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15 Abstract

16 Soil profiles keep records of the legacies of historical land uses on soil physicochemical
17 properties with deep-soil (3m depth) profiles providing information on centuries-old
18 dynamics. By combining geohistorical archives on past land-uses and management practices
19 together with soil pH data from 19 plots scattered across five study areas in North-France,
20 we analyzed the effect of two contrasting historical land-use change trajectories
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
23 covariance (ANCOVA) and a generalized additive model (GAM) were used to test the
24 interaction effect between depth and the two studied chronosequences levels on soil pH.
25 Results show two contrasting alkalinization dynamics for formerly acidic forest soils
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
28 alkalinization dynamic was much slower (2-4 centuries are necessary to neutralize the
29 relictual acidity below 2m) under liming based on chalk-blocks supply. Per contra,

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 deep-soil pH profiles with chronosequences is a powerful tool to understand the potential impact of future land-use change trajectories on soil physicochemical properties.

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

Introduction

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.*, 2015). Since the Neolithic, farmers and foresters throughout their activities are influencing the pedogenesis and evolution of anthropogenic landscapes (Van Vliet-Lanoe *et al.*, 1992; Langhor, 2001; Emadodin *et al.*, 2009). Imprints of past land uses (e.g., croplands, grasslands, woodlands) are still visible in current biodiversity patterns (Closset-Kopp & 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.*, 2008). Among soil physico-chemical parameters, potential hydrogen (pH) is a very good indicator of land-use changes (LUCs) from forest to arable land and *vice versa* (Blake *et al.*, 1999; Bossuyt *et al.*, 1999; Verheyen *et al.*, 1999). Soil pH controls pedogenetic processes 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 animal biodiversity (see for example, Chauchan, 2014; Fichtner *et al.*, 2014), hence soil pH is a key parameter in pedogenetic dynamics through time.

The dynamic of soil alkalization results from a negative proton balance which can be triggered by calcic mineral amendment (e.g., limestones, lime, marls) in agricultural regions under temperate humid climate. These agronomic practices, first described during the 1st century BC by Varron (Ed. 1864: book I - chapter VIII) and much more detailed later on by Pliny the Elder (Pliny the Elder, 1877: book XVII - chapter 4), are very old in north-western Europe and varies in nature (Goulding, 2016), amount and frequency of inputs (Tables S1-S2). Liming is required as cereals growth is seriously limited on acid soils of formerly forested lands which were deforested to extend land cultivation. Thus in North 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 deforestation are from the 12th century (Fossier, 1987) (i.e., the “sartier” contracts). Other farming practices than liming such as fertilization, product removal and crop rotations may also affect the proton (H^+) and hydroxyl (OH^-) budgets (Merry, 2009; Schroder *et al.*, 2011) and thus the dynamic of soil alkalization. Not only the dynamic of soil alkalization due to 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 investigated (de Schrijver *et al.*, 2012). For instance, the impact of LUC on soil pH is known to persist over 30-80 years of forest growth on formerly agricultural lands irrespective to the pH measurement method, i.e., pH_{water} , pH_{KCl} or pH_{CaCl_2} (Ritter *et al.*, 2003; Falkengren-Grerup *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 Schrijver *et al.*, 2012), albeit land-use history seems to influence soil properties more than the selected tree species (Ritter *et al.*, 2003).

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) nicely emphasized this issue by asking: “*What do ecologists miss by not digging deep enough?*”. Drouet *et al.* (2015) provided some answers by showing that deep nutrients could significantly contribute to tree growth. Only a handful of studies, focusing on loessic luvisols exclusively, examined the long-term acidification or alkalinisation dynamics through deep soil profiles. The experimental site of Rothamsted farm (UK) is a good example where soil profiles were sampled up to 90cm depth during a period of 110 years after the afforestation of cultivated chromic luvisols. The $\text{pH}_{\text{CaCl}_2}$ profiles of this experimental site revealed a net surface acidification from 6.2 to 3.8, whereas the deepest horizons of the investigated soil 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 Belgium and found a very marked decline in pH_{KCl} during the first 30 years after afforestation followed by a slower decline in the next 20 years after which pH_{KCl} was similar to the level observed in an ancient forest. Regarding the long term dynamic of soil alkalinisation, Pécrot (1956) focused on cultivated soils after deforestation in Belgium and concluded that the 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 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 ($\text{pH}_{\text{water}} = 5.3$) after 100 years of cultivation while a neutral (around 6.5) soil profile was observed up to 165cm depth after 250 years of cultivation. Hence, deep-time (>100 years) acidification and alkalinization dynamics following LUCs still remain unknown and yet could provide important 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

pH_{water} profiles on luvisols in North France were examined to answer the following questions: (i) How long are the impacts of past LUCs (e.g., cultivation after deforestation and afforestation after cultivation) still noticeable on soil pH? (ii) What are the vertical and temporal dynamics of soil alkalization and acidification? (iii) Is soil pH a good proxy to date past LUCs?

