Geoderma 406 (2022) 115529

Contents lists available at ScienceDirect

Geoderma

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

Soil organic carbon saturation in cropland-grassland systems: Storage potential and soil quality

Thomas Guillaume [a], David Makowski [b], Zamir Libohova [c], Luca Bragazza [a], Fatbardh Sallaku [d], Sokrat Sinaj [a,*]

[a] Agroscope, Field-Crop Systems and Plant Nutrition, Research Division Plant Production Systems, Route de Duillier 50, P.O. Box 1012, CH-1260 Nyon, Switzerland
[b] INRAE, AgroParisTech, Université Paris-Saclay, UMR MIA 518, 75231 Paris, France
[c] USDA-ARS Dale Bumpers Small Farms Research Center, 6883 South State Hwy 23, Booneville, AR 72927, USA
[d] Agriculture University of Tirana, Rruga Paisi Vodica 1025, Tirana, Albania

ARTICLE INFO

Handling Editor: Ingrid Kögel-Knabner

Keywords:
Mineral-associated organic matter
Fine particulate organic matter
Soil fractions
Carbon deficit
Switzerland

ABSTRACT

Reliable estimations of soil organic carbon (SOC) deficits in agroecosystems are crucial in evaluating the atmospheric C sequestration potential of agricultural soils and supporting management decisions. Nonetheless, the co-benefit on soil quality resulting from SOC accrual is rarely considered. Here, we assessed SOC saturation and soil physical quality in permanent grasslands (PG) and croplands (CR) by applying the C-saturation concept and the SOC:clay ratio as an indicator of soil physical quality to a set of long-term monitoring sites in western Switzerland. For this goal, we produced a new relationship between the silt + clay (SC) particles and the C stored in the mineral-associated fraction ($MAOM_C$) and we assessed the assumption that grasslands can be used as C-saturated reference sites. The saturation in PG was not coincidental as it depended on the C accrual history. Hence, PG with the lowest $MAOM_C$ have not reached their C-saturation level and present a potential SOC storage under optimal management. The $MAOM_C$ saturation in CR was low (62 ± 4%) and corresponded to a deficit of −8.8 ± 1.2 mg C g^{-1} soil as compared to the current level in PG. The saturation was mainly affected by the proportion of temporary grassland in the crop rotation. The relative distribution of C between MAOM (~80%) and the fine and coarse particulate organic matter (POM) was not affected by land-use types. The $MAOM_C$ saturation in this study ($MAOM_C = 0.372 \times SC + 4.23$) was similar to that reported in the litterature, but discrepancies appeared when the silt and clay contents were considered separately. SC was by far the main factor explaining $MAOM_C$ amount in PG (semi-partial R^2: 0.66). In contrast to other studies, the C content of MAOM in PG (43 mg C g^{-1} SC) was not related to the SC content, suggesting a fixed maximal value in C-saturated soils. Nonetheless, $MAOM_C$ saturation may be underestimated as the least saturated PG might still accumulate $MAOM_C$. Finally, the SOC:clay ratio was correlated with $MAOM_C$ saturation level in CR, but not in PG suggesting that targeting SOC accrual in CR optimizes the benefits between soil C storage and soil quality.

1. Introduction

As most cropland soils are strongly depleted in soil organic carbon (SOC) (Sanderman et al., 2017), increasing SOC in these soils is especially relevant when addressing, simultaneously, several global challenges, such as climate change mitigation and adaptation as well as food security (Chenu et al., 2019).

To estimate the carbon (C) storage potential of agricultural soils, it is necessary to determine the maximum attainable SOC level in these soils. The C-saturation concept defines an upper limit of SOC that is not subject to decomposition due to mineral protection (Schmidt et al., 2011), a limit that is not dependent on climate or land-use type, but only on soil physicochemical characteristics (Stewart et al., 2007). It is commonly accepted that once the mineral fraction is saturated, additional SOC accrual may occur only for poorly protected SOC fractions with a fast turnover rate (Six et al., 2002). It has been recently shown that the maximum levels of organic C in the mineral-associated organic matter fraction ($MAOM_C$; < 50 μm) in grasslands and forests across Europe from

* Corresponding author at: Field-Crop Systems and Plant Nutrition, Research Division Plant Production Systems, Route de Duillier 50, P.O. Box 1012, CH-1260 Nyon, Switzerland.
E-mail address: sokrat.sinaj@agroscope.admin.ch (S. Sinaj).

https://doi.org/10.1016/j.geoderma.2021.115529
Received 18 August 2021; Received in revised form 5 October 2021; Accepted 6 October 2021
Available online 18 October 2021
0016-7061/

the Land Use and Coverage Area frame Survey (LUCAS) is about 50 mg C g^{-1} soil, while the amount of organic C in the particulate organic matter fraction (POM_C; 50–2000 µm) continues to increase with increasing bulk SOC content (Cotrufo et al., 2019). Nonetheless, achieving the theoretical maximum $MAOM_C$ level (i.e. $MAOM_C$ saturation) is not realistic everywhere as climatic constraints are difficult to overcome and not all land-use types are interchangeable. Thus, an "effective" $MAOM_C$-saturation level can be determined for a specific pedoclimatic and land-use type combination that corresponds to the level at which an increase of soil C input does not lead additional $MAOM_C$ accrual (Stewart et al., 2007).

Grasslands have higher SOC contents than croplands (Guillaume et al., 2021) and both croplands and forests are depleted in $MAOM_C$ compared to grasslands (Cotrufo et al., 2019; Poeplau and Don, 2013; Wiesmeier et al., 2014b). As grasslands, which experience high soil C inputs and little soil disturbance, are expected to be saturated in $MAOM_C$, soil measurements collected on this land-use type have been used as a reference in determining a relationship for $MAOM_C$ saturation as a function of the proportion of silt + clay (SC) particles < 20 µm (Hassink, 1997). This relationship has been widely adopted to estimate the impact of land-use type and management on SOC deficit (Carter et al., 2003; Gregorich et al., 2009; Liang et al., 2009; Sparrow et al., 2006; Zhao et al., 2006) as well as the SOC storage potential in different countries (Angers et al., 2011; Chen et al., 2019; McNally et al., 2017; Wiesmeier et al., 2015, 2014a). The relationship between $MAOM_C$ and the mass proportion of silt + clay particles was initially established by linear regression on a wide range of soils from temperate and tropical climates (Hassink, 1997). However, Six et al. (2002) showed that this relationship was not universal and was influenced by the dominant clay type and the upper limit of the particle-size fraction (<20 vs. < 50 µm). More recently, few studies attempted to develop new relationships adapted to local conditions. For example, two studies were conducted in natural grasslands located in a dry and cold area in northern China to derive new equations that showed lower saturation capacity (Liang et al., 2009; Wiesmeier et al., 2015). Another study, which was conducted in a cool and humid area of Canada, found a relationship similar to the one proposed by Hassink (1997) (Carter et al., 2003). Furthermore, it was argued that the use of least squares regression was underestimating the level at which $MAOM_C$ saturates as the methodology implies that about half of the data points that fall above the regression line are apparently oversaturated (i.e. positive residuals) while the other half below the regression line are apparently under-saturated (i.e. negative residuals) (Beare et al., 2014; Feng et al., 2013). Thus, a boundary line analysis using a top quantile of the data points (e.g. 10 %) was subsequently proposed and used in several studies to investigate the relationship between silt + clay and $MAOM_C$ fractions (Baldock et al., 2019; Beare et al., 2014; Cai et al., 2021; Fujisaki et al., 2018).

The maximum attainable SOC storage potential is, however, a target that is practically difficult to achieve (Amundson and Biardeau, 2018). Besides important socio-economic constraints, changing the steady-state status of SOC stocks for a specific land-use type requires either an increase of C inputs to the soil or a decrease of SOC mineralization resulting from a change in management or land use (Chenu et al., 2019; Wiesmeier et al., 2020). Hence, the availability of organic amendments and the management of crop residues are major factors determining the maximum attainable SOC storage capacity for a cropping system (Blanchet et al., 2016; Koishi et al., 2020; Maltas et al., 2013). Similarly, the trade-offs between C sequestration and the resulting economic and environmental costs associated with nutrient sequestration may drive down the targeted SOC levels (Lugato et al., 2018; Van Groenigen et al., 2017).

The role of SOC on soil quality and functioning is unanimously acknowledged and used repeatedly along with C sequestration to justify research on soil organic C accrual (Baveye et al., 2020; Bossio et al., 2020; Rumpel et al., 2020). While it is implicitly assumed that targets defined for C storage using, for example, the C-saturation concept are also adequate and applicable to reach optimum soil quality, no study to our knowledge has evaluated the relationships and trade-offs between both in order to address the question of whether simultaneously attaining SOC saturation and soil quality targets is realistic. In theory, the aim of sequestrating C in soil should first promote large SOC pools with slow turnover rates, such as the $MOAM_C$ fractions. Nevertheless, small labile SOC pools, such as POM_C in croplands and grasslands, are also important in maintaining the quality of soil physical, chemical and biological characteristics (Duval et al., 2018; Haynes, 2005). It has been shown that some important soil properties related to soil physical quality (e.g. clay dispersability, matrix porosity and bulk density) were more related to the bulk SOC-to-clay ratio (SOC:clay) than to the bulk SOC alone (Dexter et al., 2008). A SOC:clay threshold of 1:10 was identified in several studies from different European countries and, based on independent methodologies, determined to be critical for maintaining soil physical quality (Dexter et al., 2008; Jensen et al., 2019; Johannes et al., 2017a; Prout et al., 2020). One of these studies conducted on grasslands and croplands found that below the SOC:clay threshold of 1:10 the macroporosity (>150 µm) is likely strongly reduced and the risk of structure collapse increases due to a loss of hydrostructural stability, making the soil more prone to mechanical stress (Johannes et al., 2017a; Johannes et al., 2017b).

