Geoderma 266 (2016) 46–57

Contents lists available at ScienceDirect

Geoderma

journal homepage: www.elsevier.com/locate/geoderma

Illuviation intensity and land use change: Quantification via micromorphological analysis

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ARTICLE INFO

Article history:
Received 11 August 2015
Received in revised form 25 November 2015
Accepted 29 November 2015
Available online xxxx

Keywords:
Clay translocation
Cultivation
Clay coatings
Luvisols
Pedogenesis

ABSTRACT

Among soil processes governing the clay size fraction distribution with depth, illuviation is not only widespread but also particularly poorly understood. Using a micromorphological approach, this study aims at i) quantifying the intensity of illuviation independently of all other competing soil processes, ii) testing the sensitivity of illuviation to different land uses, and iii) assessing the relative contribution of illuviation to the genesis of textural differentiation. Two Luvisols developed from loess in Northern France were selected: the first under a deciduous forest and the second under conventional agricultural management, both with no change in land use for at least 100 years. In addition to classical mass balance calculations, clay illuviation features such as limpid and dusty clay coatings were used as diagnostic features of the illuviation process and were quantified by a point-counting approach in large thin sections. In the two Luvisols under study illuviation was found to be responsible for 75% and 86% of the textural contrast, respectively in the cultivated soil and the forested one. Illuviation was thus by far the dominant factor causing textural differentiation, even if accessory processes involving the clay size fraction were also at play such as lithological discontinuity, clay neoformation or biological and physical reworking. The recognition of dusty clay coatings considered as an indicator of cultivation in the soil under forest underlines the sensitivity of illuviation to past periods of cultivation and the legacy of anthropedogenesis on the present soil properties. A qualitative change in the nature of the translocated particles without significant changes in the intensity of illuviation was identified in the soil under cultivation in comparison with the soil under forest. These results support the conclusion that illuviation is an active process in soils under present climatic conditions and must no more be considered as a fossil soil process.

1. Introduction

Humanity has strongly influenced soil evolution since the beginning of the Anthropocene Era through cultivation or urbanization manipulation purposes and is now considered as a primary geomorphologic agent (Lin, 2011; Richter, 2007). In order to predict the effects of land use on soil function and finally prevent soil degradation, land managers and policy makers need information about soil change and the associated natural or anthropic-driving factors (Tugel et al., 2005). Dynamic soil processes with short characteristic times (years to decades) are used to be more investigated than processes involved in soil formation and supposed to evolve over larger time scales (centuries to millennia) (Minasny et al., 2008; Targulian and Krasilnikov, 2007). Soil was indeed classically considered as a slowly changing natural system and its formation is seen as a very slow process, with mature soil ages averaging between 10^4 to 10^5 years (Wilkinson and Humphreys, 2005). Nevertheless, various authors have documented significant changes of soil formation processes dynamics on time scales as short as a few decades, and they have argued that soils are evolving permanently (Chadwick and Chorover, 2001; Cornu et al., 2012; Montagne et al., 2008).

Illuviation is one of the earliest recognized soil-forming process (Bockheim et al., 2005) and is involved in the genesis of many soil types under many climate types. It consists of a substantial vertical translocation of fine particles up to 10 μm in size (Quénard et al., 2011) and results in the formation of an eluviated subsurface horizon called E-horizon and, an illuviated one underneath due to the accumulation of the fine fraction and also called Bt-horizon (WRB, 2014). This process is mainly responsible for the formation of Luvisols that extend over 500–600 million ha worldwide according to WRB (2014). Some authors consider illuviation as a fossil process (Slager and van de Wetering, 1977; Van Vliet-Lanoë, 1990) whereas others have documented significant changes of illuviation intensity as a result of anthropic-driving forces (Cornu et al., 2012; Kozlovskii et al., 2001; Montagne et al., 2008). Despite considerable past research and because

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http://dx.doi.org/10.1016/j.geoderma.2015.11.035

of a lack of accurate quantitative data about illuviation, two main issues are still particularly controversial: the present relevance of the illuviation process and its sensitivity to land use management.

Illuviation is manifested through a typical granulometric clay distribution along soil profiles due to the textural differentiation between the eluviated and the illuviated horizons, and the observations at a macromorphological or micromorphological scale of clay coatings in the Bt-horizons. Illuviation intensity is generally measured through a textural differentiation index between the eluvial and illuvial horizons. Unfortunately, data related to this differentiation are difficult to interpret unambiguously, because it can be caused by several different processes such as in-situ weathering, bioturbation, and microdivision (Phillips, 2004).

An alternative to the use of multi-processes textural differentiation indexes may be the quantification of diagnostic soil features specifically associated with the illuviation process, i.e., clay coatings. Such a morphological approach is acknowledged to have succeeded in quantifying the intensity of the genesis of albic materials in soils (Montagne et al., 2013). Using a micromorphological approach makes it possible to distinguish features that are due to illuviation process among others (Kemp, 1998). Clay coatings are indeed considered to have illuvial origin, depending on specific indicators such as the degree of clay particle orientation, internal laminations, and the sharpness of boundaries with the surrounding groundmass (McCarthy et al., 1999; Ufnar, 2007). Furthermore, describing textural pedofeatures at a microscopic scale helps to understand their genesis and associated pedoclimatic context of formation. For example microlaminated limpid clay coatings are associated with typic clay illuviation, i.e., under permanent vegetation cover (Fedoroff, 1997) whereas layered dusty or impure clay coatings are associated with heavy rain showers and drastic conditions and are generally met under cultivation (Fedoroff and Courty, 1994; Jongmans et al., 2001; Kühn et al., 2010). At this stage, a detailed comparison of the classical granulometric quantitative approach with the micromorphological one has yet to be carried out.

In this context, the aim of the present study is to carry out the micromorphological description and quantification of clay coatings in soil thin sections from a forested and a cultivated Luvisol. The key objectives of the research are to provide a proxy for the intensity of the illuviation process independently of other competing soil processes, to test the sensitivity of illuviation intensity to different land uses, and, to quantify the relative contribution of illuviation to the genesis of soil textural differentiation through a comparison to classical multi-processes granulometric calculations. Our working hypothesis is that this feature-based approach can provide a more accurate quantification of illuviation intensity than classical granulometric measurements in soils facing different anthropic-driving forces.

2. Materials and methods

2.1. Study area

In our investigation, we studied two Luvisols (WRB, 2014; Table 1) of approximately 1.5 m depth, which have developed in decarbonated quaternary loess deposits in the Paris basin. As discussed in Jagercikova et al. (2015), three successive loess deposits may be distinguished. The first one is observed from the surface horizon to the Bt-horizon and shows a coarse to fine silt ratio ranging from 1.4 to 1.7. The second deposit is characterized by a coarse to fine silt ratio higher than 2, whereas the third one has a ratio around 1.9 (Table 2). The third deposit was dated at ~22 ka ^{14}C BP and the two upper deposits correspond to the youngest loess unit of 15 to 16 kyr. One Luvisol is under conventional agricultural management (CULT) and is part of the so-called Qualiagro long-term soil experiment located on the Plateau des Alluets (Feucherolles, Yvelines, France). Since 1998, the CULT soil is plowed once every year in October or November to a depth of 28 cm. It used to be limed regularly in order to keep a pH of 7 approximately in soil surface. The other plot (FOR), considered as a reference, was sampled in the Flambertins Forest, Yvelines, France. This deciduous forest with oaks and hornbeams is actually the only one i) extending on the Plateau des Alluets and consequently on Luvisols and, ii) sufficiently close to the Qualiagro long-term experiment (less than five hundred meters away) to ensure that both Luvisols have developed on a similar parent material.

Historical records indicate that the cultivated plot experienced no change in land use for at least the past two centuries. However, the forest may not be so old as initially thought, and the FOR plot may have been cultivated before being reforested at least 100 years ago (IGN/EHESS, 2015). Both soils present the typical horizons of Luvisols according to WRB (2014): A or Ap E Bt C (Fig. 1). Both sites are relatively flat (<1% slope) and under similar climatic conditions, with a mean annual precipitation of 582 mm and a mean annual temperature of 11 °C (Mercier et al., 2011).

