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1 **Title:** Soil carbon stocks under different land uses and the applicability of the soil
2 carbon saturation concept

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Abstract

Increasing soil organic carbon (SOC) stocks via land management has been proposed as a temporary climate change mitigation measure. An upper limit of soil stable SOC storage, which refers to the concepts of SOC saturation has been proposed. Using systematic grid sampling of topsoil in mainland France and an equation that predicts this SOC saturation, we derived estimates of the SOC sequestration potential density (SOC_{spd}) for various land uses. First, using French database and data from the literature we estimated the proportions of the SOC stored in the fine fraction (SOC_{fine}) in total SOC for grassland (69%), cropland (85%) and forest (66%). Then, SOC_{spd} was calculated as the difference between the theoretical SOC saturation value and SOC_{fine} . The SOC_{spd} stocks for French topsoil were estimated at about 1.1 Pg, in which cropland, forest and grassland accounted for 66%, 17% and 17%, respectively. Varying the proportions of SOC_{fine} in the calculations by assuming a possible range of 0.8-0.9 for cropland and 0.6-0.9 for grassland and forest soils led to variations of total SOC_{spd} stocks of about 0.1 Pg for each land use. Most importantly, we demonstrate that the distribution of SOC_{spd} in forest soils is exactly centred at zero, which suggests that on average, forest topsoils are saturated in SOC_{fine} , and thus Hassink's equation provides a valid estimate for the SOC sequestration potential for French topsoil.

Keywords

Soil organic carbon; Soil carbon saturation; Carbon sequestration potential; Land use; Forest soils

1. Introduction

Soil organic carbon (SOC) plays a major role in the global greenhouse gas balance (Lal, 2004, 2010). Soils throughout the world store four times more SOC than the biosphere and approximately two to three times more carbon than the atmosphere (Batjes, 1996; Stockmann et al., 2015; Le Quéré et al., 2016). Thus, a relatively small change in this reservoir may have a large effect on CO₂ balance. Anthropogenic perturbations have strongly depleted SOC stocks (Bellamy et al., 2005; Sanderman et al., 2017) contributing significantly to increased CO₂ concentration (Canadell et al., 2007). Increasing SOC is beneficial not only for mitigating climate change but also for restoring degraded soils with major potential impacts on crop yields, food security, and the well-being of smallholder farmers (Smith et al., 2008, 2012; Chabbi et al., 2017; Paustian et al., 2016). At the 21st session of the United Nations Framework Convention on Climate Change (UNFCCC, COP21), a voluntary action plan, the “4 per 1000 Initiative: Soils for Food Security and Climate” was proposed (<https://www.4p1000.org/>). The 4 per 1000 initiative is intended to foster food security and reduce atmospheric CO₂ concentration by increasing the SOC stocks at an annual rate of 0.4%.

Interest around the feasibility, range and duration of additional SOC sequestration has rapidly risen (Lu et al., 2011; Smith et al., 2012; Bradford et al., 2016; Paustian et al., 2016; Chabbi et al., 2017; Dignac et al., 2017; Minasny et al., 2017; van Groenigen et al., 2017). But it has also generated controversy, such as the need for additional nitrogen input to meet stoichiometric demand, the feasibility of implementing changes in management, and the non-permanence of the SOC stocks (Smith, 2005; van Groenigen et al., 2017; Poulton et al., 2018). However, there is a general consensus regarding beneficial practices that may increase SOC (Smith et

al., 2008; Lal, 2010; Smith et al., 2012; Paustian et al., 2016; Chabbi et al., 2017; Dignac et al., 2017; Minasny et al., 2017; Chenu et al., 2018).

Bonding of SOC to the fine mineral particles is considered one of the most significant SOC stabilization mechanisms for mineral soils (Hassink, 1997; Baldock and Skjemstad, 2000; Six et al., 2002), and as the amount of fine particles is finite, the hypothesis of an upper limit of soil stable SOC storage, which refers to SOC saturation, is generally accepted (Hassink, 1997; Baldock and Skjemstad, 2000; Six et al., 2002; Stewart et al., 2007; Dignac et al., 2017). Hassink (1997) proposed an equation to describe the relationship between maximum SOC storage in the fine fraction, which is assumed to be more stable than the coarse SOC, and the soil fine fraction ($<20\ \mu\text{m}$, clay and fine silt) for world soils. In this context, the fine fraction represents a proxy for the mineral surface. The SOC saturation deficit or SOC sequestration potential (SOC_{sp}) can be calculated as the difference between the theoretical SOC saturation and the SOC stored in the fine fraction (SOC_{fine}).