Based on the above considerations, the main goals of this study were: (i) to assess the C deficit in the cropland-grassland system of western Switzerland using Hassink's approach and determine the factors affecting the level of $MAOM_C$ saturation in croplands; (ii) to investigate the relationship between the level of $MAOM_C$ saturation and soil physical quality here indicated by the SOC:clay ratio; and (iii) to assess the potential limitations in Hassink's approach to determine the maximum capacity of soil C storage. In particular, we investigated the assumption of $MAOM_C$ saturation in grasslands, the particle-size limit used in the methodology and the impact of factors related to site conditions, other than texture, on MAOMc levels in grasslands.

2. Materials and methods

2.1. Study sites

The study was carried out in the Canton of Fribourg (Switzerland) using long-term soil monitoring sites of the FRIBO network established by the Agricultural Institute of the Fribourg Canton (Guillaume et al., 2021; Levasseur et al., 2019). The Fribourg Canton (167,000 ha) is located north of the Alps in the western part of Switzerland (46° − 47°N, 7°E). It lies on the Swiss Midland between the Jura Mountains' piedmont (NW) and the Western Alps' foothills (SE), representing a NW-SE gradient of soils, elevation and climate. The Midland elevation gradually increases from 429 m a.s.l. in the NW to about 900 m a.s.l, where steeper slopes occur in the Alps' foothills (<2389 m a.s.l.). The geology is composed of Tertiary molasse (sandstone, marl) partly covered by glacier moraine deposits. The lowest part of the NW area is mostly covered by lake and alluvial sediments. This part is relatively flat, while the rest of the Midland presents a smooth, hilly topography. Soils are dominantly Cambisols, Gleysols and Fluvisols (Frau et al., 2020). The climate is temperate continental (MAT: 8.9 °C; MAP: 1075 mm) in the Midland, but for every increase of 100 m elevation the amount of precipitation increases by about 80 mm and temperature decreases by about 0.5 °C (Dumas, 2013; Sevruk, 1997). Agricultural land (AL) occupies about 75,516 ha, with 67 % under permanent (PG) and temporary (TG) grasslands and 30 % under annual crops, half of which are cereals and the rest mostly maize, rapeseed, potatoes and sugar-beets (DIAF, 2019). Temporary grasslands, according to Swiss legislation (OTerm 910.91), are areas that are designated mainly for forage production and occasionally for grazing for ≤ 6 years as part of crop rotations.

The FRIBO network is composed of 250 sites, which were established in 1987 and based on a 2 × 2 km grid (Julien and Morand, 1995). The number of sites is intentionally biased towards croplands since one of the major focuses of the network is to monitor soil degradation. Out of the 184 well-drained cropland and grassland sites (Guillaume et al., 2021), SOC fraction analysis was conducted on 58 well-drained (Cambisols or Fluvisols) sites that were purposely selected to represent the diversity of land-use trajectories and site conditions (Table S1). The current data set includes 24 sites under croplands (CR) and 24 sites under permanent grasslands (PG) during the whole 30-year monitoring period, as well as 10 sites that experienced one to several land-use changes (LUC) between CR and PG. One CR site with extreme cPOM value (13 mg C g^{-1} soil representing 64% of bulk SOC) was kept only for analysis on MAOM. Over the 30-year monitoring period the main crop for each year was recorded at each site. Sites that have never been cropped for the last 30-year period were classified as PG. PG are used for grazing or forage production. The most intensive managed grasslands are cut up to five times per year to produce forage and are fertilized with manure and slurry (Huguenin-Elie et al., 2017). Sites that remained under crop rotation during the 30-year period were classified as CR. Most CR sites included TG in the crop rotation, and the proportion of years with TG (*Grass*) or with cereals (*Cer*) over the 30-year period varied between CR sites. Sites with TG > 6 consecutive years were considered to have experienced LUC, even if they were subsequently converted to croplands. Similarly, PG that were cultivated for at least 1 year were considered as sites that experienced LUC.

2.2. Soil analysis

Each FRIBO site consists of a 10 × 10 m plot that is sampled every 5 years since the beginning of the monitoring in 1987. Soil samples (0–20 cm) were collected in 2016 (the 6th sampling cycle) according to the FRIBO methodology (Guillaume et al., 2021). Each sample is a composite sample from 25 sampling points collected on a 2 × 2 m grid. Thirteen of the 24 PG sites and 3 of the 10 LUC sites were additionally sampled in 2020 using the same methodology to increase the number SOC size fraction measurement. Soil size fractions were isolated to standard procedures (NF ISO and NF X methods). More specifically, three size fractions of organic matter, namely the coarse particulate organic matter (cPOM, 2000–200 μm), the fine particulate organic matter (fPOM, 200–50 μm) and the mineral-associated organic matter (MAOM, < 50 μm), were separated by sieving after shaking 50 g of soil for 16 h in 180 ml of water with 20 glass beads to break the aggregates (NF X 31 516). Each isolated SOC size fraction as well as bulk soil were analyzed for organic C content (NF ISO 14235, sulfochromic oxydation) and total nitrogen (NF ISO 13878, elemental analyzer). The particle size-analysis was done using the pipette method and the proportions of clay, silt and sand are reported as mass proportion of total particules mass. The mass proportion of clay determined in the particule size-analysis was used along with the mass proportion of SOC in bulk soil to calculate the SOC:clay ratio. The rate of SOC changes in the last 30 years and soil property data used for the multiple regression analysis [pH H_2O, ammonium acetate EDTA exchangeable potassium (K_{AAE}) and magnesium (Mg_{AAE}), available phosphorous (P_{NaHCO3})] correspond to a subset of the data previously published by Guillaume et al. (2021) and are presented in Table S1.

2.3. Calculation of $MAOM_C$ saturation

Mineral-associated organic matter C saturation was determined using Hassink (1997) methodology. Permanent grasslands (n = 24) were considered as saturated and used to establish the relationship between $MAOM_C$ at saturation and the proportion of silt + clay particles (sum of silt and clay proportions determined in the particule-size analysis) using a linear least squares regression.

The level of $MAOM_C$ saturation at each site was calculated by dividing the measured $MAOM_C$ with the expected amount of $MAOM_C$ at saturation for similar silt + clay proportion (SC) determined for FRIBO using permanent grassland sites. The C deficit in croplands corresponds to the difference between the $MAOM_C$ expected at saturation with the current level of $MAOM_C$. The same approach was used for nitrogen (N). When specified, the $MAOM_C$ saturation according to Hassink was calculated using the silt + clay content from the FRIBO sites and the parameters published in Hassink (1997); i.e. MAOMC saturated = 0.370 × SC + 4.09 . Finally, we also tested the approach based on quantile regression on PG sites instead of least squares regression to determine $MAOM_C$ saturation (see *Statistical analysis* section).

In order to compare the amount of $MAOM_C$ and the relation with silt + clay found for PG in our study region with grasslands from a broader area, we used $MAOM_C$ data published by Cotrufo et al. (2019) for the Land Use/Cover Area frame Survey (LUCAS) across Europe. In their study, the $MAOM_C$ values for about 10′000 forest and grassland sites had been estimated from the prediction based on a random forest model fitted with $MAOM_C$ data measured from a subset of sites (n = 186). For the comparison, we selected grassland sites (land cover = E20) from LUCAS that were within the range of pedoclimatic conditions found in our study, i.e. mean annual temperature of 6.5–10 °C, annual precipitation of 1000–1400 mm, and pH of 5.2 – 7.3. In total, 13 sites with measured $MAOM_C$ values and 285 sites with estimated $MAOM_C$ values from LUCAS met the criteria.

2.4. Statistical analysis

All statistical analyses were performed using R software 3.6.1 (R Core Team, 2020). Effects of land-use type and co-variables such as silt + clay (% of mass) on SOM fractions were assessed by ANCOVA models (function *lm*). If the interaction between explanatory variables was significant, differences in the effects of silt + clay on SOM fractions between land-use types were assessed by the function *emtrends* (*emmeans* package). If not significant, the interaction terms were removed from the model and differences between the three lands uses were assessed by the function *emmeans* (*emmeans* package). Variance partitioning between land-use type and the co-variable silt + clay was assessed by partial eta-squared (η^2) computed using the *etasq* function (*heplots* package). The effect of land-use type on $MAOM_C$-saturation level (Fig. 2) was assessed by ANOVA (function *aov*). The performance of models including silt and clay separately or together was evaluated based on Akaike information criterion (AIC). Significance of AIC difference between models was determined with the function *anova*. ANCOVA and ANOVA model assumptions were tested by the Shapiro and Bartlett tests, and data were log-transformed if necessary. A quantile regression was performed to determine the relationship between $MAOM_C$ and silt + clay for the top quartile in permanent grasslands with the function *rq* of the *quantreg* package (Koenker, 2017).

The importance of pedoclimatic factors on the amount of $MAOM_C$ in PG and the saturation level in CR was determined by multiple linear regressions with stepwise selection (function *stepAIC*), and the semi-partial coefficient of determination was calculated with standardized variables (function *r2beta*). Only variables with a Pearson correlation coefficient < 0.70 were used in the multiple regression to limit collinearity effects. The model with data from LUCAS in 2009 (dataset ID 35277), as well as the associated R script and data produced by Cotrufo et al. (2019), were provided by the European Soil Data Center. In order to compare the relationship between $MAOM_C$ and silt + clay for our study region with the one for similar grasslands across Europe, the average response of $MAOM_C$ to silt + clay content (Fig. S1) was computed from a partial dependence plot (PDP) of the random forest model developed by Cotrufo et al. (2019). This model computes $MAOM_C$ as a function of silt + clay content, pH, total SOC, total soil N, N deposition, MAT, land cover, and extractable K. The PDP reported in Fig. S1 describes the marginal effects of silt + clay content on the $MAOM_C$ values predicted by the random forest model. It is determined by averaging the model outputs over all model inputs except silt + clay content. If not specified, data are presented as mean ± standard error

(SE) and discussed differences are significant at p-value < 0.05.