Table 1
Soil macromorphology[a,b].

Horizon	Depth (cm)	Water state class	Color (moist)	Texture	Structure	Roots	Pedofeatures	Boundary
FOR								
A	0–10	Moist	10YR 3/3	sil	2mgr	1co, 2m, 2f	–	cw
E	10–40	Moist	10YR 4/4	sil	1fsbk	1vco, 1co, 2m, 2f	–	ai
Bt1	40–65	Moist	10YR 4/6	sil	1cosbk	2m, 2f	Clay coatings	cs
Bt2	65–90	Moist	7.5YR 4/6	sil	2mpr	2f	Clay coatings	gs
Bt/C	90–120	Dry	10YR 5/8	sicl	2mpr	2f	Clay coatings	gs
C/Bt	120–145	Dry	10YR 6/6	sil	m	1vf	Clay coatings	as
Ck	>145	Dry	10YR 7/6	sil	m	1vf	Secondary carbonates	
CULT								
Ap1	0–25	Moist	10YR 4/4	sil	2fsbk	3vf	–	as
Ap2	25–31	Moist	10YR 4/4	sil	1vfbk	2vf	–	as
E/Bt	31–45	Moist	10YR 4/6	sil	1fbk	2vf	Clay coatings, few Fe–Mn masses/nodules	aw
Bt	45–80	Moist	7.5YR 4/6	sicl	2mpr	1vf	Clay coatings, few Fe–Mn masses/nodules	gs
Bt/C	80–100	Moist	10YR 5/6	sicl	2mpr	1vf	Clay coatings, few Fe–Mn masses/nodules	gs
C/Bt	100–140	Moist	10YR 5/6	sil	m	–	Clay coatings, few Fe–Mn masses/nodules	as
C	>140	Wet	10YR 5/6	sil	m	–		

[a] Soil description according to the terminology of Soil Survey Staff.
[b] Abbreviations: Texture – sil = silt loam; scl = silty clay loam. Structure grade – 1 = weak; 2 = moderate; 3 = strong. Structure size – vf = very fine; f = fine; m = medium; co = coarse. Structure type – gr = granular; sbk = subangular blocky; bk = blocky; pr = prismatic; m = massive. Root quantity – 1 = few; 2 = common; 3 = many. Roots size – vf = very fine; f = fine; m = medium; co = coarse; vco = very coarse. Boundary distinctness – a = abrupt; c = clear; g = gradual. Boundary configuration – s = smooth; w = wavy; i = irregular.

Table 2
Selected physical and chemical properties.

Horizon	Depth (cm)	Particle size distribution ($g \cdot kg^{-1}$)[a]						Bulk density ($kg \cdot m^{-3}$)	pH (H_2O)	$CaCO_3$ ($g \cdot kg^{-1}$)	OC[b] ($g \cdot kg^{-1}$)	Total N ($g \cdot kg^{-1}$)	C/N
		cl	fsi	csi	fs	cs	csi/fsi						
FOR													
A	0–10	176	284	463	64	13	1.63	0.99	5.6	–	30.5	2.44	12.5
E	10–40	167	288	463	66	17	1.61	1.36	5.1	–	10.9	1.08	10.1
Bt1	40–65	216	284	428	56	16	1.51	1.51	5.6	–	3.6	0.48	7.5
Bt2	65–90	250	278	396	58	18	1.42	1.56	5.9	–	2.3	0.36	6.4
Bt/C	90–120	320	208	441	28	4	2.12	1.51	6.2	–	1.9	0.35	5.4
C/Bt	120–145	240	222	510	27	1	2.30	1.50	6.6	–	1.7	0.29	5.9
Ck	>145	137	240	457	19	2	1.90	1.49	8.6	143	0.9	0.21	4.3
CULT													
Ap1	0–25	142	296	495	54	13	1.67	1.34	6.6	–	9.4	0.88	10.7
Ap2	25–31	144	300	483	61	13	1.61	1.34	6.6	–	9.5	0.91	10.4
E/Bt	31–45	202	297	437	45	18	1.47	1.49	6.9	–	4.3	0.48	9.0
Bt	45–80	288	271	389	39	13	1.44	1.55	7.3	–	2.8	0.35	8.0
Bt/C	80–100	314	203	460	21	2	2.27	1.53	7.5	–	2.3	0.30	7.7
C/Bt	100–140	245	243	480	29	2	1.98	1.51	7.6	–	1.4	0.20	7.0
C	>140	190	266	525	17	1	1.97	1.54	7.7	–	1.3	0.18	7.6

[a] cl = clay (<2 μm); fsi = fine silt (2–20 μm); csi = coarse silt (20–50 μm); fs = fine sand (50–200 μm); cs = coarse sand (200–2000 μm).
[b] OC = Organic carbon.

2.2. Sampling procedures and associated measurements

In April 2011, soil pits, to a depth of 2 m, were dug in both sites. Bulk soil samples obtained in each soil horizon were oven-dried at 40 °C before being sieved to 2 mm. Particle size analysis of soil was performed by sieving and Robinson's pipette methods following the AFNORX31-107 methodology after removal of organic compounds with H_2O_2 and dispersion with Na-hexametaphosphate. pH was measured in a soil:water suspension of v:v ratio 1:5 (ISO 10390). Organic carbon content was quantified after dry combustion according to the NF ISO 10694. Soil

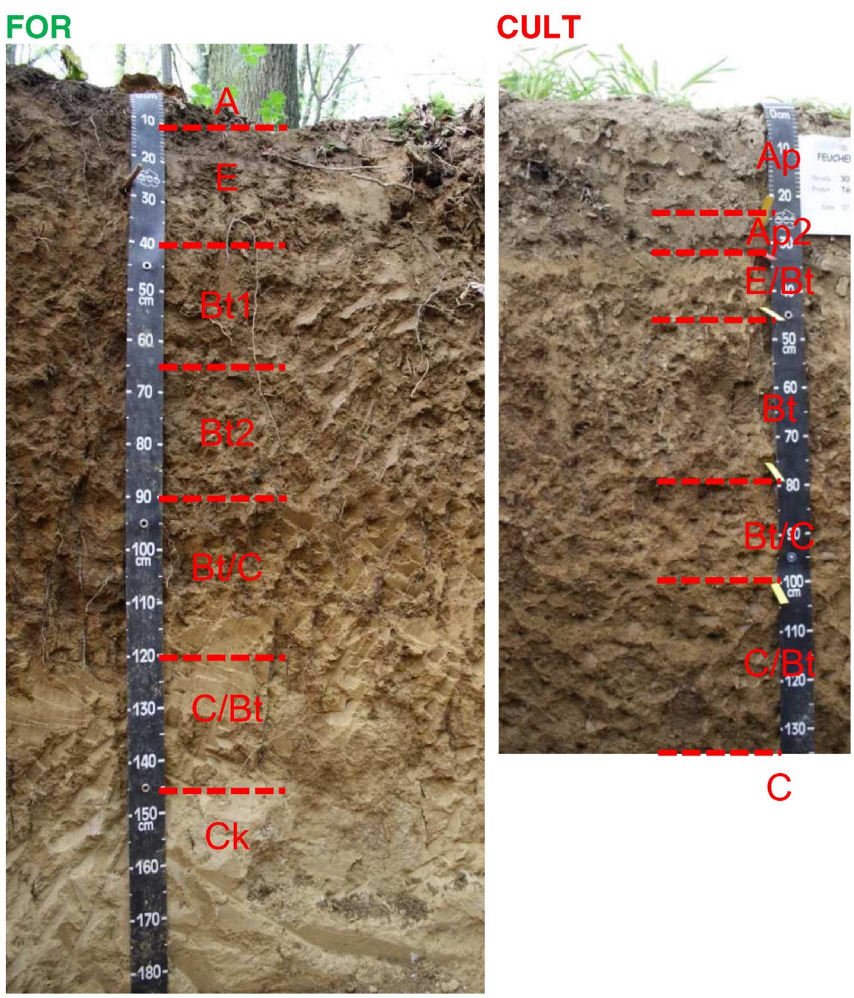

Fig. 1. Photographs of the Luvisols under study.

bulk densities were determined in triplicate using the cylinder method (500 cm^3) for each soil horizon.