Here, we make a distinction between SOC sequestration, defined as long-term stabilized SOC associated with the soil fine fraction, and soil SOC storage, which refers to the whole-soil SOC content (including the coarse or sand-size fraction) (Chenu et al., 2018). Recent discussions have emphasized that the saturation deficit concept may not be suitable to estimate short- to medium-term (10-30 years) whole-soil (including the sand-size fraction) SOC storage potential under varying agro- and climatic conditions (Barré et al., 2017). Indeed, the SOC saturation concept only applies to the soil fine fraction and is theoretically related to the inherent capacity of soils to stabilize (sequester) SOC in the long term in the fine fraction (Hassink, 1997). Other tools (e.g., data-driven approaches and process-based prediction models) may

be more appropriate to estimate the whole-soil SOC storage potential under varying management and climatic conditions for a given period of time (Barré et al., 2017).

Changes in land cover and land use have a major effect on SOC stocks (Post and Kwon, 2000; Guo and Gifford, 2002; Lal, 2010; Smith et al., 2012; Stockmann et al., 2015). Forests and grasslands usually show the highest SOC stocks among land covers (Carter et al., 1998; Post and Kwon, 2000; Conant et al., 2001; Guo and Gifford, 2002; Soussana et al., 2004; McNally et al., 2017). Forest and grassland soils are therefore often used as reference for the maximum reachable SOC stocks and SOC saturation (Conant et al., 2001; Guo and Gifford, 2002; Lugato et al., 2014).

Here we analyse the statistical distribution of SOC sequestration potential density (SOC_{spd}) from an unbiased sampling of topsoil in mainland France. Our objectives were to apply the carbon saturation concept for French topsoil, to estimate their SOC_{spd} , and to determine if some land uses could be used as a reference for estimating soil SOC sequestration potential.

2. Material and methods

2.1. Sampling and measurements

Data come from the systematic grid of the French Soil Monitoring Network (RMQS). This network is based on a 16 km x 16 km square grid and the sites are selected at the center of each grid cell resulting in about 2000 soil sampling sites (Martin et al., 2011). Twenty-five individual core samples were collected for topsoil (0-30 cm) based on an unaligned sampling design within a 20 m x 20 m square, and then were mixed into a composite sample for each site. In the south border of the 20 m x 20 m square, a soil pit was dug to record main soil characteristics including bulk density with three replicates. Soil organic carbon content was measured using an automated C-N

analyzer by the dry combustion method (Thermofisher NA 2000) and bulk density was determined by taking samples of known volume and drying them to constant weight. Particle-size analysis was determined using the pipette method. More details about the sampling strategy are found in Chen et al. (2018).

2.2. Calculation of SOC sequestration potential

The SOC saturation of soil fine fraction (<20 µm, clay and fine silt) was calculated according to the equation proposed by Hassink (1997):

$$SOC_{sat} = 4.09 + 0.37 \times \text{fine fraction} \quad (1)$$

where SOC_{sat} is the SOC saturation (g kg⁻¹) and *fine fraction* is the content of soil particle-size < 20 µm (%).

The SOC saturation deficit was calculated as the difference between SOC saturation and the SOC stored in soil fine fraction (SOC_{fine}). The SOC_{fine} content of cropland soils was assumed to comprise 85% of the total SOC based on previous studies (Balesdent, 1996; Jolivet et al., 2003; Angers et al., 2011). We also summarized previous studies from regions with similar climate (Table 1) and derived the proportions of SOC_{fine} in the total SOC by weighted averaging (according to the number of samples per study) for grassland (69±9%) and forest (66±2%).

The SOC saturation deficit or SOC sequestration potential (SOC_{sp}) was calculated as follows:

$$SOC_{sp} = SOC_{sat} - SOC_{fine} \quad (2)$$

where SOC_{sp} is the SOC sequestration potential (g kg⁻¹) and SOC_{fine} is SOC in fine fraction (g kg⁻¹).