Materials and methods

Study area

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 forests sites: (1) the Haye of Aubenton (AU; 4 plots); (2) the Val-Saint-Pierre forest (VSP; 4 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 sea level and the climate is humid temperate: mean annual air temperature is 8.75°C; 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 (*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%) to avoid soil erosion and spatial heterogeneity in acid neutralizing capacity due to topography (Brahý *et al.*, 2000).

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

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 ranging from “recently afforested” to “afforested since a very long time”: a medieval strip field afforested 15 years ago ($C \rightarrow F_{1999}$); a field afforested 80 years ago ($C \rightarrow F_{1935}$); two medieval strip fields afforested 150 years ago ($C \rightarrow F_{1865ck}$ & $C \rightarrow F_{1865}$; with and without Ck loessic lower horizon, respectively); one medieval strip field afforested 500 to 1000 years ago ($C \rightarrow F_{1000-1500}$); and four ancient forests ($C \rightarrow F_{500AU+VSP+PL+AN}$). These four ancient forests were already forested during the 12th or 13th century (Table S3) but could have been cultivated during bronze-iron ages and/or antiquity as many west European ancient forests (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 Medieval or Modern sites were found suggesting that these areas could have been cultivated during antiquity and afforested as early as the troubled period of the end of the Western Roman Empire during the 5th century AD. The absence of the upper part of albeluvic tonguing on these luvisols (ISSS Working Group RB, 1998) as well as the brownish 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→F₅₀₀ plots.

The deforestation chronosequence contains six different levels along an ordinal scale ranging from “recently deforested” to “deforestation since a very long time”: several formerly ancient forests deforested about 5 (F→C₂₀₀₉); 30 (F→C₁₉₈₅); 140 (F→C₁₈₇₅); 150 (F→C₁₈₆₅); and 170 (F→P₁₈₄₅→C₁₉₂₀ & F→C₁₈₄₅; with F→P₁₈₄₅→C₁₉₂₀ being a pasture without liming practices, but manured, during the first 85 years following deforestation and then being limed & cultivated during the next 85 years until now) years ago. In addition to those six levels, four cultivated ancient fields (C→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 deforestation chronosequence excepting the control plots were located in sites previously described as medieval forests (F→C₂₀₀₉, F→C₁₉₈₅, F→C₁₈₇₅, F→C₁₈₆₅, F→P₁₈₄₅→C₁₉₂₀ and F→C₁₈₄₅).

Following Pliny the Elder (Pline l’ancien, 1877), liming practices were already used by Gallic, Germanic and Britannic farmers for cereals culture during Antiquity. In North France, the use of white chalk (sometimes miscalled marl) is confirmed from the beginning of the high medieval period and marling contracts were recorded for lands after deforestation (Derville, 1999; Fossier, 1987). Note that liming practices (amount and periodicity of inputs) were likely variable over time (Table S1). Actual practices from the farmers who currently 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 Rothamsted farm, that CaCO₃ mean loss rate were ranging from 368kg/Ha/yr (for 5T/Ha 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 greater amount once and/or more frequently over time can help to increase soil pH values 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 maintain and even raise soil pH values (Tables S1 and S2).

Soil sampling and measurement methods

Between 2013 and 2016, a total of 6 soil profiles were sampled for each of the 19 studied plots (C→C_{AN}, C→C_{VSP}, C→C_{AU}, C→C_{BP}, C→F_{500AN}, C→F_{500VSP}, C→F_{500AU}, C→F_{500BP}, F→C₂₀₀₉, F→C₁₉₈₅, F→C₁₈₇₅, F→C₁₈₆₅, F→P₁₈₄₅→C₁₉₂₀, F→C₁₈₄₅, C→F₁₉₉₉, C→F₁₉₃₅, C→F_{1865ck}, C→F₁₈₆₅, C→F₁₀₀₀₋₁₅₀₀) excepting C→C_{AN}, C→C_{VSP}, C→C_{AU} and F→C₁₈₇₅, where only 3 soil profiles were sampled, thus totalizing 102 soil profiles across the entire study area (Table S4). By using an 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 plot. The soil augering descriptions were carried out following the FAO standard guidelines. Soil horizon depths and textures were noted and Munsell colors were determined on moist samples. For each of the 102 sampled soil profiles, we characterized the humus (Brethes *et al.*, 1995) and noted the humus index (Ponge & Chevalier, 2006) as well as the colors of the soil horizons (Table S5). None of the 102 profiles had any archaeological remains, topographic anomalies, natural (windthrow) or anthropogenic disturbances, anomaly of humus thickness (Bossuyt *et al.*, 1999) or a tree trunk that was closer than 2m (Beniamino *et 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.