After manual sorting of clay coatings found in FOR Bt-horizon (63–73 cm), their mean bulk density was determined by the kerosene method (Monnier et al., 1973) using 21 replicates. Aggregates that have been manually sorted for bulk density determination had a granulometric clay content of 311 $g.kg^{-1}$, whereas the corresponding horizon had a clay content of 217 $g.kg^{-1}$. This clay size fraction enrichment confirmed the fact that these aggregates were actually clay coatings.

Kubiena boxes of 150 × 80 × 50 mm were used to extract undisturbed soil samples. After each eluviated horizon was sampled, soil samples were taken at regular 10–20 cm intervals in the Bt-horizons in which clay coatings were supposed to be abundant (Fig. 2). Then we collected one box for each Bt/C horizon and C/Bt horizon, respectively, because clay coatings were hypothesized to be less abundant in these horizons. In the case of CULT, we extracted a supplementary box from the C-horizon at 1.5 m depth.

2.3. Micromorphological approach and quantification

Once we had taken vertically undisturbed soil blocks in both profiles, we oven-dried and impregnated them under vacuum with polyester resin. After polymerization, large thin sections (6 × 14 cm) were prepared following the procedures of Guilloré (1985). A micromorphological examination was done from 20× until 400× with a polarizing light microscope (Eclipse 50iPol, Nikon) and all thin sections were described according to the terminology of Bullock et al. (1985) and Stoops and Vepraskas (2003). Clay illuviation features such as clay coatings, clay infillings, clay intercalations and papules, i.e., fragments of clay coatings, were quantified by the same micromorphologist using a point-counting approach (Amonette, 1994; Murphy and Kemp, 1987). A mean of 480 points per thin section was counted at regular intervals of 4 mm. These parameters were selected in order to limit absolute counting error while avoiding excessive counting time (Grove and Jerram, 2011; Ulery and Drees, 2008; Van der Plas and Tobi, 1965). This approach enabled us to detect and quantify the frequency of the features of interest in each thin section, and to determine their areal proportions (Gargiulo et al., 2013; McKeague et al., 1980; Weibel et al., 1966). As a first approximation, areal proportions were used to estimate volumetric proportions, as proposed by Anderson and Binnie (1961) and FitzPatrick (1984). Point count errors were estimated by using Van der Plas and Tobi's (1965) chart, which is a standard tool to assess uncertainty in point-counting results (Howarth, 1998). We verified that the operator who did the determination tended to be consistent between point counts by recounting selected sets of thin sections (standard deviation of 3% maximum, Table A in Supplementary data file).

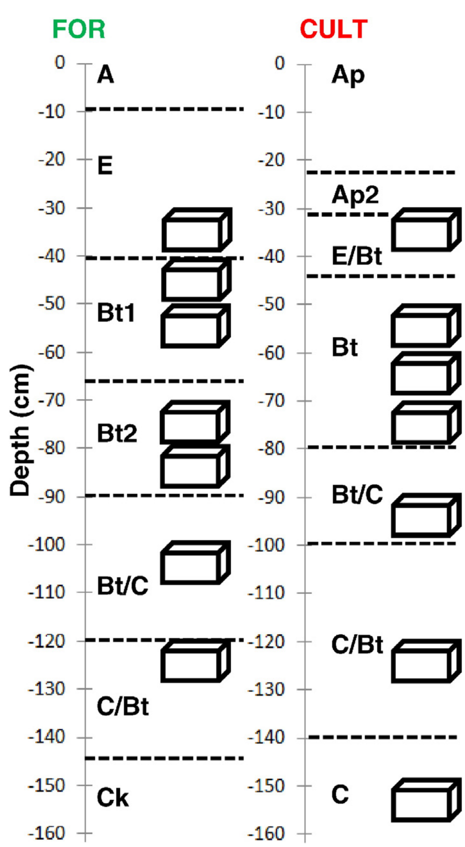

Fig. 2. Sampling procedure in FOR and CULT profiles.

2.4. Mass balance calculations

Mass balance is a classical way to estimate the amount of fine particles lost by the A/E horizons and gained by the Bt-horizons. Calculations were performed considering i) granulometric clay fraction as mobile element *j* and, ii) coarse granulometric fraction as invariant within soil profile (Agbenin and Felix-Henningsen, 2001; Nordt et al., 2004; Oh and Richter, 2005; Quénard et al., 2011). The coarse granulometric fraction was approximated using the coarse silt and sand content of each soil horizon.

The overall mass flux for any soil volume $m_{j,flux}$ in $g \cdot cm^{-2}$ was then calculated for each element *j* using the following equation proposed by Brimhall et al. (1991) and modified by Egli and Fitze (2000):

$$m_{j,flux} = \frac{\frac{1}{100} \times \rho_{ref} \times C_{j,ref} \times Th \times \tau_{j,w}}{1 + \varepsilon_{i,w}} \quad (1)$$

where the subscripts *ref* and *w* refer, respectively, to the soil volume taken as a reference and to the weathered product, $C_{j,ref.}$ is the concentration of *j* in the reference parent material in weight percent, *Th* is the thickness of the considered horizon, and ρ_{ref} is the bulk density of the reference material in $g \cdot cm^{-3}$. The two functions $\varepsilon_{i,w}$ and $\tau_{j,w}$ refer, respectively, to the strain and to open-system mass-transport. They were defined by Brimhall et al. (1991) to represent, for $\varepsilon_{i,w}$, the soil-volume change over time using an immobile element *i* and, for $\tau_{j,w}$, the mass fraction of element *j* gained or lost from the weathered product with respect to the mass originally present in the parent material. They were calculated according to the following equations:

$$\varepsilon_{i,w} = \frac{\rho_{ref} \times C_{i,ref}}{\rho_w \times C_{i,w}} - 1 \quad (2)$$

$$\tau_{j,w} = \frac{C_{j,w} \times C_{i,ref}}{C_{i,w} \times C_{j,ref}} - 1 \quad (3)$$

Volumetric strains and mass fluxes were calculated using the parent material characteristics based on the C-horizon characteristics of the CULT profile. Errors involved in the calculations were estimated according to the following rules:

$$\text{for } C = A + B \text{ or } C = A - B, \ \Delta C = \Delta A + \Delta B \quad (4)$$

$$\text{and for } C = A \times B \text{ or } C = \frac{A}{B}, \frac{\Delta C}{C} = \frac{\Delta A}{A} + \frac{\Delta B}{B} \quad (5)$$

with *A*, *B*, and *C* being variables and Δ*A*, Δ*B*, and Δ*C* the uncertainties related to *A*, *B*, and *C*. The analytical errors were set to 5% for the contents in the fine and coarse soil fraction and for the volume occupied by the different soil volumes. Errors related to bulk density measurements were determined thanks to their respective standard deviations.

2.5. Statistical analysis

The effect of land use change on illuviation was analyzed thanks to a second-order polynomial model taking into account the land use, the depth and the square depth. Such square depth model was chosen in order to represent the typical distribution with depth of the clay size fraction that increases with depth until it reaches a maximum in the Bt-horizons before decreasing towards the C-horizon. Denoting by *Yuk* the percentage of illuvial features in a thin section, the linear model (M1) is defined by:

$$\text{(M1)}\quad Y_{uk} = m + a_u + b \times depth_{uk} + c \times depth^2_{uk} + d_u \times depth_{uk} + f_u \times depth^2_{uk} + E_{uk}$$

where u stands for the land use, k the observation number, $depth_{uk}$ (respectively $depth^2{}_{uk}$) the depth (respectively the square depth) for observation k with land use u and E_{uk} independent identically distributed random normal variables.

The effect of land use u is captured by the coefficients a_u, d_u, f_u, therefore the model M1 is compared (thanks to a nested model test) to the simpler model M0:

$$\text{(M0)}\quad Yuk = m + b \times depth_{uk} + c \times depth^2_{uk} + E_{uk}$$

where no coefficient depends on the land use.