3. Results

3.1. Land-use impact on C and N pools

The proportion of silt + clay particles (SC) and the land-use types were good predictors (R^2_{adj}: 0.81, n = 58) of the amount of organic C associated with silt + clay particles ($MAOM_C$; mg C g^{-1} soil) (Fig. 1a). The slope was steeper in PG [$MAOM_C = 0.372\ (\pm 0.087) \times SC + 4.23\ (\pm 3.89)$] compared to CR [$MAOM_C = 0.199\ (\pm 0.054) \times SC + 4.073\ (\pm 2.48)$], but the interaction between land use and silt + clay content was not significant (F = 1.99, p = 0.147). Based on the difference between intercepts and regardless of mineral particle content, CR and LUC were depleted by −8.76 ± 1.22 and −5.34 ± 1.40 mg C g^{-1} soil, respectively.

The average organic C content of silt + clay particles ($MAOM_C$ content; mg C g^{-1} SC) was affected by land-use type (F = 29.4, eta^2 = 0.53), but not by silt + clay content (F = 0.002, p = 0.96, eta^2 < 0.01) and there was no interaction between land-use and silt + clay content variables (F = 0.28, p = 0.76, eta^2 = 0.01) (Table 1). The mean $MAOM_C$ content for PG sites was 43 mg C g^{-1} SC with a standard deviation (SD) of 7 mg C g^{-1} SC and a maximum of 54 mg C g^{-1} SC (Fig. 1b). C:N ratios of MAOM ($MAOM_{CN}$ mean = 8.1, SD = 0.7) were not affected either by silt + clay content (p = 0.21) or land-use type (p = 0.34) (Fig. 1b).

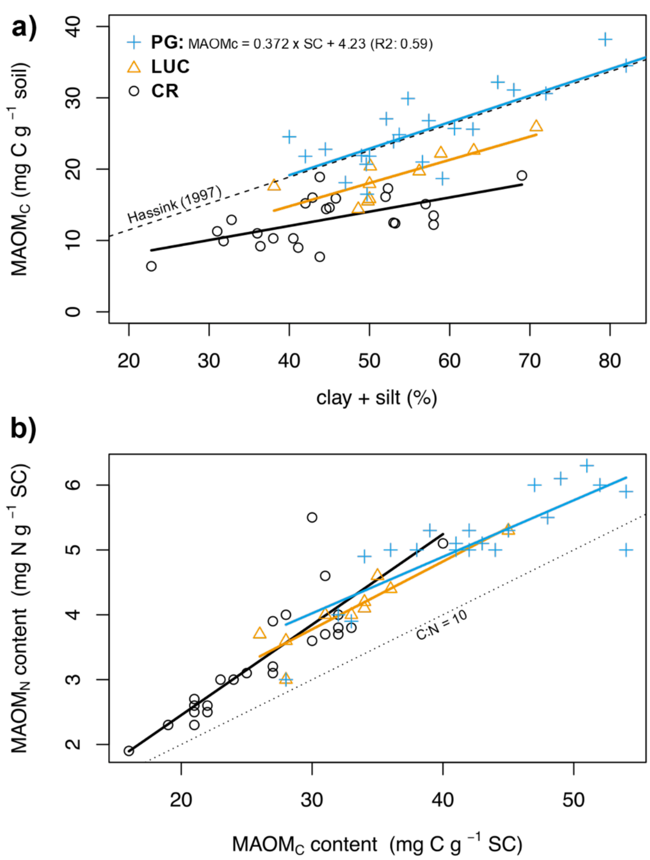

Fig. 1. Effect of land-use type on the relationships between (a) mineral-associated organic C ($MAOM_C$) and the mass proportion of silt + clay particles (<50 μm, SC) and between (b) the contents of organic C ($MAOM_C$) and total N ($MAOM_N$) in the silt + clay fraction. Solid lines represent the regression for permanent grasslands (PG), croplands (CR) and sites having experienced at least one land-use change in the last 30 years (LUC). The dashed line in (a) represents the relationship for permanent grassland found by Hassink (1997), whereas the dashed line in (b) represents the carbon to nitrogen (C:N) ratio = 10 of MAOC.

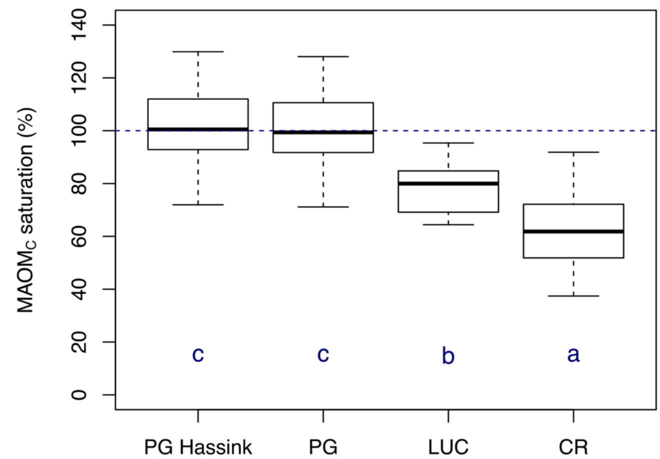

Fig. 2. $MAOM_C$ saturation in croplands (CR) and land-use change sites (LUC) using $MAOM_C$ in permanent grasslands (PG) as reference level for saturated soils following the equation determined in this study: $MAOM_C$ saturated = 0.372 × % SC + 4.23 (Fig. 1). PG Hassink represents the saturation level of PG calculated using the silt + clay proportion of PG and the relationship between the proportion of silt + clay particles and $MAOM_C$ determined by Hassink (1997): $MAOM_C$ saturated = 0.370 × SC + 4.09. Letters represent significant differences at p < 0.05 modeled by ANOVA.

Most SOC was concentrated in the $MAOM_C$ (<50 μm, 66–87 %), followed by the fine particulate organic matter fraction ($fPOM_C$, 50–200 μm, 8–26 %) and the coarse particulate organic matter fraction ($cPOM_C$, 200–2000 μm, 3–10 %) (Table 1). $MAOM_C$ (r = 0.98) and $fPOM_C$ (r = 0.84) were strongly correlated with bulk SOC and with each other (r = 0.73). The correlation between $cPOM_C$ and other fractions was weaker as correlation coefficients decreased to 0.64 for bulk SOC, 0.62 for $fPOM_C$ and 0.56 for $MAOM_C$.

The land-use type had no effect on the relative distribution of organic C in the three size-fractions (p > 0.24, Table 1), but the proportion of SOC in MAOM slightly increased with increasing proportion of silt + clay particles (0.09 ± 0.04% per % SC) while the proportion of SOC in cPOM decreased (-0.09 ± 0.02% per % SC). In fact, $cPOM_C$ did not increase with increasing silt + clay content as was the case for the other two finer fractions (Table 1).

The C:N ratio decreased in the finer fractions (Table 1). Within each fraction, C:N ratios were not affected by land-use type nor by silt + clay content, except for the fPOM where the corresponding C:N ratio was higher in CR than in PG and LUC. Hence, C and N were similarly affected by land-use type and silt + clay content in all fractions. The only exception was that fPOM in CR was more depleted in N than other fractions.

3.2. Mineral-associated organic C saturation

The $MAOM_C$ levels in PG corresponded to the levels expected for saturated soil (101 ± 3%) according to Hassink (1997). Using the $MAOM_C$ observed in PG as reference for saturated soils, the level of $MAOM_C$ saturation in LUC (79 ± 4%) and CR (62 ± 4%) indicated a strong deficit in $MAOM_C$ (Fig. 2).

The relationship between $MAOM_N$ and silt + clay content determined in PG [$MAOM_N$ (mg g^{-1}) = 0.037 (±0.006) × SC + 0.983 (±0.387)] was slightly weaker (R^2_{adj} = 0.60) than for $MAOM_C$. Although the slope was similar to the one found by Hassink's model ($MAOM_N$ = 0.037 × SC + 0.40), the intercept was higher, indicating an over-saturation in $MAOM_N$ (123 ± 4 %) for PG.

The model selected by multiple regression with stepwise selection explained 64% of the variation in saturation levels observed in CR (Table 2). Only two factors, namely the proportion of temporary grasslands in the crop rotation (*Grass*) and the exchangeable K (K_{AAE}) had a

Table 1
Effect of land-use type and mass proportion of silt + clay particles on SOM fractions in permanent grasslands (PG), croplands (CR) and land-use changes (LUC) sites.

	Land-use type (mean ± SE)			Variables			
	PG	CR	LUC	Land use	Silt + clay	Interaction	R^2_{adj}
SOC†	34.0 ± 1.8c	14.4 ± 0.8 a	25.0 ± 1.8b	***¥	+ ***	n.s.	0.78
$MAOM_C$ content$	42.5 ± 1.4c	26.5 ± 1.2 a	33.0 ± 1.7b	***	n.s.	n.s.	0.56
$MAOM_C$†	26.6 ± 1.5c	13.3 ± 0.6 a	19.7 ± 1.1b	***	+ ***	n.s.	0.81
$fPOM_C$†	5.3 ± 0.6b	2.2 ± 0.2 a	3.5 ± 0.4 ab	***	+ m.s.	n.s.	0.46
$cPOM_C$†	2.2 ± 0.1b	1.2 ± 0.1 a	1.8 ± 0.2b	***	- m.s.	n.s.	0.42
$MAOM_C$ rel‡	78.3 ± 1.1	79.5 ± 0.7	78.8 ± 1.6	n.s.	+ **	n.s.	0.10
$fPOM_C$ rel‡	15.0 ± 1.1	13.3 ± 0.5	13.8 ± 1.3	n.s.	n.s.	n.s.	−0.05
$cPOM_C$ rel‡	6.7 ± 0.4	7.3 ± 0.4	7.3 ± 0.7	n.s.	-***	n.s.	0.38
$MAOM_{CN}$	8.3 ± 0.1	8.1 ± 0.2	8.0 ± 0.1	n.s.	n.s.	n.s.	−0.02
$fPOM_{CN}$	11.4 ± 0.3 a	13.2 ± 0.3b	11.9 ± 0.4 a	***	n.s.	n.s.	0.29
$cPOM_{CN}$	17.6 ± 0.6	18.3 ± 0.5	18.0 ± 0.8	n.s.	n.s.	n.s.	0.01

† mg C g^{-1} soil.
$ mg C g^{-1} SC.
‡ % of total C.
¥ Significance levels (p-values): n.s. > 0.10, m.s. > 0.05, * < 0.05, ** < 0.01 and *** < 0.001. The direction of the relationship for silt + clay (mass proportion) is indicated by + and – signs.