Data analyses

To detect an imprint of past LUCs on soil pH profiles and represent the inter- and intra-plot variability in the data ($n = 1044$), we ran several statistical models testing the impact of different LUC trajectories (the main explanatory variable) on soil pH (the response variable) while controlling for the potential confounding effect of soil depth (a covariate) and its potential interaction with past LUCs. Soil pH and depth were treated as continuous variables while the variable representing the different LUC trajectories of the afforestation and deforestation chronosequences tested here was treated as a factor variable with 12 levels ($C \rightarrow C$, $C \rightarrow F_{500}$, $F \rightarrow C_{2009}$, $F \rightarrow C_{1985}$, $F \rightarrow C_{1865}$, $F \rightarrow P_{1845} \rightarrow C_{1920}$, $F \rightarrow C_{1845}$, $C \rightarrow F_{1999}$, $C \rightarrow F_{1935}$, $C \rightarrow F_{1865ck}$, $C \rightarrow F_{1865}$, $C \rightarrow F_{1000-1500}$). Note here that the $F \rightarrow C_{1875}$ level of LUC trajectory was not 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 model (GAM), we tested four candidate models: a null model (or intercept model) without any explanatory variable as a control or baseline ($m0: pH \sim 1$); a model that tests the effect of soil depth on soil pH ($m1: pH \sim \text{Depth}$); a model that tests the effect of the different LUC trajectories while controlling for soil depth ($m2: pH \sim \text{Depth} + \text{LUC}$); and a model that tests the interaction effect between soil depth and LUC trajectories ($m3: pH \sim \text{Depth} * \text{LUC}$). The OLS regression approach assumes a linear relationship between soil pH and depth while the 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 depth. In total, we ran 3x4 different models (Table 2) and we used the Akaike information criterion (AIC) to select the best model (Burnham & Anderson, 2002), which is the one with

the lowest AIC value (AIC_{min}). We also computed the relative support for each model, using the differences in AIC values, $\Delta AIC_i = AIC_i - AIC_{min}$, where AIC_i is the observed AIC value of the focal model i and AIC_{min} 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 ($CI = 1.96 \times SEM$; 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).

Results

Soils’ description

The 102 soils exhibited a contrasted development of eluvial (E) and illuvial (Bt) horizons. The diffuse transition horizon (BC) and C decarbonated loess material mostly constitute the lower half part of the soil profile whereas calcareous loess material was completely absent except for C→F_{1865ck}, which exhibited calcareous loess at 220-250cm depth. For afforested soils, the humus index score increased from 1 (i.e., eumull) to 3.6 (i.e., oligomull-dysmull) with increasing afforestation age (Table S5) and the A/E transition appeared between 12cm (C→F₅₀₀) and 25cm (C→F₁₉₉₉) depth. For tilled cultivated soils, the Ap/E transition showed a lesser variation with a range between 26cm (F→C₁₈₄₅) and 35cm (F→C₂₀₀₉).

The E horizons were bioturbated with some soils sampled in the Aubenton site (C→F_{500AU}, C→F₁₈₆₅ and C→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 found between 33cm (F→C₁₈₆₅) and 60cm (AF) depth. Importantly, the upper parts of the transitional albeluvic tonguing between the E and Bt horizons were systematically truncated and leveled by bioturbation, even on C→F₅₀₀ luvisols which suggests cultures as past land uses. The Bt horizon of most plots showed light grey polygonal crack networks typical of a fragipan. Hence, the soils are classified (IUSS Working Group WRB, 2006) in luvisols but still bear relict patterns of albeluvisols. All soils' horizons textures are very homogeneous, from silt loam in the surface to silty clay loam for the deep part (Figure S2), albeit the sand content of C→F₁₉₉₉ raised from 100cm towards the deepest part.