The integration of the parametric expression of the expected curve linking depth to the proportion of illuvial clay provides the area under the curve (AUC) given by: $(m+a_u)^*(dephthmax_u\text{-}depthmin_u) + (b+d_u)^*1/2^*(depthmax^2_u\text{-}depthmin^2_u) + (c+f_u)^*1/3^*(depthmax^3_u\text{-}depthmin^3_u)$, where $depthmax_u$ and $depthmin_u$ are respectively the maximal and minimal depths for land use u. Therefore, the difference between two AUC corresponding to different land uses u and v can be tested with the contrast: $(m+a_u)^*(dephthmax_u\text{-}depthmin_u)\text{-}(m+a_v)^*(dephthmax_v\text{-}depthmin_v) + (b+d_u)^*1/2^*(depthmax^2_u\text{-}depthmin^2_u)\text{-}(b+d_v)^*1/2^*(depthmax^2_v\text{-}depthmin^2_v) + (c+f_u)^*1/3^*(depthmax^3_u\text{-}depthmin^3_u)\text{-}(c+f_v)^*1/3^*(depthmax^3_v\text{-}depthmin^3_{v\cdot})$

According to standard linear regression theory, this contrast follows a Student distribution. All statistical analyses have been carried out with the R software (R Core Team, 2013).

3. Results and discussion

3.1. Macromorphological analysis and analytical measurements

The surface A-horizon of FOR is 10-cm thick (Fig. 1), whereas the Ap-horizon of CULT is 25-cm thick and is overlying a plow pan of several centimeters (Ap2). While the E-horizon is subangular blocky and 30-cm thick in the FOR profile, the CULT profile displays a 14-cm thick heterogeneous blocky E/Bt horizon.

Underneath, both profiles present a progressive development of the illuviated horizons (105-cm thick for FOR and 95-cm thick for CULT) including separate Bt/C and C/Bt illuviated horizons mixed with decarbonated loess. Clay coatings are visible to the naked eye until the C-horizon reached at 145 cm depth for the FOR profile and at 140 cm depth for the CULT profile. Redoxic features are detected along profiles especially in the CULT one undoubtedly due to a change in parent material at the bottom of profiles (clay with millstone at 160 cm depth) and to regular perched water table (Table 1).

Both soils are showing similar clay size fraction distribution with depth with i) an expected clay content increase in Bt-horizons and ii) a maximum clay content reached in the Bt/C horizons (Fig. 3 and Table 2). The maximum clay content of 314 $g \cdot kg^{-1}$ is observed at 90 cm depth in CULT whereas the maximum content of 320 $g \cdot kg^{-1}$ appears deeper (105 cm depth) in FOR. While FOR profile displays greater amounts of granulometric clay from 0 to 30 cm and from 95 to 140 cm depth, CULT profile shows higher clay contents than FOR from 30 to 95 cm.

The organic carbon content in the cultivated plot displays an homogeneous content of 9 $g \cdot kg^{-1}$ until 30 cm depth due to plowing homogenization whereas in the forested soil, it is mainly restricted to the A-horizon (0–10 cm) with a content of 30.5 $g \cdot kg^{-1}$. From 31 cm to the basis of both soil profiles, the organic carbon content stays beneath 5 $g \cdot kg^{-1}$ (Fig. 3 and Table 2). The pH in the forested soil ranges from 5.1 (E-horizon) to 8.6 (Ck-horizon) and from 6.6 (Ap-horizon) to 7.7 (C-horizon) under cultivation. The pH in FOR profile is theoretically more favorable for illuviation (particle mobilization) than under cultivation (Quénard et al., 2011; Van Breemen and Buurman, 2002).

3.2. Micromorphological analysis

3.2.1. General characteristics

While the organization between solid and pores is complex in both profiles (Table 3), porosity abundance tends to vary between 5 to 15% surface area (Table A in Supplementary data file). Porosity is mostly related to biological activity as it is made up essentially of channels, packing voids, and mammilated vughs (resulting from the loose packing of soil components such as excremental features). Excremental features such as rough ellipsoidal organo-mineral enchytraeids excrements or infillings with a bow-like fabric produced by earthworms are well established all along both profiles (Table 3).

3.2.2. Eluviated horizons

In FOR E-horizon and CULT E/Bt-horizon, large fabric units occur in a dense groundmass of smaller units with a close c/f related distribution pattern (Table 3). In case of cultivation, the b-fabric tends to be undifferentiated as tillage in case of cultivation and reworking through faunal activity under forest may have prevented particle orientation. The eluviated E-horizon of FOR profile displays some indications of past disturbance: a channel to massive microstructure at 30–35 cm depth overlying a complex microstructure (granular to vughy) at 35–40 cm depth with i) aboveground faunal activity features suggesting past clearance, and ii) subhorizontal planes undoubtedly due to compaction. In good agreement with historical records, these micromorphological data confirm that the soil under forest experienced a more complex land use history than initially thought including episodic cultivation.

3.2.3. Illuviated horizons

Fissure and prismatic microstructure made of physical planar voids can be regularly observed in subsurface illuviated horizons. Vughs are still abundant in the C-horizon of the CULT profile (150–160 cm). While single spaced porphyric c/f related distribution is more regular in these subsurface horizons, b-fabric tends to be more oriented with depth (stipple speckled or mosaic speckled to granostriated b-fabrics).

Different types of textural pedofeatures were identified in each horizon of both soils: limpid or dusty clay coatings or intercalations, papules, compound layered of silty clay coatings, unsorted or silt infillings, sedimentary crust fragments (Table 3). Clay translocation due to illuviation is responsible for the formation of clay coatings, i.e., well sorted and oriented clay bodies with sharp boundaries (Brewer, 1964; McCarthy et al., 1999; Ufnar, 2007). In the thin sections under study, two kinds of clay coatings are distinguished: i) yellow to yellow brown, limpid clay, often microlaminated ones, with sharp extinction lines and strong interference colors (Fig. 4), ii) brown to dark brown dusty clay, comprising small organic particles or microparticles of up to 5 μm in diameter, moderately to well sorted, laminated to layered with diffuse extinction lines (Fig. 5).

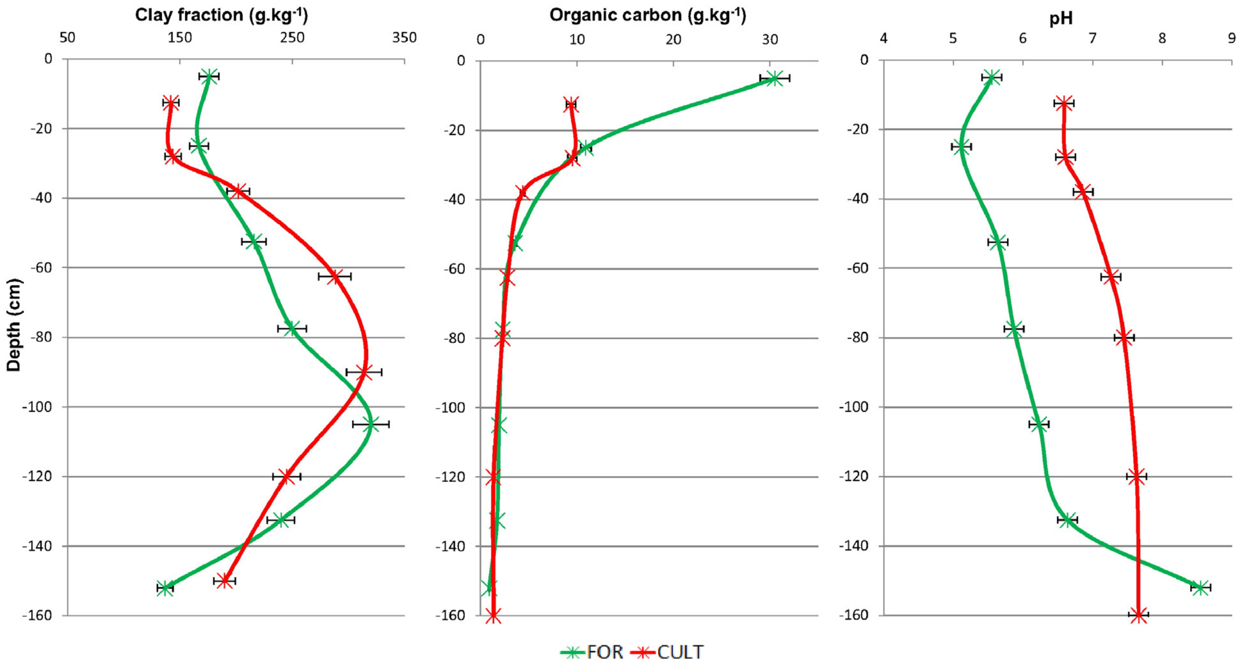

Fig. 3. Distribution with depth in FOR and CULT profiles of a/granulometric clay size fraction (g·kg^{-1}) b/organic carbon content (g·kg^{-1}) c/pH.