Table 2
Variables influencing the saturation level of organic C in MAOM fraction in croplands (CR) and the amount of $MAOM_C$ in permanent grasslands (PG). Models selected by multiple regressions with stepwise selection of the following variables: *ΔSOC*: rate of SOC changes in the last 30 years, *pH* in water, K_{AAE}: exchangeable K, Mg_{AAE}: exchangeable Mg, P_{NaHCO3}: available P, P_{tot}: total P, *elevation, slope, Grass* and *Cer*: proportion of temporary grasslands or cereals in the crop rotation (only for CR), *clay*: clay particles content (only for CR), *silt + clay*: Mass proportion of silt + clay particles (only for PG).

Response variables	Explanatory variables	R^2	Estimates†	p-values
Saturation level in CR (n = 24)	*Model*	*0.64*	*54 ± 26*	*<0.001*
	Grass (%)	0.55	0.41 ± 0.09	<0.001
	K_{AAE} (g kg^{-1})	0.30	75 ± 26	<0.05
	P_{tot} (g kg^{-1})	0.23	28 ± 12	<0.05
	Mg_{AAE} (g kg^{-1})	0.19	−64 ± 31	<0.10
	- Elevation/slope/clay/ΔSOC/Cer/pH_{H2O}/P_{NaHCO3}: removed during the stepwise regression			
$MAOM_C$ in PG (n = 24)	*Model*	*0.80*	*− 47 ± 19*	*< 0.001*
	Silt + clay (%)	0.66	0.36 ± 0.06	< 0.001
	pH_{H2O}	0.22	5.5 ± 2.4	< 0.05
	ΔSOC (mg g^{-1} yr^{-1})	0.21	11 ± 5	< 0.05
	Elevation (m a.s.l.)	0.18	0.02 ± 0.01	< 0.10
	-P_{NaHCO3}/P_{TOT}/K_{AAE}/Mg_{AAE}/slope: removed during the stepwise regression			

† Estimates (mean ± SE) correspond to the slope for variables and to the intercept for the model.

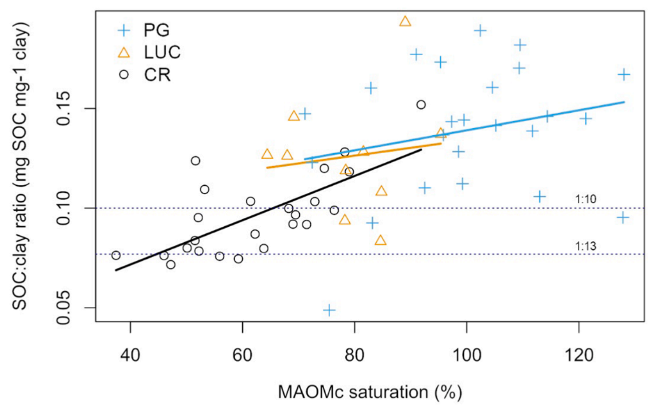

Fig. 3. Relationship between soil physical quality defined as the SOC:clay ratio and organic C saturation in the MAOM fraction in permanent grassland (PG), land-use change (LUC) and cropland (CR) sites. The thresholds for good (ratio > 0.10) and poor (ratio < 0.077) soil physical quality as defined by Johannes et al. (2019) are indicated by horizontal dotted lines. The PG site with SOC:clay ratio < 0.05 and high clay content is represented in the figure, but it is excluded from the ANCOVA analysis (n = 57).

significant effect on the saturation levels in CR but P_{tot} and Mg_{AAE} were also kept in the model by the stepwise selection. *Grass* had the highest semi-partial R^2 (0.55), i.e. almost doubled as compared to K_{AAE}. Factors related to other site pedoclimatic conditions (clay, elevation, pH and slope) or history (*Cer* and *ΔSOC*) were not selected in the model.

3.3. Influence of C-saturation level on soil physical quality

The SOC:clay ratio was < 1:10 in 62 % of the CR sites, but only 2 sites in LUC and 3 in PG were below this threshold (Fig. 3). Twenty percent of CR sites were even < 1:13. The strength of the relationship between organic C saturation in the MAOM fraction and the SOC:clay ratio varied depending on the land-use type, and the interaction between the two variables was significant (Fig. 3). For this analysis the data were log-transformed to meet the normal distribution criteria for ANCOVA, and one PG site with high clay content (61 %) and low SOC:clay ratio (<0.05) was considered as an outlier and excluded. The slope for CR (0.011 ± 0.003) was higher compared to PG (0.001 ± 0.003), but not compared to LUC (0.002 ± 0.006). The confidence interval (CI) for the PG and LUC slopes included zero but the CR slope was significantly different from zero ($F = 2.55$, $p < 0.04$, CI: 0.005–0.017). The SOC:clay ratio was negatively correlated with $MAOM_C$ content for PG (Fig. 4). For the PG sites, the SOC:clay ratio was positively correlated only with the proportion of $cPOM_C$ in bulk SOC. By contrast, the apparent $MAOM_C$ saturation was negatively correlated with the proportion of $cPOM_C$ and positively correlated with the proportion of $fPOM_C$ (Fig. 4).

The apparent $MAOM_C$ saturation in PG sites was not correlated with the SOC:clay ratio. One oversaturated site (>100 %) was below the optimum SOC:clay ratio (0.10), while several unsaturated sites were above this optimum. In CR sites, the relationship between saturation and SOC:clay ratio [SOC:clay = 0.0011 (±0.0002) × $MAOM_C$ saturation + 0.028 (±0.016)] indicated that a saturation of 65 % is required to reach a SOC:clay of 1:10. Nonetheless, a site with an apparente saturation as low as 52 % had already reached this level, i.e. the minimum threshold for good soil structure (Fig. 3).

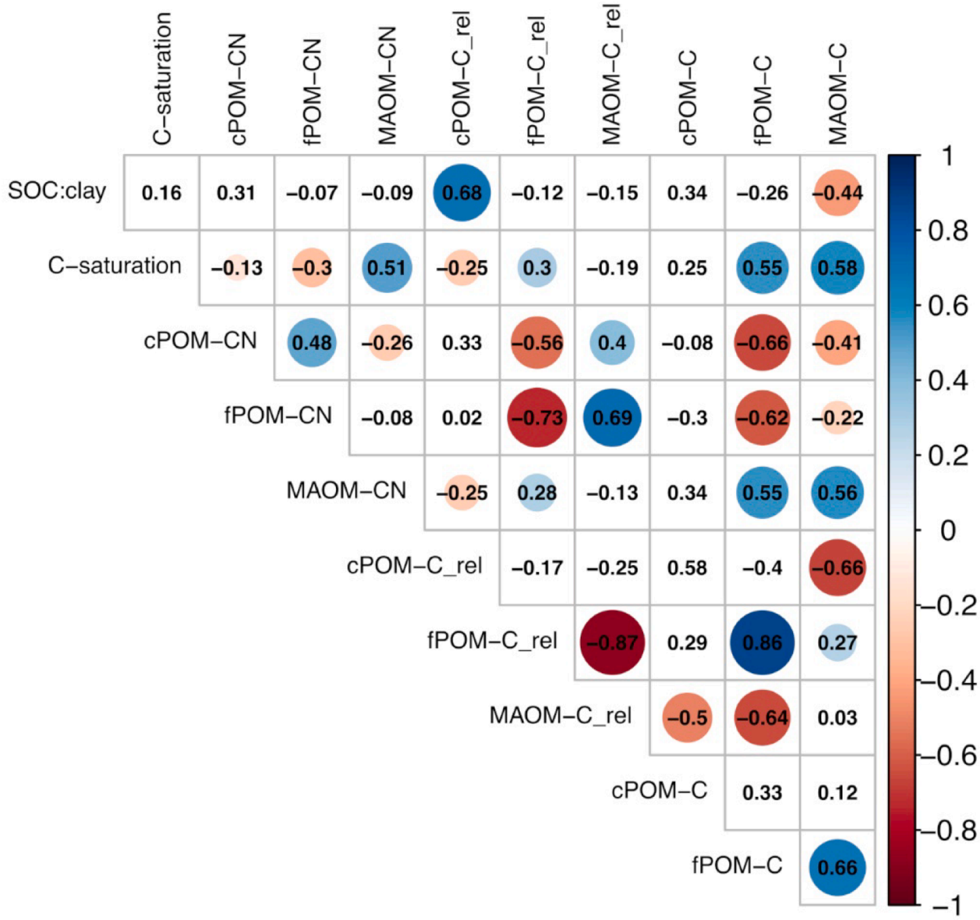

Fig. 4. Pearson correlations for permanent grasslands (PG) between SOC:clay ratio, apparent deviation of MAOMc from saturation (C-saturation) and different parameters (–CN═C:N mass ratio, -C_rel = proportion of bulk SOC in the fraction [%], -C = amount of SOC in the fraction [mg C g^{-1} soil]) in the coarse (cPOM; > 200 μm) and fine (fPOM; < 200 and > 50 μm) POM fractions as well as in the MOAM fraction (<50 μm). Colored circles indicate significant correlations at $p < 0.05$.

3.4. *Mineral-associated organic C in grasslands*

The silt + clay content was by far the factor that explained the most variation of $MAOM_C$ amount in PG (Table 2). The semi-partial R^2 for silt + clay (0.66) was at least three times higher than the ones for the two other significant variables in the model, namely pH_{H2O} (0.22) and ΔSOC (0.21). The factor *Elevation* was kept in the model but was not significant. Together these variables were sufficient to explain 80 % of the variation on $MAOM_C$ in PG. The relationship between silt + clay and $MAOM_C$ in the multiple regression model that included site pedoclimatic condition and the variable related to the land-use history (slope = 0.36, semi-partial $R^2 = 0.55$) was similar to the relationship that included only silt + clay (slope = 0.37, $R^2 = 0.59$); i.e. corresponding to Hassink's approach.

