968 565 Van Vliet-Lanoe B, Fagnart J-P, Langhor R, Munaut A. 1992. Importance de la succession des phases 566 écologiques anciennes et actuelles dans la différenciation des sols lessivés de la couverture 567 lœssique d'Europe occidentale: argumentation stratigraphique et archéologique. Science du Sol 30: 75-93 568 569 Verheyen K, Bossuyt B, Hermy M, Tack G. 1999. The land use history (1278–1990) of a mixed 570 hardwood forest in western Belgium and its relationship with chemical soil characteristics. Journal of Biogeography 26: 1115–1128. DOI: 10.1046/j.1365-2699.1999.00340.x 571 Von Oheimb G, Härdtle W, Naumann PS, Westphal C, Assmann T, Meyer H. 2008. Long-term effects 572 573 of historical heathland farming on soil properties of forest ecosystems. Forest Ecology and 574 Management 255: 1984–1993. DOI: 10.1016/j.foreco.2007.12.021 575 Wigley BJ, Coetsee C, Hartshorn AS, Bond WJ. 2013. What do ecologists miss by not digging deep 576 enough? Insights and methodological guidelines for assessing soil fertility status in ecological 577 studies. Acta Oecologica 51: 17-27. DOI: 10.1016/j.actao.2013.05.007 578

**Table 1.** Summary statistics (mean and standard deviation) of soil pH values at different soil depths and for each of the 13 modalities characterizing the factor variable of land-use change (LUC) trajectories (cf. the afforestation and deforestation chronosequences). The meaning of each of the levels of the factor variable of LUC trajectories is fully described in the main text of the manuscript (see the Materials and Methods section). The total number of soil profiles (i.e., sample size) per LUC trajectory and per soil depth is provided in Table S4.

Depth (cm)	Depth C→F <sub>500</sub> (cm)		C→F <sub>1000-1500</sub>		$C \rightarrow F_{1865ck}$		$C \rightarrow F_{1865}$		C→F <sub>1935</sub>		C→F <sub>1999</sub>		F→C <sub>2009</sub>		F→C <sub>1985</sub>		F→C <sub>1875</sub>		F→C <sub>1865</sub>		$F \rightarrow P_{1845} \rightarrow C_{1920}$		F→C <sub>1845</sub>		C→C	
()	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ	Mean	σ
5	4,61	0,34	4,90	0,27	4,78	0,41	5,16	0,26	6,10	0,21	6,03	0,33	7,08	0,61	6,19	0,39	7,88	0,24	7,67	0,18	7,63	0,23	6,09	0,47	7,19	0,37
25	4,63	0,28	4,90	0,05	4,84	0,36	5,18	0,31	5,99	0,32	6,22	0,25	7,05	0,83	6,18	0,43	7,77	0,28	7,76	0,25	7,65	0,21	6,72	0,10	7,28	0,43
50	4,74	0,21	5,05	0,18	5,50	0,32	5,61	0,34	6,11	0,12	6,66	0,27	4,68	0,35	4,81	0,18	7,91	0,15	7,70	0,25	7,70	0,15	6,98	0,08	7,33	0,30
75	4,98	0,21	5,34	0,21	5,92	0,10	5,80	0,29	6,31	0,17	6,90	0,18	4,65	0,18	4,77	0,26	7,86	0,13	7,69	0,26	7,68	0,15	7,02	0,15	7,34	0,22
100	5,17	0,18	5,60	0,22	6,20	0,27	6,02	0,27	6,43	0,21	7,13	0,15	4,87	0,29	5,07	0,15	7,84	0,11	7,66	0,27	7,39	0,42	6,73	0,34	7,34	0,25
125	5,34	0,19	5,65	0,18	6,56	0,17	6,44	0,20	6,53	0,28	7,22	0,16	5,12	0,24	4,99	0,21	7,79	0,15	7,49	0,35	6,79	0,69	6,60	0,48	7,24	0,25
150	5,46	0,26	5,75	0,17	6,84	0,21	6,58	0,29	6,40	0,33	7,29	0,17	5,27	0,26	4,95	0,20	7,72	0,09	7,36	0,40	6,09	0,68	6,61	0,50	7,22	0,28
175	5,49	0,28	5,83	0,17	7,07	0,23	6,71	0,30	6,33	0,23	7,24	0,17	NA	NA	4,91	0,19	NA	NA	7,36	0,38	NA	NA	6,57	0,48	7,16	0,36
200	5,54	0,28	5,78	0,20	7,34	0,46	6,78	0,28	6,41	0,23	7,21	0,17	5,33	0,27	4,91	0,18	7,71	0,18	7,27	0,31	5,78	0,72	6,54	0,45	7,15	0,30
250	5,66	0,47	5,91	0,30	7,91	0,43	6,80	0,30	6,44	0,26	7,24	0,20	5,18	0,14	4,98	0,61	7,71	0,22	7,30	0,28	5,51	0,36	6,33	0,62	6,95	0,29
300	5,71	0,52	5,91	0,37	8,18	0,05	6,87	0,25	6,74	0,26	7,18	0,24	5,30	0,12	5,07	0,51	7,64	0,15	6,98	0,61	5,44	0,23	6,25	0,60	6,99	0,24

**Table 2.** The 12 tested models clustered in 3 different categories of four candidate models (m0, m1, m2, m3): ordinary least square (OLS) regression models with a linear relationship between soil pH and soil depth; OLS regression models with a second-order polynomial (mXp) relationship between soil pH and soil depth; and generalized additive models (GAMs: mXg) allowing more complex nonlinear relationships. Difference in Akaike Information Criterion (AIC) values between the best model and the focal model ( $\Delta$ AIC), Akaike weights (w), adjusted R<sup>2</sup> values (R<sup>2</sup><sub>adj</sub>) as well as degrees of

7 freedom (Df) are provided for comparison purposes.