The first kind of clay coatings is typical of primary illuviation (Jamagne et al., 1987) and is associated with homogeneous and low energy stratified flow accumulations (Bullock et al., 1985; Fedoroff, 1997; Fedoroff and Courty, 1994; Kühn et al., 2010). The second type of clay coatings is similar to the organo-argillans described by Brewer (1964), to the clayey part of 'type b' coatings described by Jongmans et al.

Table 3
Soil micromorphology.

Horizon	Depth (cm)	Related distribution pattern[a]	Birefringence fabric[a]	Microstructure	Main pedofeatures	
					Textural	Other features
FOR						
E	30–40	Close-por	Undiff.	30–35: channel to massive 35–40: granular to vughy	Unsorted compound layered features	Fe–Mn nodules, excremental
Bt1	40–50	Close-por	Stipple-speckled	Granular to vughy	Dusty clay coatings and intercalations, thin limpid clay coatings, papules	Fe–Mn nodules, excremental
Bt1	50–60	Close-por	Stipple-speckled	Channel, fissure and vughy	Dusty and limpid clay coatings and infillings, papules	Fe–Mn nodules, excremental
Bt2	70–80	ss-/close-por	Granostriated/ mosaic speckled	Channel, fissure and vughy	Unsorted compound layered features, laminated dusty and microlaminated limpid clay coatings and infillings, papules	Fe–Mn nodules, passage
Bt2	80–90	ss-por	Granostriated/ mosaic speckled	Vughy, channel and prismatic	Unsorted compound layered features, silty limpid clay coatings, laminated dusty coatings and infillings	Fe–Mn nodules
Bt/C	100–110	ss-/close-por	Granostriated/ mosaic speckled	Prismatic, channel, fissure, vughy	Dusty and limpid clay coatings, papules	Passage, excremental
C/Bt	120–130	Close-por	Stipple-speckled	Channel, vughy, fissure, platy, vesicular	Unsorted compound layered features, dusty and limpid clay coatings, papules	Passage, excremental
CULT						
E/Bt	30–40	Close-por	Undiff.	Channel, fissure, prismatic, granular, subangular blocky	Sedimentary crust, unsorted compound layered features, papules	Excremental
Bt	50–60	Close-por	Stipple-speckled	Channel, granular to vughy, fissure	Dusty and limpid clay coatings, papules	Fe–Mn nodules, passage
Bt	60–70	ss-/close-por	Stipple-speckled	Fissure, prismatic, vughy to crumb, channel	Dusty and limpid clay coatings and infillings, papules	Fe–Mn nodules, excremental
Bt	70–80	ss-por	Granostriated/ mosaic speckled	Fissure to blocky, channel, granular, crack	Unsorted compound layered features, dusty and limpid clay coatings, papules	Fe–Mn nodules, passage
Bt/C	90–100	ss-/close-por	Stipple-speckled	Channel, fissure, blocky	Dusty and limpid clay coatings, papules	Fe–Mn nodules, excremental, passage
C/Bt	120–130	ss-/close-por	Stipple-speckled	Channel, blocky, platy	Unsorted compound layered features, dusty and limpid clay coatings, papules	Fe–Mn nodules, excremental, passage
C	150–160	Close-por	Stipple-speckled	Platy, vughy	Unsorted compound layered features, silty limpid clay coatings, dusty clay coatings	Fe–Mn nodules, excremental, passage

[a] Undiff = undifferentiated; ss = single-spaced; por = porphyric.

(1991) and to the organo-clay coatings observed by Miedema et al. (1999). They are also due to clay illuviation but as they may contain a low amount of silt-sized particles (Jamagne et al., 1987) and as they have a medium to poor orientation (Fedoroff, 1997), they are more related to successional patterns of deposition and concentration in soils affected by a change in internal drainage (Adderley et al., 2010). Even if it is still under discussion (Adderley et al., 2010; Goldberg and Macphail, 2006; Usai, 2001), dusty clay coatings are generally associated with cultivation (Brammer, 1971; Gebhardt, 1988) and agric horizons (Stoops and Vepraskas, 2003; WRB, 2014) whereas the limpid ones are related to unexposed soils with a permanent vegetation cover (Fedoroff, 1997).

Papules (i.e., fragments of clay coatings (Brewer, 1964)) are observed regularly all along both profiles (Fig. 6). As they are mostly located into biological infillings, we firmly believe that they are related to biological activity. While papules are detected in both cases from the E-horizon to the C/Bt horizon, limpid and dusty clay coatings are clearly identified from the top of both Bt-horizons and were still observable in the C-horizon of the CULT profile.

Thick (>250 μm width) and coarse-textured features with a mixture of clay, silt and organic matter are seen locally in both profiles but especially in the E/Bt horizon of the CULT one (Fig. 7). These unsorted or clayey silt compound layered features are similar to the "agricutans" defined by Jongerius (1970) and are classically associated with cultivation (Jongmans et al., 1991, 2001).

These qualitative results confirm previous macromorphological observations and granulometric data. They clearly suggest that illuviation has contributed to the textural contrast and to the specific clay size fraction distribution of both soil profiles. These micromorphological data provide important additional information regarding macromorphological description of both soils: i) clay coatings observed in the C-horizon of CULT profile were too scarce to be detected in the field, ii) papules detected all along both profiles were too small (<100 μm) to be directly visible to the naked eye, iii) the two main types of illuvial clay coatings (limpid and dusty ones) were not differentiable macromorphologically due to their fine textural characteristics. In light of these micromorphological descriptions, the illuviation process appears to have produced two different types of textural pedofeatures: the limpid and the dusty clay coatings, the latter being considered in the following as an indicator of cultivation.

3.3. Quantification of illuviation intensity and sensitivity to land use change

After having selected the diagnostic features associated with illuviation, it was possible to quantify them according to our point-counting approach. Results in Table 4, related to illuvial clay quantification (areal percentage in thin sections) suggest, as expected, that the abundance of illuvial clay features in E-horizons is low (<1%) and essentially made of papules. A great variability of illuvial clay abundances can be underlined in Bt-horizons of both profiles: from 0.7% in the top of the Bt-horizon of FOR profile (40–50 cm) to 10.2% in the bottom of the Bt-horizon of CULT profile (70–80 cm). Unexpectedly, both profiles reach a maximum of illuvial clay percentage in the Bt/C horizon with 14.0% for the FOR profile and 12.4% for the CULT profile but not in the Bt-horizon. Deeper, the abundances decrease but remain significant nonetheless since 2.3% of total illuvial clay is still present in the C-horizon of the CULT profile. The illuviated horizons under study display more than enough amounts of illuvial oriented clay to be classified as argic horizons according to WRB (2014), i.e., more than 1%. The clay illuviation indexes of 876%·cm and 862%·cm, in FOR and CULT profiles respectively (Table 4) are indicative of a 'very high' degree of illuviation according to Miedema and Slager (1972). The data obtained in both Bt/C horizons (14.0% and 12.4% in FOR and CULT respectively) are in accordance with the highest degree of illuviation recorded in the literature for subhorizons of the argillic horizon: 6.2 to 19.9% (Kühn et al., 2010). The degree of reworking is considered as 'weak' in the illuviated horizons of both our soils according to Miedema and Slager's (1972) classes. The results depicted in Fig. 8 suggest that illuvial clay accumulations are positively correlated with granulometric clay contents ($R^2 = 0.70$). These results clearly suggest that illuviation has been quantitatively involved in textural contrast and associated clay size fraction distribution with depth in both profiles.