Soil pH profiles of ancient forested (C→F₅₀₀) and ancient cultivated (C→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→F_{1865ck}). Soil pH in C→F₅₀₀ 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→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.

Chronosequences

For the youngest afforested plot ($C \rightarrow F_{1999}$), neutro-alkaline pH values were still found below 75cm depth but decreased significantly to 6.66 and 6.03 at 50cm and 5cm depth, respectively. Soil pH values in the $C \rightarrow F_{1999}$ LUC trajectory significantly differed from the 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 than the $C \rightarrow C$ reference all along the soil profile (Figure 3), with pH dropping to 6.10 at 5cm 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 greater than the $C \rightarrow F_{500}$ reference only for the $C \rightarrow F_{1865}$ LUC trajectory (Wilcoxon signed-rank test: $W = 131$, $p = 0.0012$). At 175cm depth, soil pH increased up to 7.07 for the $C \rightarrow F_{1865ck}$ LUC trajectory, thus being not significantly different from the $C \rightarrow C$ reference. The oldest afforested plot ($C \rightarrow F_{1000-1500}$) exhibited greater mean pH values than the $C \rightarrow F_{500}$ reference 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 250cm depth (Wilcoxon signed-rank test: $W = 91$, $p = 0.0551$).

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 300cm depth) (Table 1 and Figure 3). On $F \rightarrow C_{1875}$ and $F \rightarrow C_{1865}$ plots, about 150 years after deforestation and cultivation, soil pH values were neutro-alkaline (≥ 7) from topsoil down to 300cm depth. For the $F \rightarrow P_{1845} \rightarrow C_{1920}$ LUC trajectory, the neutro-alkaline pH values observed between the surface and 100cm depth sharply declined to acid values (5.51 at 250cm depth) more or less similar to the $C \rightarrow F_{500}$ reference. The $F \rightarrow C_{1845}$ LUC trajectory showed weakly acid to neutral pH values from topsoil (pH = 6.09 at 5cm depth) to the deep C horizon (pH = 6.33 at 250cm depth) and through the Bt horizon (pH = 7.02 at 75cm depth).

Discussion

313

314 *Alkalinization and acidification dynamics*

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

320 On the deforestation and culture chronosequence, time since deforestation clearly
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
323 alkalinization process is confined within the ploughed horizon (Ap) with a sharp decline in pH
324 values towards initial acidic conditions, reflecting former land use as forest. Following the
325 Rothamsted's model ($\Delta pH = 0.109L - 0.690$; L is lime added as T/Ha $CaCO_3$) (Goulding *et al.*,
326 1986; Goulding, 2016), we found that for a topsoil pH value of 4.6 (equivalent to ancient
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
329 reaching 7.1 in the Ap horizon (Table 1).

330 Coherently, for older deforestation events that happened 140 to 170 years ago (e.g.,
331 $F \rightarrow C_{1865}$), the alkalinization process migrates (more or less rapidly) towards the deepest soil
332 layers. Interestingly and for LUC trajectories being more or less equivalent in terms of time
333 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
334 migration rate of this alkalinization front likely depends on the former management
335 practices used by farmers. For cultivated soils limed with defecation lime ($F \rightarrow C_{1875}$ and
336 $F \rightarrow C_{1865}$), the alkalisation dynamic operated all along the soil profile and soil pH values are
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 alkalisation process was much less

pronounced and pH values gently decline towards acid values in the deep soil layers. For the F→P₁₈₄₅→C₁₉₂₀ LUC trajectory, for which successive liming practices were restricted to the last 85 years, the migration rate of the alkalization front is more or less intermediate with pH values declining towards initial acidic conditions at 3m depth. These results suggest that the high solubility of Ca(OH)₂ in defecation lime (Table S6) has triggered faster and deeper soil alkalization rates through winter infiltrations of waters (Girard *et al.*, 2005) highly concentrated in OH⁻ ions (strong base). In contrast, the low solubility of coarse chalk fragments limits the basicity of infiltration waters, which bring few CO₃²⁻ (weak base).