- 8
- 9

Modelling approach	Id	Depth	LUC	Depth x LUC	AIC	ΔΑΙϹ	Wi	R <sup>2</sup> adj	Df
generalized additive model	m3g	Х	Х	Х	519	0	0	0,91	968
two order polynomial model	m3p	Х	Х	Х	818	299	<0,01	0,88	996
linear model	m3	Х	Х	Х	1026	508	<0,01	0,85	1008
generalized additive model	m2g	Х	Х		1843	1324	<0,01	0,69	1014
two order polynomial model	m2p	Х	Х		1843	1324	<0,01	0,69	1018
linear model	m2	Х	Х		1845	1327	<0,01	0,67	1019
linear model	m1	Х			2967	2448	<0,01	0,02	1030
generalized additive model	m1g	Х			2967	2448	<0,01	0,04	1026
two order polynomial model	m1p	Х			2968	2449	<0,01	0,04	1029
linear model	m0				2987	2468	<0,01	/	1031
two order polynomial model	m0p				2987	2468	<0,01	/	1031
generalized additive model	m0g				2987	2468	<0,01	/	1031

10 11 12

- 13
- 13
- 14
- 15
- 16
- 17
- 18 19
- 20



Figure 1. Map of forested areas (light green color; Corinne Land Cover 2006) within the studied spatial extent located in North France (Aisne; Hauts-de-France) with the 5 focal study sites labelled

- from 1 to 5: (1) the Haye of Aubenton; (2) the Val-Saint-Pierre forest; (3) the Andigny forest; (4) the
- 25 Plomion wood (East) and the Thenailles ancient forest now deforested (West); and (5) the Metz-en-

- 26 Couture/Havrincourt ancient forest now partly deforested. The dark green color refers to ancient
- 27 forests still afforested while the brown color refers to ancient forests now deforested.
- 28



29 30 31

32 Figure 2. Geohistorical reconstruction of the Plomion wood area: (a) old map (1766) of the Foigny's 33 abbey domain (AD02 H658b); (b) Napoleonian cadaster with associated land-use status (1814) and 34 Etat-Major map (1826); (c) aerial photograph (1949) (Source: IGN-Geoportail); (d) aerial photograph 35 (1991) (Source: IGN-Geoportail); (e) aerial photograph (2013) (Source: IGN-Geoportail); and (f) 36 geohistorical reconstruction made within a geographic information system (GIS) (Source: ArcGIS-37 ESRI). Red dots on the maps show the exact locations of the studied soil profiles (n = 24) within the 38 Plomion wood study area with some soil profiles located in the ancient forest of Plomion afforested 39 since a very long time (before 1194) and some located on different deforestation chronosequences ranging from recent ( $F \rightarrow C_{2009}$ ) to older ( $F \rightarrow C_{1985}$ ) deforestation events. 40





Figure 3. Predicted soil pH values along the soil depth profiles for each of the 12 different levels
 characterizing the factor variable of land-use change (LUC) trajectories (cf. the afforestation and
 deforestation chronosequences). Predictions are based on the best model (see Table 2) that accounts

for the interaction effect between soil depth and LUC trajectories as well as for complex non-linear

relationships between soil depth and soil pH. Shaded surfaces show the 95% confidence interval

48 (1.96xSEM; where SEM means standard error of the mean) around the estimated coefficients. The

50 meaning of each of the levels of the factor variable of LUC trajectories is fully described in the main

51 text of the manuscript (see the Materials and methods section).





Figure 4. Variation in soil pH values at 50cm depth for different afforestation chronosequences 54 depicted by the time elapsed since the afforestation. Soil profiles located on ancient cultivated fields 55  $(C \rightarrow C)$  are used as the baseline (cf. age = 0) whereas soil profiles located within ancient forests are 56 used as the other extreme situation of afforestation since a very long time (cf. age = 1500). For the 57  $C \rightarrow F_{1000-1500}$  level of the factor variable of land-use change (LUC) trajectories, data are positioned at 58 the median year after afforestation, i.e., 750 years ago. Box plots show data from this study whereas 59 red dots show pH<sub>Cacl2</sub> data from Geescroft site (Blake 1999) converted in pH<sub>water</sub> (Libohova and others 60 2014). The lower panel depicts a zooming window for the 0-150 years interval. 61

- 64 How to cite this article: Brasseur B, Spicher F, Lenoir J, Gallet-Moron E, Buridant J, Horen H. What
- 65 deep-soil profiles can teach us on deep-time pH dynamics after land use change?. Land Degrad
- 66 Dev. 2018;1–11. https://doi.org/10.1002/ldr.3065