Among the three quantified kinds of illuvial pedofeatures, the limpid clay coatings are the most abundant (Table 4). Dusty clay coatings represent around one third of total illuvial clay in both soil profiles (Table 4). Papules generally represent between 5 and 10% of total illuvial clay, except in the C/Bt-horizon of the FOR profile where papules are particularly abundant (Table 4). The recognition of dusty clay coatings in the FOR profile suggests that illuviation has not only occurred under a permanent forest vegetation cover but also during some periods of cultivation before the last afforestation at least one hundred years ago. Such a complex land use management history detected in our FOR site is in good agreement with other micromorphological indicators of past disturbances as discussed above (see Section 3.2.2) and is very common in this part of France where first local forest clearings for agro-pastoral purposes dated back from 5400 BP and were generalized before 4500 BP (Pastre et al., 2015). According to the highly significant abundance of dusty clay coatings in both soil profiles, the legacy of anthropic-driven illuviation over the past millennia is far from being negligible.

With a global mass flux of $15.1 \pm 5.7\ g \cdot cm^{-2}$ for the CULT profile and of $15.3 \pm 6.1\ g \cdot cm^{-2}$ for the FOR profile (Table 5), it appears that the last land-use change dated back from at least one hundred years for the FOR profile and two hundred years for the CULT profile did not induced a significant effect on total illuvial clay according to statistical

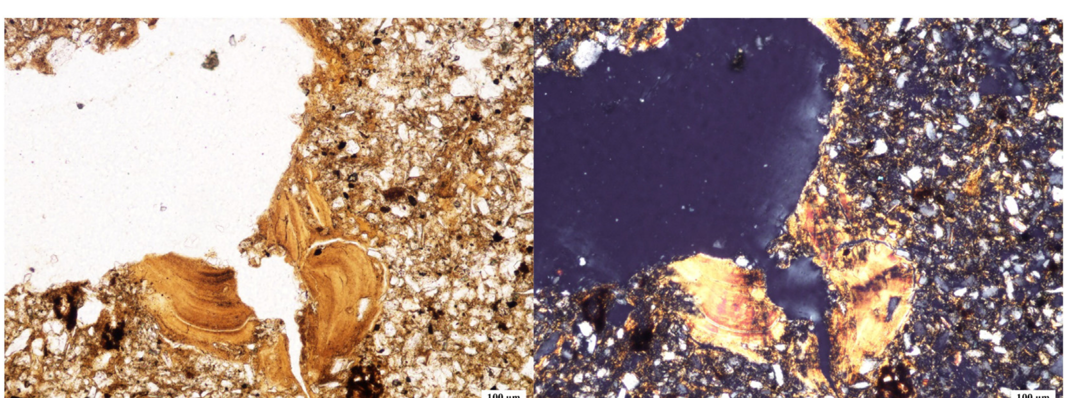

Fig. 4. Microlaminated limpid clay coating (FOR 70–80 cm) (left PPL; right XPL).

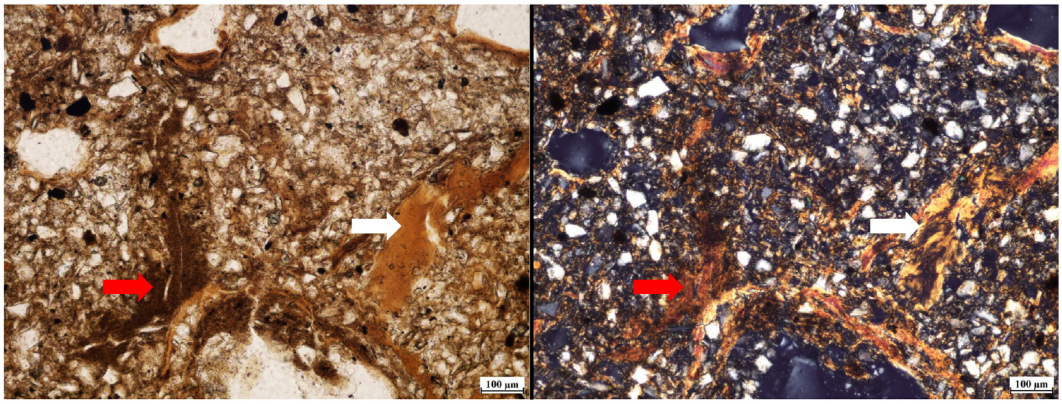

Fig. 5. Some dusty (red arrow) and limpid (white arrow) clay coatings and intercalations (FOR 80–90 cm) (left PPL; right XPL). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

results (p-value associated with Fisher test $Pr(>F) = 0.45$, Fig. 9). This result is clearly in accordance with the one obtained by Jagercikova et al. (2015) on the same soil profiles using a ^{10}Be isotopic approach and tend to confirm the results previously obtained by Slager and van de Wetering (1977). On the opposite, Thompson et al. (1990) have studied very similar soils and land uses and they observed that illuviation was more expressed in the cultivated soil than under forest. As the forest in their study has been probably established in the 17th century, their forested site experienced a longer period under forest than our FOR plot.

In addition to a higher amount of agricutans-like pedofeatures in the CULT profile than in the FOR profile (see paragraph 3.2.3), the CULT profile shows a slightly higher, but not statistically significant ($Pr(>F) = 0.14$), dusty clay illuviation index at the scale of the entire profile (Table 4). At the horizon scale, the abundance of dusty illuvial clay coatings is moreover twice higher in the Bt-horizon of the CULT profile than in the Bt-horizon of the FOR profile (Table 4). All these results are in agreement with Simpson (1997), who associated a greater frequency of coarser textural pedofeatures to more intense cultivation. These concordant results suggest i) a qualitative change in the nature of the translocated particles with coarser particles and more abundant organic rich particles being translocated in CULT than in FOR profile, and ii) a differentiation of the dusty clay illuviation index between the FOR and CULT profiles since the last shift in land use.

In good agreement with the general framework of Richter (2007), two telescoping time scales of anthropedogenesis may be distinguished: i) the legacy of human history over the past millennia with more or less episodic periods of cultivation resulting in the formation of significant amount of dusty clay coatings in both soil profiles, and ii) active human impacts since the last land-use change that mainly induced a qualitative change in the nature of the translocated particles in the direction of more abundant dusty and coarse-textured features.

3.4. Relative contribution of illuviation to the genesis of textural contrast

CULT C-horizon (with a bulk density of $1.54\ g\cdot cm^{-3}$) contains a considerable amount of illuvial clay, i.e., $26\ g\cdot kg^{-1}$ based on the estimated bulk density of $1.75\ g\cdot cm^{-3}$ for clay coatings. According to these observations and measurements, a correction of the granulometric characteristics of the CULT C-horizon was carried out. Using this new virtual reference horizon allowed us to do adequate mass balance calculations regarding a nonilluviated C-horizon.

As mass balance calculations were done regarding the corrected C-horizon of CULT profile, we can note that, as expected, the soil-volume change over time $\varepsilon_{i,w}$ of the overlying horizon (C/Bt horizon) is low (Table 6). The FOR A-horizon displays the highest $\varepsilon_{i,w}$ related to its high content of organic matter. Bt-horizons of both profiles have intermediate values of $\varepsilon_{i,w}$ due to moderate expansion related to clay accumulation in these illuviated horizons. While A and E-horizons have very low to negative mass flux values, Bt, Bt/C and C/Bt horizons have positive mass flux values as high as $8.4 \pm 3.5\ g\cdot cm^{-2}$ in the Bt-horizon of the CULT profile. These results clearly confirm the occurrence of clay accumulation in subsurface horizons.