Models using Hassink's approach but relating $MAOM_C$ to different size particle fractions such as clay alone or silt and clay were compared. The relationship between $MAOM_C$ and silt and clay fractions included separately in the same model was: $MAOM_C = 0.440\ (\pm 0.073) \times \text{clay} - 0.131\ (\pm 0.158) \times \text{silt} + 10.49\ (\pm 5.31)$. Nonetheless, only the coefficient for clay content was significant ($p < 0.001$) while the intercept was marginally significant ($p = 0.06$). Although the performance of the model was slightly higher ($R^2_{adj} = 0.62$; AIC = 144) compared to the one using Hassink's approach ($R^2_{adj} = 0.59$; AIC = 145), the performance of both models did not improve significantly ($F = 2.72$, $p = 0.11$). Finally, the model with clay as unique explanatory variable [$MAOM_C = 0.451\ (\pm 0.071) \times \text{clay} + 14.51\ (\pm 2.11)$] had a similar performance ($R^2_{adj} = 0.62$; AIC = 143) to the model including clay and silt fractions separately ($F = 0.68$, $p = 0.42$).

The range of clay content in PG was wider (16–61 %) than the range of silt content (23–44 %, Fig. 5, Table S1). When silt accounted for a high proportion of the silt + clay particles, Hassink's equation tended to overestimate $MAOM_C$ for the FRIBO sites (Fig. 5a). When compared with data from Cotrufo et al. (2019) for LUCAS grasslands with pedoclimatic conditions similar to FRIBO, the measured values of $MAOM_C$ from LUCAS (LUCAS_measured) were close to the ones expected at FRIBO sites with similar silt and clay content. Nonetheless, at LUCAS sites with modeled values (LUCAS_estimated) of $MAOM_C$, the $MAOM_C$ was much higher for low silt + clay content than what would be expected for similar soils in FRIBO.

Similarly, $MAOM_C$ content (mg C g^{-1} SC) for LUCAS_measured sites was higher at low silt + clay content compared to FRIBO (Fig. 5b). In FRIBO, the relationship between $MAOM_C$ content and the silt and clay fractions included separately followed the equation: $MAOM_C$ content = $0.15\ (\pm 0.11) \times \text{clay} - 0.35\ (\pm 0.24) \times \text{silt} + 5.00\ (\pm 0.81)$. None of the particle-size fractions tested separately in the same model ($F = 1.65$, $p = 0.22$), together as silt + clay ($F = 0.19$, $p = 0.66$), or with only clay ($F = 1.14$, $p = 0.30$) had significant effects on the $MAOM_C$ content. The highest $MAOM_C$ content in PG was 54 mg C g^{-1} SC.

Finally, the relationship between $MAOM_C$ (mg C g^{-1} soil) and silt + clay determined from a quantile regression on the quartile of PG sites having the highest $MAOM_C$ as a function of their silt + clay content [$MAOM_C = 0.441\ (\pm 0.114) \times SC + 3.12\ (\pm 6.45)$] showed a slope that was 19% steeper than the common approach using least squares regression including all PG sites.

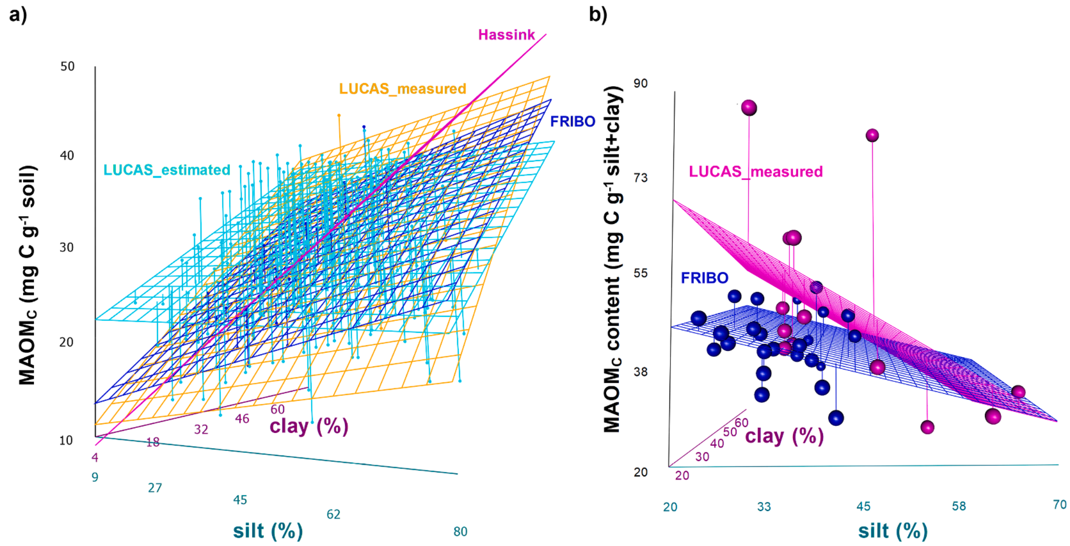

Fig. 5. Three-dimensional plots showing (a) $MAOM_C$ and (b) $MAOM_C$ content in permanent grasslands from various sources as a function of silt and clay contents. Data comes from the FRIBO monitoring network (n = 24), from comparable grasslands in LUCAS with measured (n = 13) or estimated (n = 285, only in Fig. 5a) values, and, for (a), from saturation levels expected in FRIBO sites as calculated using Hassink's equation. The grid represents the regression plan for the respective data sources. Note that Hassink's regression plan in Fig. 5a appears as a line because of the orientation of the 3D view.

4. Discussion

4.1. Impact of land use on SOM fractions and C saturation

The C storage potential of cropland soils was considerable, varying from 2 to 14 mg C g^{-1} soil in the top 20 cm of the studied cropland sites, corresponding to a current average level of $MAOM_C$ saturation of 62%. The relative C deficit was not influenced by soil texture as indicated by the non-selection of *clay* in the multiple regression model (Table 2). This means that a larger absolute C amount could potentially be sequestered in soils with finer texture.

The proportion of temporary grasslands within the crop rotation was the main factor affecting the level of $MAOM_C$ saturation in croplands, explaining 55% of the variation even after the inclusion of all other factors. The importance of including temporary grasslands in farm management to increase bulk SOC has previously been highlighted (Guillaume et al., 2021). It demonstrates that farmer's choices about their crop rotation directly affect soil C storage (Table 2). The other factors explaining the saturation levels in croplands (K_{AAE} and P_{tot}) were related to the nutrient status of the soil. Higher SOC at sites with higher chemical fertility may be a consequence of the positive effect of fertilization on biomass production, resulting in higher soil C inputs that is not fully compensated by an increase of SOC mineralization (Rüegg et al., 2019).

The importance of nutrient limitations on the level of $MAOM_C$ saturation in croplands may also be highlighted by the stronger depletion of the N pool in the fPOM fraction of croplands, leading to a higher $fPOM_{CN}$ than in other land-use type (Table 1). Moreover, in permanent grasslands, high N contents in the fPOM fraction (i.e. low $fPOM_{CN}$) were associated with high $MAOM_C$ (Fig. 4). This fraction, and especially its heavy component (>1.7 g cm^{-3} following a density fractionation), has already been identified by Samson et al. (2020) as the most responsive fraction to agronomic pratices and it is expected to represent a crucial step during the SOC stabilization process. Our findings suggest that this pool might serve as N mining source for soil microorganisms in N-limited condition. The fPOM isolated with the fractionation scheme of our study is likely composed of fragmented organic residues free or coated with fine mineral particles because micro-aggregates were disrupted with glass beads (Maillard et al., 2015). Nonetheless, a detailed characterization of this fraction is necessary to further investigate the mechanisms and the consequences of higher N depletion in that pool.

Overall, the findings of the present study, in line with Jilling et al. (2020), show that the type of rotation does not only affect organic matter pools with rapid turnover such as cPOM and fPOM, but it has also a rapid and extensive impact on $MAOM_C$: a C pool with an allegedly slow turnover rate due to mineral protection (Lavallee et al., 2019).

Finally, contrary to other studies (Barré et al., 2017; Cotrufo et al., 2019), the proportion of organic C in different fractions was relatively similar for all land-use types. A similar observation was made in allophanic and non-allophanic soils in New Zeland (McNally et al., 2017). This indicates that, within one land-use type or one system with frequent conversion (cropland-grassland), the POM fraction was also constrained to a maximum level. This challenges the concept that substantial additional C sequestration can be achieved via POM fractions, once $MAOM_C$ saturation is reached, without a drastic change in land-use type (e.g. a change from agricultural land to forest) (Cotrufo et al., 2019). Furthermore, the relatively fixed ratio between $MAOM_C$ and POM_C means that the C-saturation concept of $MAOM_C$ is adequate to estimate the "whole-soil OC" storage potential (Barré et al., 2017). In our study, about 20% of the C accrual in MAOM could potentially be added in POM fractions (Table 1). Even if POM are labile fractions with faster turnover rates (Meyer et al., 2017), and thus more sensitive to climate change, this additional organic C storage in POM would represent a net gain of C storage, as long as land-use type and management remain constant (Chenu et al., 2019). Nonethless, all greenhouse gas emissions generated in order to increase SOC storage should be accounted to determine the net atmospheric C sequestration (Chenu et al., 2019).

4.2. SOC accrual for C sequestration and soil physical quality

The relationship between SOC:clay ratio (as indicator of soil physical quality) and the degree of saturation was clear in croplands because an increase of the level of $MAOM_C$ saturation was associated with a gain in soil physical quality as indicated by the increase in SOC:clay ratio (Fig. 3). One possible mechanism explaining the direct effect of $MAOM_C$ saturation on soil physical quality is that the dispersability of clay is reduced when the particles are coated with organic matter (Dexter et al., 2008; Schjønning et al., 2012). The majority of cropland sites were below the 1:10 threshold (Fig. 3), as already observed in other regions of Switzerland (Dupla et al., 2021) and in the UK (Prout et al., 2020). This indicates that the increase of C storage in those soils would also be highly beneficial for their resistance to mechanical stress from field machinery operations or extreme climatic events such as long droughts along with the overall soil quality (Jensen et al., 2019; Johannes et al., 2017a; Kuzyakov et al., 2020).