As expected for the afforestation chronosequence, we found a progressive acidification process over time, from topsoil to deeper soil horizons, with the oldest afforestation events (C→F₁₀₀₀₋₁₅₀₀) showing the largest decrease in soil pH values relative to the ancient culture (AC) reference. The magnitude and rate of this acidification dynamic, relatively to the C→C reference, is more important towards the soil surface where the main sources of acidity are concentrated: litter decomposition (Markewitz *et al.*, 1998; Reich *et al.*, 2005); root exudation and respiration (Hinsinger *et al.*, 2003); and atmospheric precipitations (Van Breemen *et al.*, 1983). Below pH = 5.5, soil Al solubilization trigger acidity buffering, which decrease acidification kinetic on the acid forest plots. Hence, depending on the time since afforestation, the imprint of the former cultivation on soil pH can still be visible in the deepest part of the soil profile. For instance, 150 years after afforestation (C→F₁₈₆₅), soil pH values below 200cm depth are more or less similar to the C→C reference. Noteworthy, our results suggest that the alkalisation process after deforestation and culture is faster (F→C₁₈₄₅; chalk) to much faster (F→C₁₈₆₅; defecation lime) than the acidification process after afforestation.

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 (B₂₁ horizon) and 125cm (B₃) 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 by Ha, granulometry) there is a high variability of alkalinisation dynamics after deforestation 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 slower. Because of this variability, it appears difficult to date a deforestation event just from 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 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→F₅₀₀ 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→F₁₉₃₅ & C→F₁₈₆₅ 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_{CaCl2} values, converted in pH_{water} (Libohova *et al.*, 2014), from the Rothamsted

experimental station (Figure 4, Blake *et al.*, 1999) where the climatic conditions are close to our study area. Both datasets show high consistency and suggest that soil pH at 50cm depth is negatively correlated with afforestation age during the first 150 years (see Figure 4). The 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 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→F_{1865ck} LUC trajectory which still show high pH values at 3m depth due to loess initial calcium carbonate. As 2-4 centuries of chalk liming practices are necessary to complete deep soil deprotonation and re-saturate exchange complex with Ca²⁺ (Colinet, 2003), only the afforestation plots on ancient fields display deep neutro-alkaline profiles when acidification began.

Conclusion

Our findings suggest that one century of liming practices with fine-grained hydrated lime is enough to reach a complete deprotonation of formerly acid forest soils. A much slower alkalization is observed when coarse chalk (or limestone) blocks are used during 170 years 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 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 depend upon the wide diversity of liming practices through time rather than time elapsed since deforestation and culture. Per contra, imprints of former agricultural lands limed during at least two to four centuries will still be visible in the deepest soil layers more than millennia after afforestation. The acidification dynamic is more regular and progressive from 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.

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Supporting information

Figure S1: Histogram of residuals on the best AIC score model (m3g)

Figure S2: Soil textures (Pipette method NF X31-107) ratio values for Clay-Silt-Sand

Figure S3: Mean pH of the plots studied in the two chronosequences: a-afforestation on culture and b-deforestation and culture

Table S1: Local liming practices from the lower medieval period to the modern period and antique liming practices over Gaul and Britain territories.

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) afforestation and b) deforestation chronosequences

Table S4: Number of soil samples at each soil depth and for each of the 13 levels 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

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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)	C→F ₅₀₀		C→F ₁₀₀₀₋₁₅₀₀		C→F _{1865ck}		C→F ₁₈₆₅		C→F ₁₉₃₅		C→F ₁₉₉₉		F→C ₂₀₀₉		F→C ₁₉₈₅		F→C ₁₈₇₅		F→C ₁₈₆₅		F→P ₁₈₄₅ →C ₁₉₂₀		F→C ₁₈₄₅		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 non-linear relationships. Difference in Akaike Information Criterion (AIC) values between the best model and the focal model (ΔAIC), Akaike weights (w_i), adjusted R^2 values (R^2_{adj}) as well as degrees of freedom (Df) are provided for comparison purposes.

Modelling approach	Id	Depth	LUC	Depth x LUC	AIC	ΔAIC	w_i	R^2_{adj}	Df
generalized additive model	m3g	X	X	X	519	0	0	0,91	968
two order polynomial model	m3p	X	X	X	818	299	<0,01	0,88	996
linear model	m3	X	X	X	1026	508	<0,01	0,85	1008
generalized additive model	m2g	X	X		1843	1324	<0,01	0,69	1014
two order polynomial model	m2p	X	X		1843	1324	<0,01	0,69	1018
linear model	m2	X	X		1845	1327	<0,01	0,67	1019
linear model	m1	X			2967	2448	<0,01	0,02	1030
generalized additive model	m1g	X			2967	2448	<0,01	0,04	1026
two order polynomial model	m1p	X			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