As gains are markedly larger than losses, it can be suggested that additional processes were involved in addition to illuviation. In order to assess the relative contribution of illuviation, present mass balance calculations were compared with illuvial clay mass flux calculations

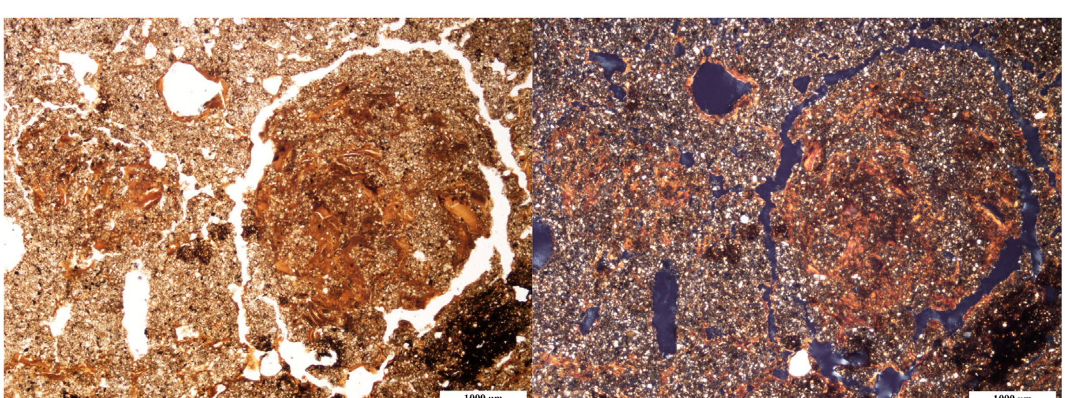

Fig. 6. Papules corresponding to fragments of clay coatings in faunal infillings (FOR 90–100 cm) (left PPL; right XPL).

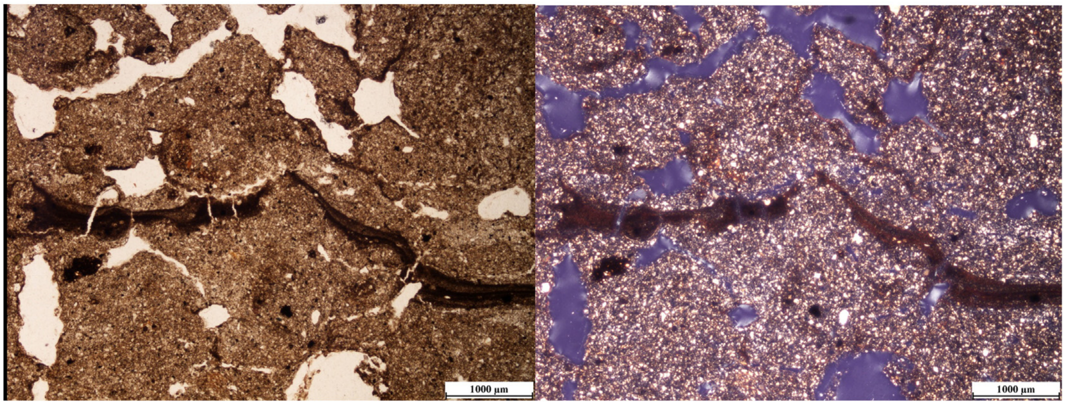

Fig. 7. Layered silty clay intercalation: agricutan-like feature (CULT 30–40 cm) (left PPL; right XPL).

presented in Table 5. Three cases may be identified: i) mass balance flux is lower than illuviation mass flux in FOR E-horizon, ii) mass balance fluxes are higher than illuviation mass fluxes in all Bt-horizons and, iii) mass balance flux values are close to illuviation mass flux values in the Bt/C and C/Bt horizons of both profiles.

Mass balance flux is negligible in FOR E-horizon whereas its illuviation mass flux obtained thanks to the micromorphological approach is equal to $0.5 \pm 0.6\ g\cdot cm^{-2}$. This low gain in clay size fraction, partly due to the presence of papules, is undetectable by the mass balance approach as it is counterbalanced by a higher loss in clay size fraction thanks to eluviation. Quantification of the cumulative effect of these opposite phenomena shows that mass balance fluxes are lower than mass fluxes, as determined with the micromorphological approach, and finally lead to an underestimation of the intensity of the illuviation process.

Bt-horizons display higher clay size fraction gains with the mass balance approach than with the micromorphological quantification (Tables 5 and 6). Several complementary hypotheses could explain such a difference. According to the description of loess deposits, the first loess deposit, in which Bt-horizons formed, is slightly finer than the third deposit corresponding to the present C-horizon. It has resulted in an overestimation of the ratio between the contents of the coarse silt and sand fraction between the C-horizon taken as reference and the Bt-horizons, and finally in overestimations of the ε (Eq. (2)) and τ (Eq. (3)) functions. Contrastingly, the micromorphological quantification is not sensitive to such lithological discontinuities. In addition, as soon as clay neoformation or microdivision is involved, some gains in clay size fraction in Bt-horizons unrelated to the illuviation process may be quantified by the mass balance approach but not by the micromorphological approach. Finally, illuvial features could have been integrated in the soil groundmass through biological or physical reworking and reorganization processes (Schuylenborgh et al., 1970; Ufnar, 2007) and consequently may have induced an underestimation of illuvial fluxes by the micromorphological approach.

Contrastingly, the Bt/C and C/Bt horizons show mass balance fluxes very close to their illuviation mass fluxes (Tables 5 and 6) as well as a similar or a higher intensity of biological reworking than the Bt-horizons as measured by the relative abundance of papules (Table 4). Such results suggest that i) in the Bt/C and C/Bt horizons, lithological discontinuity, clay neoformation and reorganization processes did not interfere with illuviation that was the main active process, ii) the integration of illuvial features through biological reworking did not induced significant underestimations of the micromorphological fluxes and may consequently be neglected at least in the studied soils, and iii) the clay coatings were particularly stable over time in these two horizons. Finally, at least in soils with a 'weak' reworking intensity, the micromorphological quantification of illuvial features provide a more accurate quantification of the intensity of the illuviation process than a mass balance approach particularly sensitive to lithological discontinuities as well as to competing (E-horizon) or synergic (Bt-horizons) processes.

Whatever the occurrence of soil processes like neoformation, microdivision or lithological discontinuities in some of the different soil horizons, the total mass flux obtained through micromorphological

Table 4
Clay coatings and illuviation features amounts (surface percentage) in the different horizons of the FOR and CULT profiles. Absolute counting error with a 95% confidence was determined according to Van der Plas and Tobi (1965). Illuviation indexes (in %·cm) were calculated according to Miedema and Slager (1972).

FOR					CULT				
Depth (cm)	Limpid (%)	Dusty (%)	Papules (%)	Total (%)	Depth (cm)	Limpid (%)	Dusty (%)	Papules (%)	Total (%)
E									
30–40	0 ± 1%	0.5 ± 1%	0.5 ± 1%	1.0 ± 1%	30–40	0 ± 1%	0 ± 1%	0.3 ± 1%	0.3 ± 1%
Bt									
40–50	0.7 ± 1%	0 ± 1%	0 ± 1%	0.7% ± 1%	50–60	1.3 ± 1%	1.0 ± 1%	0.6 ± 1%	2.9 ± 1%
50–60	2.0 ± 1%	2.2 ± 1%	0.7 ± 1%	4.9 ± 2%	60–70	5.1 ± 2%	4.0 ± 2%	0.2 ± 1%	9.3 ± 3%
70–80	4.9 ± 2%	2.0 ± 1%	0.2 ± 1%	7.1 ± 2%	70–80	5.4 ± 2%	4.7 ± 2%	0.1 ± 1%	10.2 ± 3%
80–90	7.7 ± 2%	0.5 ± 1%	0 ± 1%	8.2% ± 2%					
Bt/C									
100–110	11.0 ± 3%	2.3 ± 1%	0.7 ± 1%	14.0 ± 3%	90–100	8.4 ± 2%	3.2 ± 2%	0.9 ± 1%	12.4 ± 3%
C/Bt									
120–130	2.8 ± 1%	2.3 ± 1%	1.6 ± 1%	6.7 ± 2%	120–130	6.4 ± 2%	1.7 ± 1%	0.6 ± 1%	8.7 ± 2%
C									
					150–160	1.6 ± 1%	0.7 ± 1%	0 ± 1%	2.3% ± 1%
Illuviation index (%.cm)									
	590	202	85	876		560	245	57	862