By contrast, SOC:clay ratio in permanent grasslands, which was above the 1:10 threshold in almost all sites, was not related to apparent $MAOM_C$-saturation level. As permanent grasslands are supposed to be close to $MAOM_C$ saturation, the POM fraction is likely playing a more important role in the variation of the SOC:clay ratio under this land-use type than the apparent deviation from saturation, as indicated by its positive correlation with the proportion of $cPOM_C$ (Fig. 4). While this result may suggest that promoting management practices that enhance coarse SOC fractions is a way to increase soil quality, it may also highlight a limitation of using the SOC:clay ratio as an indicator of soil physical quality in C-saturated soils. By contrast to $MAOM_C$, the amount of $cPOM_C$ did not increase proportionally with silt + clay in permanent grasslands (Table 1, Fig. 4). Consequently, the upper limit of SOC:clay ratio in saturated soils might be dependent on the clay content. It should be further investigated whether a fixed SOC:clay threshold is valid over the entire range of clay content or if it should be adapted for soils with very high sand or clay content.

Management practices favoring SOC accrual, such as organic amendments application, are not expected to result in a substantial improvement of SOC storage or soil physical quality in permanent grasslands. Indeed, C inputs are less efficient in increasing SOC in soils close to saturation because the organic C-retention rate decreases approaching the saturation level (Castellano et al., 2015; Stewart et al., 2007). Furthermore, the gain of soil physical quality per unit of sequestered C and nutrients is the highest in soils with low clay content because less SOC is necessary to increase the SOC:clay ratio and to reach the clay surface-saturation. Consequently, prioritizing the application of organic amendments in the form of plant residue or animal manure in the least saturated croplands (<65%) with the coarser soil texture seems the most effective strategy to simultaneously address climate change mitigation/adaptation as well as food security.

4.3. Challenging the C-saturation concept

Hassink's approach to determine the maximum capacity of soils to store SOC, using silt + clay content as unique factor, may be seen as an oversimplification because a multitude of other soil characteristics (e.g. mineralogy, pH and Ca content) and climatic factors are known to affect SOC dynamics (Beare et al., 2014; Six et al., 2002; Wiesmeier et al., 2014a). Yet the relationship between $MAOM_C$ in permanent grasslands and the silt + clay content found in our study was almost identical to the one found by Hassink (1997) for a mix of temperate and tropical grasslands.

Within the relatively large range of pedoclimatic conditions in our study region (Table S1), silt + clay remained by far the variable explaining most of the $MAOM_C$ variation (55%) in grasslands (Table 2). With only four variables, the model explained as much as 80 % of the variation, leaving little explanatory power to other variables not measured in this study. The three other variables had a similarly small explanatory power. The effect of soil pH likely reflects an effect of parent material and possibly calcium on SOC stabilization as soils with and without carbonates (pH up to 7.3) were included in the study (Rowley et al., 2018). In the study region, differences in climate (MAT and precipitation) between sites are driven by differences in elevation. The inclusion of elevation in the model thus suggests an effect of climate on the amount of $MAOM_C$ at saturation. Nonetheless, within the 500 m elevation range of the study, corresponding to differences of about 2.5C° and 400 mm rain (Dumas, 2013; Sevruk, 1997), the influence of climate was not significant (Table 2).

The C-saturation concept relating the maximum SOC storage capacity to a particle-size fraction is supported by no effect of clay or silt content on the $MAOM_C$ content in permanent grasslands (Fig. 5b). This suggest that the saturation of silt + clay fraction occurs at a fixed C content of this particle-size fraction independent of the proportion of silt + clay particles: around 43 mg C g^{-1} SC according to the average in permanent grasslands. This C content corresponds to the inflexion point where the relationship between $MAOM_C$ and SOC from Cotrufo et al. (2019) is no longer related to SOC but reaches a plateau (Fig. S1a). The plateau's level also corresponds to the $MAOM_C$ amount expected for soil with 100 % silt + clay saturated in $MAOM_C$. The hypothesis that $MAOM_C$ saturates at a fixed organic C content is further supported by the near zero intercept of the relationship between $MAOM_C$ and silt + clay determined by Hassink (1997), in FRIBO or observed in the measured data of LUCAS (Fig. 5a).

Nonetheless, the Pan-European data from LUCAS shows that there are limitations on the applicability of the same model to large areas. Some LUCAS sites with low silt + clay content had very high measured $MAOM_C$ contents (Fig. 5b and Fig. S1d) and the response of $MAOM_C$ to silt + clay as extracted from the random forest model from Cotrufo et al. (2019) was not linear (Fig. S1c). Thus, the model determined in our study is not expected to be applicable in conditions where specific SOC stabilization mechanisms occur and are not correlated to the silt + fraction. This may be the case for instance where the role of Fe-/Al-oxides and hydroxides becomes prominent as in allophanic soils (Beare et al., 2014), in sandy acidic soils under former heathland vegetation (Wiesmeier et al., 2014a), in poorly drained soils (Vos et al., 2018) or riparian areas (Hennings et al., 2021), which were not included in our study. Hence, models developed using sites where the mechanisms of SOC stabilization are very different might not be sufficiently accurate. Nonetheless, boundary conditions still need further investigation.

The particle-size upper limit for fractions chosen by studies using Hassink's approach has varied between 20 µm vs. 53 µm (Gregorich et al., 2009; Liang et al., 2009; Six et al., 2002). Including coarse silt particles should have a diluting effect because larger soil particles have lower C content (Arrouays et al., 2006; Lutfalla et al., 2019). This is supported by the tendency for lower $MAOM_C$ content to be associated with higher silt content in the FRIBO network (Fig. 5b). However, the fact that models explaining $MAOM_C$ content or amount were not improved by including silt separately or together with clay as compared to clay alone, suggests that the silt fraction and its size limit play only a marginal role in the $MOAM_C$ fraction. Further investigations on the role of silt fraction are needed because the range of silt content in the FRIBO grasslands (23–44 %) was smaller than that of LUCAS sites with measured $MAOM_C$ values (30–69 %), where a stronger negative effect of silt content on $MAOM_C$ content could be observed (Fig. 5b). Nonetheless, the 50 µm limit necessitates separating only the sand fraction from the bulk soil. This greatly simplifies the soil texture and SOC fraction measurements, facilitating the determination of $MAOM_C$ on a large number of soil samples. In addition, the contrasted responses of fPOM and cPOM to land use and silt + clay proportion (Table 1 and Fig. 4) suggest that conceptualizing SOM as a three-pool system (MAOM, fPOM and cPOM) instead of a two-pool system (MAOM and POM), as is currently advocated (Cotrufo et al., 2019; Haddix et al., 2020; Lavallee et al., 2019), might offer better insights into the current SOC storage mechanisms and dynamics.

The second aspect of Hassink's approach, namely the assumption that grasslands are saturated and thus are an appropriate reference to determine the maximum SOC storage, has not yet been tested to our knowledge. The similarity found between Hassink's relationship and the one from our study suggests that permanent grasslands in our study were indeed saturated. However, this assumption does not fully hold given the factors affecting the $MAOM_C$ amounts in permanent grasslands (Table 2). When all factors were included, the rate of SOC accrual in the last 30 years, as estimated by Guillaume et al. (2021), still influenced the $MAOM_C$ amount. In addition, the effect of soil pH, besides being related to soil type, might also indicate that permanent grassland management has affected $MAOM_C$ because intensively managed grasslands tend to experience acidification (Huguenin-Elie et al., 2017; Malhi et al., 1998). Even if we cannot assess whether permanent grasslands with apparent oversaturation are still currently gaining SOC, permanent grasslands may still not represent the maximum attainable SOC levels for the respective pedoclimatic conditions. This would justify using an approach based on quantile regression instead of least square regression (Beare et al., 2014; Feng et al., 2013; McNally et al., 2017).

In conclusion, the soil C-saturation concept based on the silt + clay size fraction showed some limitations for determining the maximum C storage potential due to the difficulties in identifying the maximum SOC storage attainable in grasslands. Even if known, a target for SOC level defined on maximal attainable SOC storage may not have a practical value for land managers given that plant C inputs to the soil and the availability of organic amendments are limited under current agronomic practices. Consequently, the current SOC level as observed in permanent grasslands represents a realistic benchmark for estimating the potential of agricultural soil to sequester atmospheric C and for setting agronomic management priorities for agricultural soils. Furthermore, the approach of using the silt + clay content as a unique factor to determine the saturation level of soils is efficient in capturing most of the heterogeneity within a large landscape but may be inadequate for marginal pedoclimatic conditions.

Declaration of Competing Interest

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

Acknowledgements

The research was funded by Agroscope. We thank the "Institut agricole de l'Etat de Fribourg" for giving access to data and soil samples, as well as the farmers of the FRIBO network for granting access to their fields. We also thank the European Soil Data Center for sharing the data and the associated R script of soil organic matter fractions from LUCAS.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.geoderma.2021.115529.

References

Amundson, R., Biardeau, L., 2018. Soil carbon sequestration is an elusive climate mitigation tool. PNAS 115, 11652–11656. https://doi.org/10.1073/pnas.1815901115.

Angers, D.A., Arrouays, D., Saby, N.P.A., Walter, C., 2011. Estimating and mapping the carbon saturation deficit of French agricultural topsoils. Soil Use Manag. 27, 448–452. https://doi.org/10.1111/j.1475-2743.2011.00366.x.

Arrouays, D., Saby, N., Walter, C., Lemercier, B., Schvartz, C., 2006. Relationships between particle-size distribution and organic carbon in French arable topsoils. Soil Use Manag. 22 (1), 48–51. https://doi.org/10.1111/j.1475-2743.2006.00020.x.