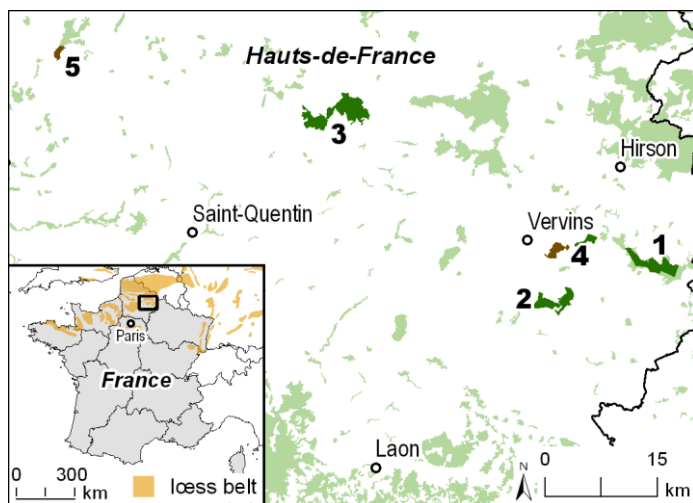


Figure 1. Map of forested areas (light green color; Corinne Land Cover 2006) within the studied spatial extent located in North France (Aisne; Hauts-de-France) with the 5 focal study sites labelled from 1 to 5: (1) the Haye of Aubenton; (2) the Val-Saint-Pierre forest; (3) the Andigny forest; (4) the Plomion wood (East) and the Thenailles ancient forest now deforested (West); and (5) the Metz-en-

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

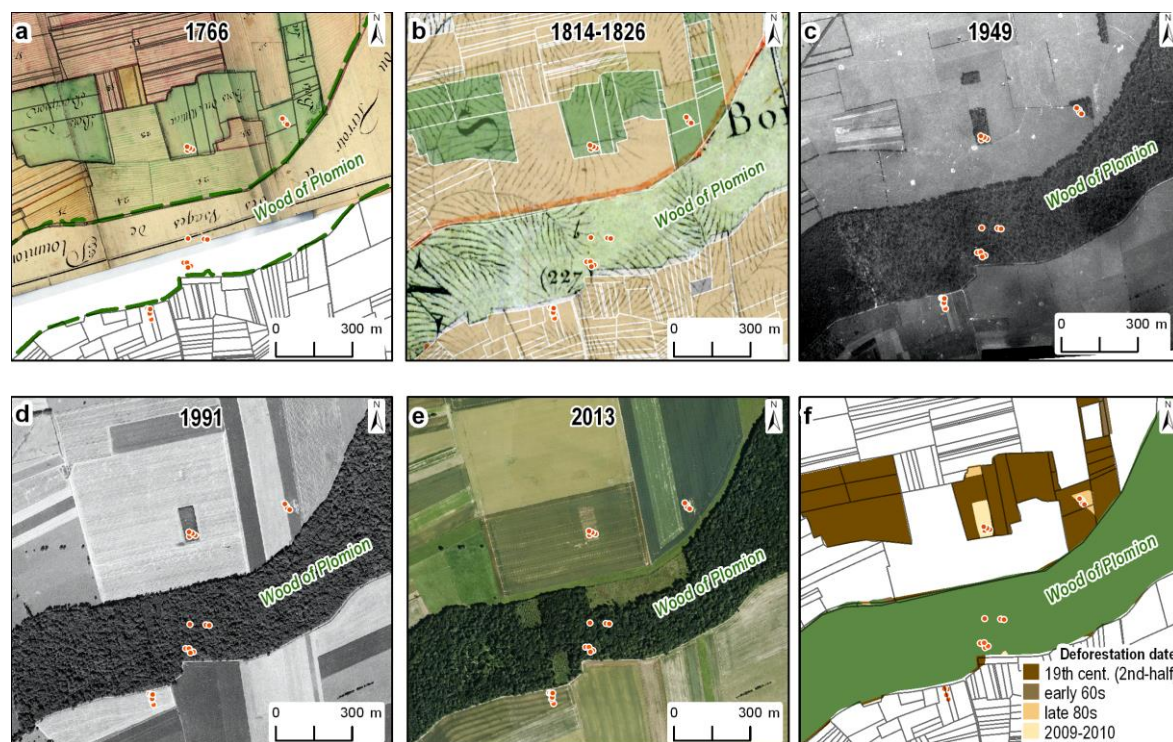


Figure 2. Geohistorical reconstruction of the Plomion wood area: (a) old map (1766) of the Foigny's abbey domain (AD02 H658b); (b) Napoleonian cadaster with associated land-use status (1814) and Etat-Major map (1826); (c) aerial photograph (1949) (Source: IGN-Geoportail); (d) aerial photograph (1991) (Source: IGN-Geoportail); (e) aerial photograph (2013) (Source: IGN-Geoportail); and (f) geohistorical reconstruction made within a geographic information system (GIS) (Source: ArcGIS-ESRI). Red dots on the maps show the exact locations of the studied soil profiles ($n = 24$) within the Plomion wood study area with some soil profiles located in the ancient forest of Plomion afforested 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.