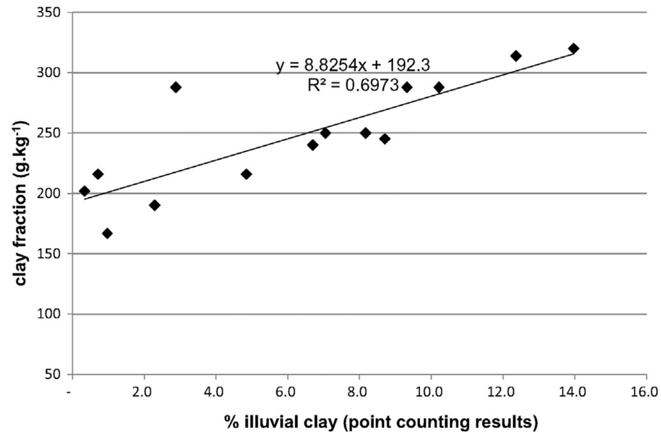

Fig. 8. Correlation between illuvial clay from point-counting results (areal percentage) and granulometric clay size fraction ($g \cdot kg^{-1}$) for all studied thin sections.

quantification represents between 75% in the CULT profile and 86% in the FOR profile of the gains calculated thanks to the mass balance approach. Even if the micromorphological point-counting quantification can lead to an overestimation of the illuvial clay amounts partly due to edge effect (Murphy and Kemp, 1984), illuviation is thus by far the most important process in the genesis of the textural contrast in the soils under study.

4. Conclusion

To quantify illuviation matter fluxes separately from all other soil processes implying clay size fraction, the method proposed here relies on an exhaustive quantification of clay coatings abundance at a microscopic scale and the determination of their bulk density after selective manual sampling. In addition to being in very good agreement with more classical macromorphological descriptions and bulk analyses, demonstrating that its results can be interpreted with a good level of confidence, this method is not invalidated by lithological discontinuities or by the presence of other accessory soil processes like clay neoformation or microdivision, frequently encountered under the field conditions. The micromorphological quantification of illuvial clay coatings is a promising way to improve the accuracy of the quantification of illuviation intensity and consequently to provide new insight on the process of illuviation and on its sensitivity to land use change.

Table 5
Illuvial clay mass flux in FOR and CULT profiles (clay coatings bulk density = 1.75 ± 0.12 $g \cdot cm^{-3}$).

Horizon	Thin sections used	Horizon thickness (cm)	% Illuvial clay	Flux ($g \cdot cm^{-2}$)
FOR				
E	30–40	30	1.0	0.5 ± 0.6
Bt1	40–50	25	2.8	1.2 ± 0.6
	50–60			
Bt2	70–80	25	7.6	3.3 ± 1.3
	80–90			
Bt/C	100–110	30	14.0	7.3 ± 2.5
C/Bt	120–130	25	6.7	2.9 ± 1.2
Sum (profile)				15.3 ± 6.1
CULT				
E/Bt	30–40	14	0.3	0.1 ± 0.3
Bt	50–60	35	7.5	4.6 ± 1.8
	60–70			
	70–80			
Bt/C	90–100	20	12.4	4.3 ± 1.6
C/Bt	120–130	40	8.7	6.1 ± 2.1
Sum (profile)				15.1 ± 5.7

As a first example, the micromorphological quantification of clay coatings result in demonstrating that illuviation was responsible for 75% and 86% of the textural contrast in the cultivated and forested soils, respectively. Illuviation was thus by far the dominant factor causing textural differentiation in the two Luvisols under study, even if accessory processes involving the clay size fraction were also at play, such as lithological discontinuity, clay neoformation, or biological and physical reworking.

In addition, the quantification of high amounts of dusty clay coatings in the soil profile under forest for at least the last hundred years clearly underlines the sensitivity of illuviation to past periods of cultivation and the legacy of anthropedogenesis on the present soil properties. Similarly, the recognition of "agricutans" and the slight increase of dusty clay coatings abundance in the cultivated soil in comparison with the forested one, taken as a reference, point out the fact that illuviation is strongly sensitive and reacts rapidly, at least qualitatively, to changes in soil properties impacted by land-use management such as soil cover, soil water dynamics, soil pH or soil organic matter content, to name just a few.

All these results strongly support the conclusions that illuviation is active in soils under present climatic conditions and must no more be considered as a fossil process. More work is needed, however, to better constrain the temporal dynamics of the illuviation process and its sensitivity to key soil properties.

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.geoderma.2015.11.035.

Acknowledgments

This research was conducted in the framework of the Agriped project (ANR-10-BLANC-605), which was supported by the French National Research Agency (ANR). The authors are grateful to the Agriped team for contributing to the sampling and to INRA Grignon for providing access to their long-term experimental site and its associated data. We would also like to thank Hervé Gaillard for providing soil bulk density data, Christophe Labat for helping with impregnation and thin section preparation and Philippe Baveye for providing editorial comments on an early draft of this article.

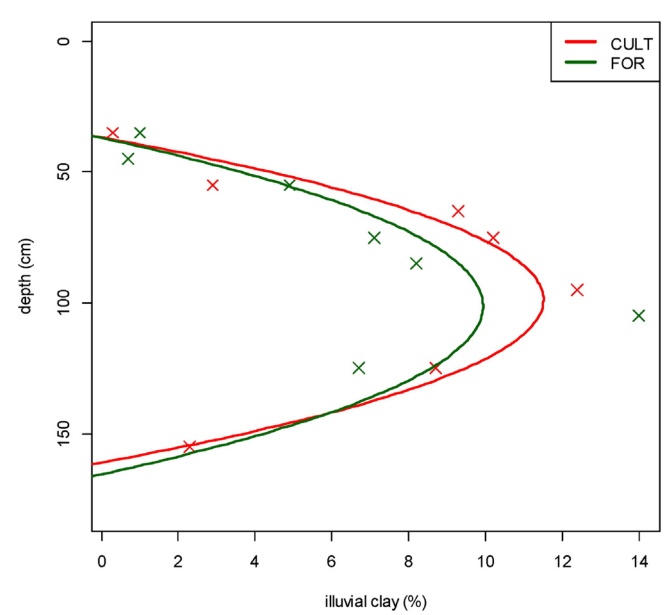

Fig. 9. Statistical modeling of illuvial clay distribution with depth in CULT and FOR profiles.

Table 6
Final mass balance calculations in FOR and CULT profiles.

Horizon	Depth (cm)	Thickness (cm)	εHorCcult	ζj.HorCcult	Flux ($g\cdot cm^{-2}$)
FOR					
A	0–10	10	0.6	0.1	–
E	10–40	30	0.2	0.0	–
Bt1	40–65	25	0.1	0.4	2.5 ± 1.0
Bt2	65–90	25	0.2	0.8	4.2 ± 1.8
Bt/C	90–120	30	0.2	1.2	8.0 ± 3.4
C/Bt	120–145	25	0.1	0.5	2.9 ± 1.2
Gains sum					*17.6* ± 7.5
CULT					
Ap	0–25	25	0.1	−0.2	−0.9 ± 0.4
A2	25–31	6	0.1	−0.1	−0.2 ± 0.1
E/Bt	31–45	14	0.2	0.3	1.1 ± 0.4
Bt	45–80	35	0.3	1.2	8.4 ± 3.5
Bt/C	80–100	20	0.2	1.2	5.2 ± 2.2
C/Bt	100–140	40	0.1	0.6	5.5 ± 2.3
C	>140	10	0.0	0.0	0.0 ± 0.0
Gains sum					*20.1* + 8.4

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