The SOC sequestration potential density was calculated using the following equation:

$$SOC_{spd} = p \times SOC_{sp} \times BD \times (100 - ce) \times 10^{-2} \quad (3)$$

where SOC_{spd} is the SOC sequestration potential density ($kg\ m^{-2}$) in topsoil layer (0-30 cm), BD , SOC_{sp} and ce are the bulk density ($kg\ m^{-3}$), sequestration potential ($g\ kg^{-1}$ or $\%$), ce is the percentage of coarse elements ($>2\ mm$, $\%$), and p is the thickness (m) of topsoil horizon.

As the RMQS is a systematic grid covering the whole territory (mainland France), it can be used in a straightforward and unbiased way to estimate mean values and total SOC_{spd} stocks (Brus and de Gruijter, 1997). Using RMQS sites we calculated the estimates of SOC_{spd} stocks at the national scale. Estimates of SOC_{spd} stocks were calculated by multiplying the total area of each land use by its mean SOC_{spd} (negative values were replaced by 0 as they have by definition no potential to sequester SOC) of observed RMQS sites under cropland, forest and grassland.

2.3. Sensitivity analysis

As a large part of uncertainty comes from the parameter setting for the proportion of SOC_{fine} in the total SOC for each land use, we first assessed the influence of proportion setting on the estimates of French national SOC_{spd} stocks in topsoil. Then, in order to assess how the distribution of SOC_{fine} proportion may change the distribution of SOC_{spd} , we applied Gaussian distribution simulation (1000 times) for the forest soils with SOC_{spd} around 0 (-0.5 to $0.5\ kg\ m^{-2}$). In the Gaussian distribution simulation, the mean value was defined as 0.66 (mean value used for forest soils), and three different standard deviation values (σ at 0.1, 0.2 and 0.3) were tested.

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3. Results

3.1. SOC sequestration potential under land uses

The relative frequency distribution of SOC_{spd} was analysed for three main land uses: forest, grassland and cropland (Fig. 1). Positive values indicate the potential to sequester additional stable SOC, whereas negative values indicate seemingly oversaturated soils, the implication of which is discussed later. Nearly 84% of cropland soils are depleted in stable SOC, with a modal SOC_{spd} value of approximately 4 kg m^{-2} (40 Tg ha^{-1}).

The relative frequency distribution of SOC_{spd} of forest soils is exactly centred at zero. Its mean, modal and median values are 0.06, 0, and 0.28 kg m^{-2} , respectively. The distribution is skewed and exhibits a tail for most negative values, which suggests that some forest soils may be oversaturated in stable SOC.

Skewness is also observed for the relative frequency distribution of SOC_{spd} under grassland. The distribution is centered at 2 kg m^{-2} with a sharp increase in the frequency around this value, whereas its median value is close to zero (-1.40 kg m^{-2}). Among the three frequency distributions, the values under forest are the most scattered and show a large proportion of negative values.

3.2. SOC_{spd} sequestration potential density and soil fine fractions

The SOC_{spd} is related to the fine mineral particle fraction ($0\text{--}20 \mu\text{m}$) as per Hassink's equation. There is a general increase in SOC_{spd} with an increase in fine fraction content, but the magnitude differs between land uses (Fig. 2). Cropland soils show a greater slope value (0.08) and Pearson correlation coefficient (0.47) of the fitted line between SOC_{spd} and fine fraction content than forest and grassland soils. This result suggests that the increase in SOC_{spd} with increasing fine fraction is highest for cropland.

3.3. SOC_{spd} stocks and sensitivity analysis

The SOC_{spd} stocks in French topsoil were about 1.1 Pg, of which cropland, forest and grassland accounted for 66%, 17% and 17%, respectively. Varying the proportion of SOC_{fine} in the calculation (assuming a possible range of 0.8-0.9 for croplands and 0.6-0.9 for grasslands and forests) led to variations of total SOC_{spd} stocks of about 0.1 Pg for each land use (Fig. 3). Fig. 4 shows that the SOC_{spd} distribution is very sensitive to variation in the proportion of SOC_{fine} used. The flatter and the wider is the distribution of SOC_{fine}, the larger is the range of SOC_{spd} for simulated forest soils that had a SOC_{spd} close to zero when using the fixed value of 0.66 for the SOC_{fine} proportion.