Baldock, J.A., McNally, S.R., Beare, M.H., Curtin, D., Hawke, B., 2019. Predicting soil carbon saturation deficit and related properties of New Zealand soils using infrared spectroscopy. Soil Res. 57, 835–844. https://doi.org/10.1071/SR19149.

Barré, P., Angers, D.A., Basile-Doelsch, I., Bispo, A., Cécillon, L., Chenu, C., Chevallier, T., Derrien, D., Eglin, T.K., Pellerin, S., 2017. Ideas and perspectives: Can we use the soil carbon saturation deficit to quantitatively assess the soil carbon storage potential, or should we explore other strategies? Biogeosciences Discuss. 1–12 https://doi.org/10.5194/bg-2017-395.

Baveye, P.C., Schnee, L.S., Boivin, P., Laba, M., Radulovich, R., 2020. Soil Organic Matter research and climate change: Merely Re-storing carbon versus restoring soil functions. Front. Environ. Sci. 8, 1–8. https://doi.org/10.3389/fenvs.2020.579904.

Beare, M.H., McNeill, S.J., Curtin, D., Parfitt, R.L., Jones, H.S., Dodd, M.B., Sharp, J., 2014. Estimating the organic carbon stabilisation capacity and saturation deficit of soils: A New Zealand case study. Biogeochemistry 120 (1-3), 71–87. https://doi.org/10.1007/s10533-014-9982-1.

Blanchet, G., Gavazov, K., Bragazza, L., Sinaj, S., 2016. Responses of soil properties and crop yields to different inorganic and organic amendments in a Swiss conventional farming system. Agric. Ecosyst. Environ. 230, 116–126. https://doi.org/10.1016/j.agee.2016.05.032.

Bossio, D.A., Cook-Patton, S.C., Ellis, P.W., Fargione, J., Sanderman, J., Smith, P., Wood, S., Zomer, R.J., von Unger, M., Emmer, I.M., Griscom, B.W., 2020. The role of soil carbon in natural climate solutions. Nat. Sustain. 3 (5), 391–398. https://doi.org/10.1038/s41893-020-0491-z.

Cai, A., Xu, H.u., Duan, Y., Zhang, X., Ashraf, M.N., Zhang, W., Xu, M., 2021. Changes in mineral-associated carbon and nitrogen by long-term fertilization and sequestration potential with various cropping across China dry croplands. Soil Tillage Res. 205, 104725. https://doi.org/10.1016/j.still.2020.104725.

Carter, M.R., Angers, D.A., Gregorich, E.G., Bolinder, M.A., 2003. Characterizing organic matter retention for surface soils in eastern Canada using density and particle size fractions. Can. J. Soil Sci. 83 (1), 11–23. https://doi.org/10.4141/S01-087.

Castellano, M.J., Mueller, K.E., Olk, D.C., Sawyer, J.E., Six, J., 2015. Integrating plant litter quality, soil organic matter stabilization, and the carbon saturation concept. Glob. Chang. Biol. 21 (9), 3200–3209. https://doi.org/10.1111/gcb.2015.21.issue-910.1111/gcb.12982.

Chen, S., Arrouays, D., Angers, D.A., Martin, M.P., Walter, C., 2019. Soil carbon stocks under different land uses and the applicability of the soil carbon saturation concept. Soil Tillage Res. 188, 53–58. https://doi.org/10.1016/j.still.2018.11.001.

Chenu, C., Angers, D.A., Barré, P., Derrien, D., Arrouays, D., Balesdent, J., 2019. Increasing organic stocks in agricultural soils: Knowledge gaps and potential innovations. Soil Tillage Res. 188, 41–52. https://doi.org/10.1016/j.still.2018.04.011.

Cotrufo, M.F., Ranalli, M.G., Haddix, M.L., Six, J., Lugato, E., 2019. Soil carbon storage informed by particulate and mineral-associated organic matter. Nat. Geosci. 12 (12), 989–994. https://doi.org/10.1038/s41561-019-0484-6.

Dexter, A.R., Richard, G., Arrouays, D., Czyż, E.A., Jolivet, C., Duval, O., 2008. Complexed organic matter controls soil physical properties. Geoderma 144 (3-4), 620–627. https://doi.org/10.1016/j.geoderma.2008.01.022.

DIAF, 2019. Rapport agricole. Fribourg. https://doi.org/10.1017/CBO9781107415324.004.

Dumas, M.D., 2013. Changes in temperature and temperature gradients in the French Northern Alps during the last century. Theor. Appl. Climatol. 111 (1-2), 223–233. https://doi.org/10.1007/s00704-012-0659-1.

Dupla, X., Gondret, K., Sauzet, O., Verrecchia, E., Boivin, P., 2021. Changes in topsoil organic carbon content in the Swiss leman region cropland from 1993 to present. Insights from large scale on-farm study. Geoderma 400, 115125. https://doi.org/10.1016/j.geoderma.2021.115125.

Duval, M.E., Galantini, J.A., Martínez, J.M., Limbozzi, F., 2018. Labile soil organic carbon for assessing soil quality: influence of management practices and edaphic conditions. Catena 171, 316–326. https://doi.org/10.1016/j.catena.2018.07.023.

Feng, W., Plante, A.F., Six, J., 2013. Improving estimates of maximal organic carbon stabilization by fine soil particles. Biogeochemistry 112 (1-3), 81–93. https://doi.org/10.1007/s10533-011-9679-7.

Frau, L.J., Libohova, Z., Joost, S., Levasseur, C., Jeangros, B., Bragazza, L., Sinaj, S., 2020. Regional investigation of spatial-temporal variability of soil magnesium - a case study from Switzerland. Geoderma Reg. 21, e00278. https://doi.org/10.1016/j.geodrs.2020.e00278.

Fujisaki, K., Chapuis-Lardy, L., Albrecht, A., Razafimbelo, T., Chotte, J.L., Chevallier, T., 2018. Data synthesis of carbon distribution in particle size fractions of tropical soils: Implications for soil carbon storage potential in croplands. Geoderma 313, 41–51. https://doi.org/10.1016/j.geoderma.2017.10.010.

Gregorich, E.G., Carter, M.R., Angers, D.A., Drury, C.F., 2009. Using a sequential density and particle-size fractionation to evaluate carbon and nitrogen storage in the profile of tilled and no-till soils in eastern Canada. Can. J. Soil Sci. 89 (3), 255–267. https://doi.org/10.4141/CJSS08034.

Guillaume, T., Bragazza, L., Levasseur, C., Libohova, Z., Sinaj, S., 2021. Long-term soil organic carbon dynamics in temperate cropland-grassland systems. Agric. Ecosyst. Environ. 305, 107184. https://doi.org/10.1016/j.agee.2020.107184.

Haddix, M.L., Gregorich, E.G., Helgason, B.L., Janzen, H., Ellert, B.H., Francesca Cotrufo, M., 2020. Climate, carbon content, and soil texture control the independent formation and persistence of particulate and mineral-associated organic matter in soil. Geoderma 363, 114160. https://doi.org/10.1016/j.geoderma.2019.114160.

Hassink, J., 1997. The capacity of soils to preserve organic C and N by their association with clay and silt particles. Plant Soil 191, 77–87. https://doi.org/10.1023/A:1004213929699.

Haynes, R.J., 2005. Labile Organic Matter Fractions as Central Components of the Quality of Agricultural Soils: An Overview. Adv. Agron. 85, 221–268. https://doi.org/10.1016/S0065-2113(04)85005-3.

Hennings, N., Becker, J.N., Guillaume, T., Damris, M., Dippold, M.A., Kuzyakov, Y., 2021. Riparian wetland properties counter the effect of land-use change on soil carbon stocks after rainforest conversion to plantations. Catena 196, 104941. https://doi.org/10.1016/j.catena.2020.104941.

Huguenin-Elie, O., Mosimann, E., Schlegel, P., Lüscher, A., Kessler, W., Jeangros, B., 2017. Fertilisation des herbages, in: Sinaj, S., Richner, W. (Eds.), Principes de Fertilisation Des Cultures Agricoles En Suisse (PRIF 2017). Recherche Agronomique Suisse (8) 6, publication speciale, pp. 9/1–9/21.

Jensen, J.L., Schjønning, P., Watts, C.W., Christensen, B.T., Peltre, C., Munkholm, L.J., 2019. Relating soil C and organic matter fractions to soil structural stability. Geoderma 337, 834–843. https://doi.org/10.1016/j.geoderma.2018.10.034.

Jilling, A., Kane, D., William, A., Yannarell, A.C., Davis, A., Jordan, N.R., Koide, R.T., Mortensen, D.A., Smith, R.G., Snapp, S.S., Spokas, K.A., Grandy, A.S., 2020. Rapid and distinct responses of particulate and mineral associated organic nitrogen to conservation tillage and cover crops. Geoderma 359, 114001. https://doi.org/10.1016/j.geoderma.2019.114001.

Johannes, A., Matter, A., Schulin, R., Weisskopf, P., Baveye, P.C., Boivin, P., 2017a. Optimal organic carbon values for soil structure quality of arable soils. Does clay content matter? Geoderma 302, 14–21. https://doi.org/10.1016/j.geoderma.2017.04.021.