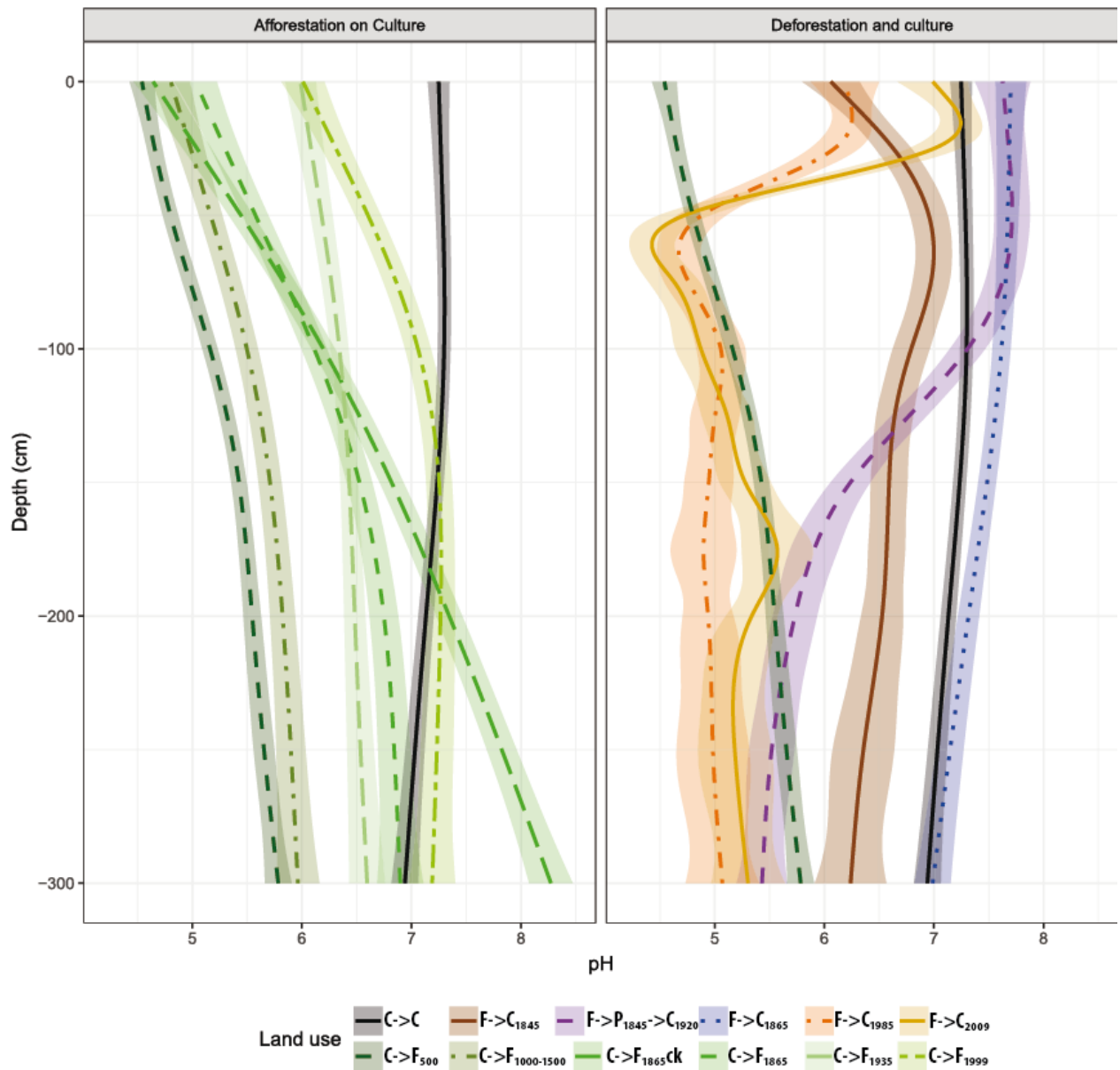


Figure 3. Predicted soil pH values along the soil depth profiles for each of the 12 different levels characterizing the factor variable of land-use change (LUC) trajectories (cf. the afforestation and deforestation chronosequences). Predictions are based on the best model (see Table 2) that accounts 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 (1.96xSEM; where SEM means standard error of the mean) around the estimated coefficients. 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).