4. Discussion

The observed differences among land uses in topsoil confirm the well-known effect of SOC depletion with cultivation (Post and Kwon, 2000; Conant et al., 2001; Guo and Gifford, 2002; McNally et al., 2017). The mean values that we obtained for cropland, grassland and forest soils are consistent with trends of SOC associated with land use change reported in the literature (Post and Kwon, 2000; Guo and Gifford, 2002).

Among the invoked limitations of the SOC saturation concept, is the lack of field validation (O'Rourke et al., 2015). Our findings indicate that the SOC saturation concept for the French climate is sensitive to land use. Similar ranges of SOC_{spd} have been identified for cultivated soils using an entirely different and independent data set (Angers et al., 2011) which indicates that our findings are robust.

Following a previous study (Angers et al., 2011), we postulated that part (15%) of the SOC measured in cropland soil was not bound to minerals and was unprotected. Similarly, we used lower values from the literature to estimate this proportion of unprotected SOC in grassland and forest soils as a higher proportion of unbound

SOC is usually observed in these soils. Although we adapted the proportion to land use, we may have under- or overestimated the protected SOC in some soils with very high SOC contents. Moreover, our sensitivity analysis shows that SOC_{spd} is very sensitive to the proportion of SOC_{fine} used in the calculations. Assuming that there is a variation around the mean values we used, a large part of the dispersion in SOC_{spd} under forest soils may be attributable to the variability in the proportion of SOC_{fine} . This may also explain why we find some negative values of SOC_{spd} (oversaturation of the fine fraction) which should not happen in theory.

To estimate stable SOC saturation, we employed an equation that was derived for soils from around the world (Hassink, 1997). Therefore, some bias and uncertainty may be associated with the derived estimates. The equation we used is linear; however, as suggested by some authors (Stockmann et al., 2013), this relation may follow a curve that reaches a plateau at high fine particle content.

Unlike many previous studies looking at national/regional estimates of SOC sequestration, our results are statistically unbiased. Indeed, our sampling scheme, which is based on a systematic grid for which the origin was not purposely chosen, corresponds to the standards of an unbiased statistical sample. Apart from noticeable exceptions (e.g., Bellamy et al., 2005, Wiesmeier et al., 2014), most of the broad-scale inventory or monitoring schemes are based on purposive sampling that is useful for understanding causes of variation but may be biased when applying statistics to determine a mean value or a total stock at a national scale.

One striking finding is that SOC_{spd} under forest exhibits a peak relative frequency distribution that is exactly centred at zero. The fact that the potential additional sequestration is centered at zero under forest suggests that "on average" their fine

fraction is saturated. This does not mean that, for a specific pedo-climate context, forest soils are saturated. However, French forest soils, considered as a whole, provide an adequate reference to estimate the maximum sequestration potential overall, for the French territory. The higher mean SOC_{spd} in grassland may result from the fact that some grassland soils have been already degraded probably due to SOC export from intensive grazing. This may also be related to the fact that grassland soils are on average more clayey (25% clay or 48% fine fraction) than forest soils (22% clay or 43% fine fraction), and, as we can see in Fig. 2, the SOC_{spd} tends to increase with fine fraction content.

The SOC_{spd} stocks for French topsoil under the three main land uses was estimated to be about 1.1 Pg. This potential additional SOC sequestration can be compared to recent estimates of total SOC stocks in French topsoil of about 3.5 Pg (Martin et al., 2011; Meersmans et al., 2012; Mulder et al, 2016). This potential additional amount of SOC stocks represents 4.0 Pg CO₂ equivalent, that is about 12.6 times the French annual CO₂ emission in 2013 (0.32 Pg) (Eurostat, 2015). Cropland has the highest potential to sequester with nearly 66% of the total. The fact that our calculations show that a large additional sequestration potential exists in theory does not necessarily mean that it can be reached in practice. Innovative management practices (Chenu et al., 2018) will need to be developed and proposed to take advantage of this high sequestration potential. Many factors may limit both the duration and potential of SOC sequestration. Among those factors limiting SOC sequestration potential are for instance N and P availability and climatic or water regime effects on net primary productivity and SOC mineralization and stabilization (van Groenigen et al., 2017). It may be that under French climate, the conditions for an optimal SOC sequestration cannot be reached. Moreover, some options for a

maximal SOC sequestration are clearly not realistic (e.g., converting all croplands to grassland or forest) or may not be feasible according to technical, economic or social constraints.