Johannes, A., Weisskopf, P., Schulin, R., Boivin, P., 2017b. To what extent do physical measurements match with visual evaluation of soil structure? Soil Tillage Res. 173, 24–32. https://doi.org/10.1016/j.still.2016.06.001.
Julien, P., Morand, D., 1995. FRIBO: Réseau fribourgeois d'observation des sols. Posieux, pp. 1987–1994.
Koenker, R., 2017. Quantile regression: 40 years on. Annu. Rev. Econom. 9 (1), 155–176. https://doi.org/10.1146/annurev-economics-063016-103651.
Koishi, A., Bragazza, L., Maltas, A., Guillaume, T., Sinaj, S., 2020. Long-term effects of organic amendments on soil organic matter quantity and quality in conventional cropping systems in Switzerland. Agronomy 10 (12), 1977. https://doi.org/10.3390/agronomy10121977.
Kuzyakov, Y., Gunina, A., Zamanian, K., Tian, J., Luo, Y., Xu, X., Yudina, A., Aponte, H., Alharbi, H., Ovsepyan, L., Kurganova, I., Ge, T., Guillaume, T., 2020. New approaches for evaluation of soil health, sensitivity and resistance to degradation. Front. Agric. Sci. Eng. 7, 282–288. https://doi.org/10.15302/J-FASE-2020338.
Lavallee, J.M., Soong, J.L., Cotrufo, M.F., 2019. Conceptualizing soil organic matter into particulate and mineral-associated forms to address global change in the 21st century. Glob. Chang. Biol. 26 (1), 261–273. https://doi.org/10.1111/gcb.14859.
Levasseur, C., Favrelière, E., von Niederhäusern, A., Brülhart, J., Rossier, N., 2019. FRIBO: Réseau fribourgeois d'observation des sols 1987-2016. Posieux.
Liang, A., Yang, X., Zhang, X., McLaughlin, N., Shen, Y., Li, W., 2009. Soil organic carbon changes in particle-size fractions following cultivation of Black soils in China. Soil Tillage Res. 105 (1), 21–26. https://doi.org/10.1016/j.still.2009.05.002.
Lugato, E., Leip, A., Jones, A., 2018. Mitigation potential of soil carbon management overestimated by neglecting N2O emissions. Nat. Clim. Chang. 8 (3), 219–223. https://doi.org/10.1038/s41558-018-0087-z.
Lutfalla, S., Barré, P., Bernard, S., Le Guillou, C., Alléon, J., Chenu, C., 2019. Multidecadal persistence of organic matter in soils: Multiscale investigations down to the submicron scale. Biogeosciences 16 (7), 1401–1410. https://doi.org/10.5194/bg-16-1401-2019.
Maillard, É., Angers, D.A., Chantigny, M., Bittman, S., Rochette, P., Lévesque, G., Hunt, D., Parent, L.É., 2015. Carbon accumulates in organo-mineral complexes after long-term liquid dairy manure application. Agric. Ecosyst. Environ. 202, 108–119. https://doi.org/10.1016/j.agee.2014.12.013.
Malhi, S.S., Nyborg, M., Harapiak, J.T., 1998. Effects of long-term N fertilizer-induced acidification and liming on micronutrients in soil and in bromegrass hay. Soil & Tillage Research 48, 91–101. https://doi.org/10.1016/S0167-1987(98)00097-X.
Maltas, A., Charles, R., Jeangros, B., Sinaj, S., 2013. Effect of organic fertilizers and reduced-tillage on soil properties, crop nitrogen response and crop yield: Results of a 12-year experiment in Changins, Switzerland. Soil Tillage Res. 126, 11–18. https://doi.org/10.1016/j.still.2012.07.012.
McNally, S.R., Beare, M.H., Curtin, D., Meenken, E.D., Kelliher, F.M., Calvelo Pereira, R., Shen, Q., Baldock, J., 2017. Soil carbon sequestration potential of permanent pasture and continuous cropping soils in New Zealand. Glob. Chang. Biol. 23 (11), 4544–4555. https://doi.org/10.1111/gcb.2017.23.issue-1110.1111/gcb.13720.
Meyer, N., Bornemann, L., Welp, G., Schiedung, H., Herbst, M., Amelung, W., 2017. Carbon saturation drives spatial patterns of soil organic matter losses under long-term bare fallow. Geoderma 306, 89–98. https://doi.org/10.1016/j.geoderma.2017.07.004.
Poeplau, C., Don, A., 2013. Sensitivity of soil organic carbon stocks and fractions to different land-use changes across Europe. Geoderma 192, 189–201. https://doi.org/10.1016/j.geoderma.2012.08.003.
Prout, J.M., Shepherd, K.D., McGrath, S.P., Kirk, G.J.D., Haefele, S.M., 2020. What is a good level of soil organic matter? An index based on organic carbon to clay ratio. Eur. J. Soil Sci. 1–11 https://doi.org/10.1111/ejss.13012.
R Core Team, 2020. R: a language and environment for statistical computing. R fundation for statistical computing, Vienna, Austria.
Rowley, M.C., Grand, S., Verrecchia, É.P., 2018. Calcium-mediated stabilisation of soil organic carbon. Biogeochemistry 137 (1-2), 27–49. https://doi.org/10.1007/s10533-017-0410-1.
Rüegg, J., Quezada, J.C., Santonja, M., Ghazoul, J., Kuzyakov, Y., Buttler, A., Guillaume, T., 2019. Drivers of soil carbon stabilization in oil palm plantations. L. Degrad. Dev. 30 (16), 1904–1915. https://doi.org/10.1002/ldr.3380.
Rumpel, C., Amiraslani, F., Chenu, C., Garcia Cardenas, M., Kaonga, M., Koutika, L.-S., Ladha, J., Madari, B., Shirato, Y., Smith, P., Soudi, B., Soussana, J.-F., Whitehead, D., Wollenberg, E., 2020. The 4p1000 initiative: Opportunities, limitations and challenges for implementing soil organic carbon sequestration as a sustainable development strategy. Ambio 49 (1), 350–360. https://doi.org/10.1007/s13280-019-01165-2.
Samson, M.-É., Chantigny, M.H., Vanasse, A., Menasseri-Aubry, S., Angers, D.A., 2020. Coarse mineral-associated organic matter is a pivotal fraction for SOM formation and is sensitive to the quality of organic inputs. Soil Biol. Biochem. 149, 107935. https://doi.org/10.1016/j.soilbio.2020.107935.
Sanderman, J., Hengl, T., Fiske, G.J., 2017. Soil carbon debt of 12,000 years of human land use. Proc. Natl. Acad. Sci. USA 114 (36), 9575–9580. https://doi.org/10.1073/pnas.1706103114.
Schjønning, P., de Jonge, L.W., Munkholm, L.J., Moldrup, P., Christensen, B.T., Olesen, J.E., 2012. Clay dispersibility and soil friability-testing the soil clay-to-carbon saturation concept. Vadose Zo. J. 11 (1) https://doi.org/10.2136/vzj2011.0067.
Schmidt, M.W.I., Torn, M.S., Abiven, S., Dittmar, T., Guggenberger, G., Janssens, I.A., Kleber, M., Kögel-Knabner, I., Lehmann, J., Manning, D.A.C., Nannipieri, P., Rasse, D.P., Weiner, S., Trumbore, S.E., 2011. Persistence of soil organic matter as an ecosystem property. Nature 478 (7367), 49–56. https://doi.org/10.1038/nature10386.
Sevruk, B., 1997. Regional dependency of precipitation-altitude relationship in the Swiss Alps. Clim. Change 36, 355–369. https://doi.org/10.1007/978-94-015-8905-5_7.
Six, J., Conant, R.T., Paul, E.A., Paustian, K., 2002. Stabilization mechanisms of soil organic matter: Implications for C-saturation of soils. Plant Soil 241, 155–176. https://doi.org/10.1023/a:1016125726789.
Sparrow, L.A., Belbin, K.C., Doyle, R.B., 2006. Organic carbon in the silt + clay fraction of Tasmanian soils. Soil Use Manag. 22 (2), 219–220. https://doi.org/10.1111/sum.2006.22.issue-210.1111/j.1475-2743.2006.00021.x.
Stewart, C.E., Paustian, K., Conant, R.T., Plante, A.F., Six, J., 2007. Soil carbon saturation: concept, evidence and evaluation. Biogeochemistry 86 (1), 19–31. https://doi.org/10.1007/s10533-007-9140-0.
van Groenigen, J.W., van Kessel, C., Hungate, B.A., Oenema, O., Powlson, D.S., van Groenigen, K.J., 2017. Sequestering Soil Organic Carbon: A Nitrogen Dilemma. Environ. Sci. Technol. 51 (9), 4738–4739. https://doi.org/10.1021/acs.est.7b01427.
Vos, C., Jaconi, A., Jacobs, A., Don, A., 2018. Hot regions of labile and stable soil organic carbon in Germany – Spatial variability and driving factors Cora. Soil 4, 153–167. https://doi.org/10.5194/soil-4-153-2018.
Wiesmeier, M., Hübner, R., Spörlein, P., Geuß, U., Hangen, E., Reischl, A., Schilling, B., von Lützow, M., Kögel-Knabner, I., 2014a. Carbon sequestration potential of soils in southeast Germany derived from stable soil organic carbon saturation. Glob. Chang. Biol. 20 (2), 653–665. https://doi.org/10.1111/gcb.2014.20.issue-210.1111/gcb.12384.
Wiesmeier, M., Mayer, S., Burmeister, J., Hübner, R., Kögel-Knabner, I., 2020. Feasibility of the 4 per 1000 initiative in Bavaria: A reality check of agricultural soil management and carbon sequestration scenarios. Geoderma 369, 114333. https://doi.org/10.1016/j.geoderma.2020.114333.
Wiesmeier, M., Munro, S., Barthold, F., Steffens, M., Schad, P., Kögel-Knabner, I., 2015. Carbon storage capacity of semi-arid grassland soils and sequestration potentials in northern China. Glob. Chang. Biol. 21 (10), 3836–3845. https://doi.org/10.1111/gcb.12957.
Wiesmeier, M., Schad, P., von Lützow, M., Poeplau, C., Spörlein, P., Geuß, U., Hangen, E., Reischl, A., Schilling, B., Kögel-Knabner, I., 2014b. Quantification of functional soil organic carbon pools for major soil units and land uses in southeast Germany (Bavaria). Agric. Ecosyst. Environ. 185, 208–220. https://doi.org/10.1016/j.agee.2013.12.028.
Zhao, L., Sun, Y., Zhang, X., Yang, X., Drury, C.F., 2006. Soil organic carbon in clay and silt sized particles in Chinese mollisols: Relationship to the predicted capacity. Geoderma 132 (3-4), 315–323. https://doi.org/10.1016/j.geoderma.2005.04.026.