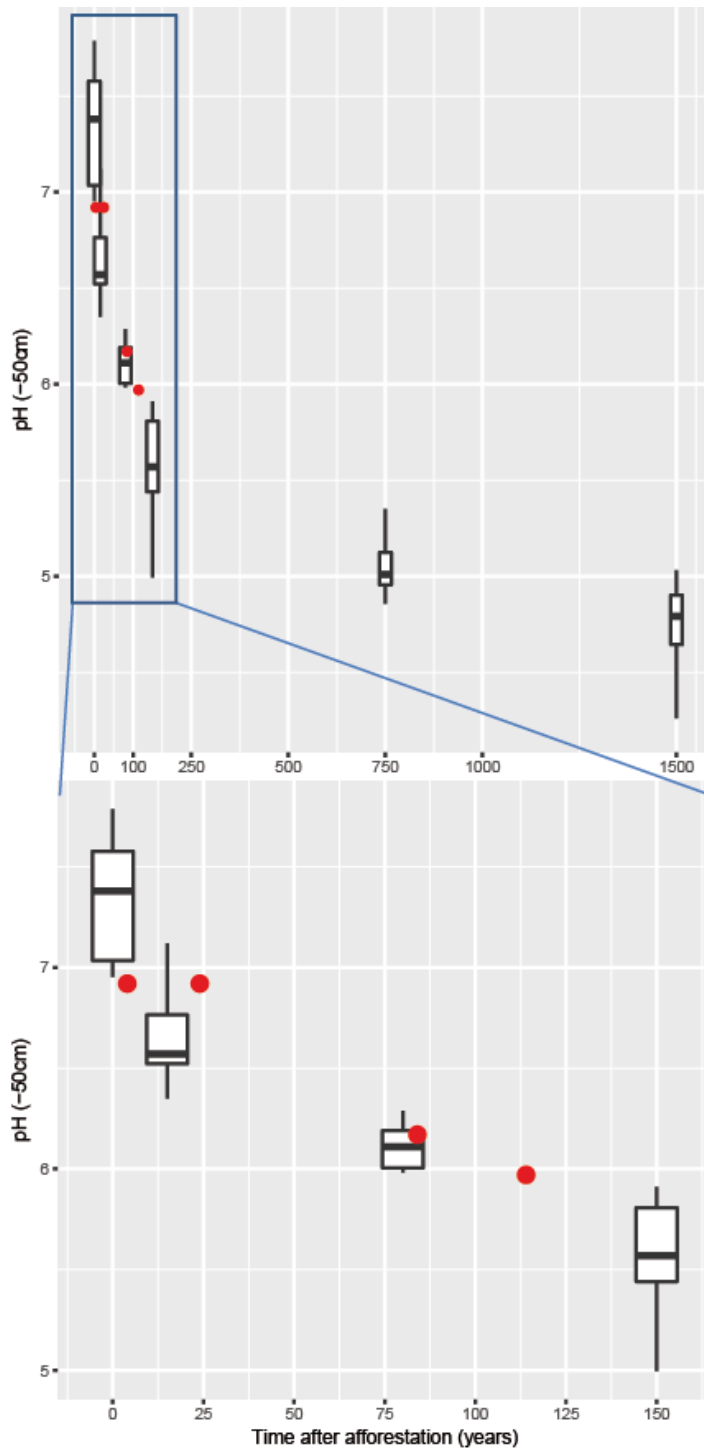


Figure 4. Variation in soil pH values at 50cm depth for different afforestation chronosequences depicted by the time elapsed since the afforestation. Soil profiles located on ancient cultivated fields (C→C) are used as the baseline (cf. age = 0) whereas soil profiles located within ancient forests are used as the other extreme situation of afforestation since a very long time (cf. age = 1500). For the C→F₁₀₀₀₋₁₅₀₀ level of the factor variable of land-use change (LUC) trajectories, data are positioned at the median year after afforestation, i.e., 750 years ago. Box plots show data from this study whereas red dots show pH_{CaCl2} data from Geescroft site (Blake 1999) converted in pH_{water} (Libohova and others 2014). The lower panel depicts a zooming window for the 0-150 years interval.

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