Finally, there is a need to refine our estimate of SOC in the fine fractions for French soils in order to derive more reliable estimates of SOC_{spd}. Further, other approaches to estimate the potential for additional SOC storage, such as data-driven or mechanistic C models (Barré et al., 2017), need to be developed in order to improve the national estimates, their driving factors, and identify suitable land management practices.

5. Conclusion

We estimated SOC_{spd} using an unbiased sampling of topsoil in mainland France and estimates of the proportion of SOC_{fine} compiled from the literature. Our results showed that French topsoils have a large potential to sequester SOC (1.1 Pg) when compared to the total SOC stocks (3.5 Pg). A sensitivity analysis suggested that actual measurements of the proportion of SOC_{fine} would be helpful to decrease the uncertainty of SOC_{spd} estimates. As the distribution of SOC_{spd} in forest soils is centred at zero, we suggest that it may be used as a reference to estimate the maximum SOC sequestration potential in France.

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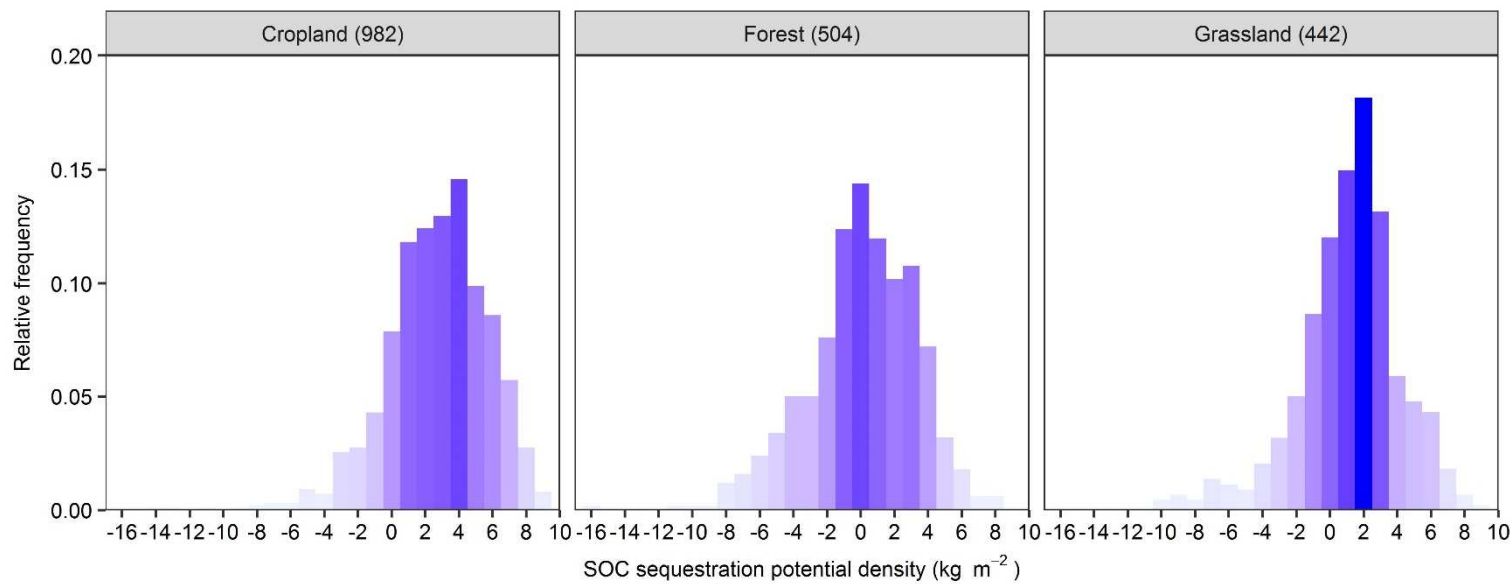
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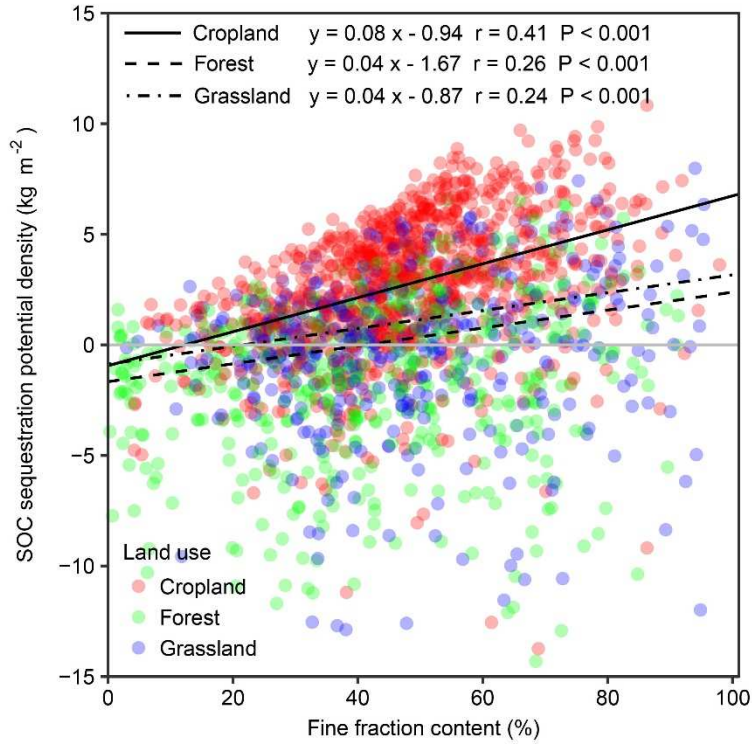
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463 Fig. 1. Relative frequency distribution of SOC_{spd} for three land uses in topsoil (0-30
464 cm). The number of samples is indicated for each land use.

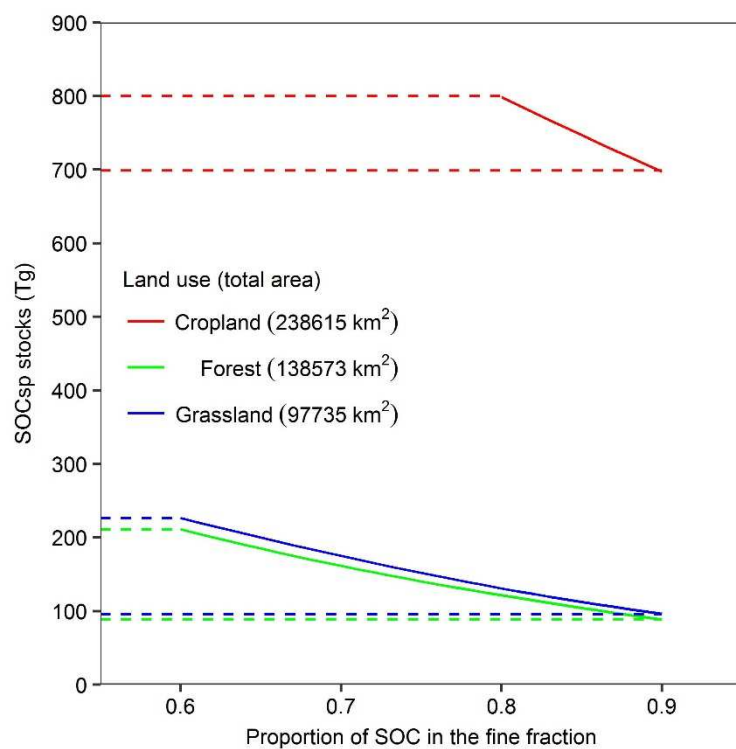


465 Fig. 2. Correlation between the fine fraction content and SOC_{spd}. Solid black lines
 466 black dashed line and black dot dashed line dashed black lines are fitted lines for
 467 cropland, forest and grassland, respectively.

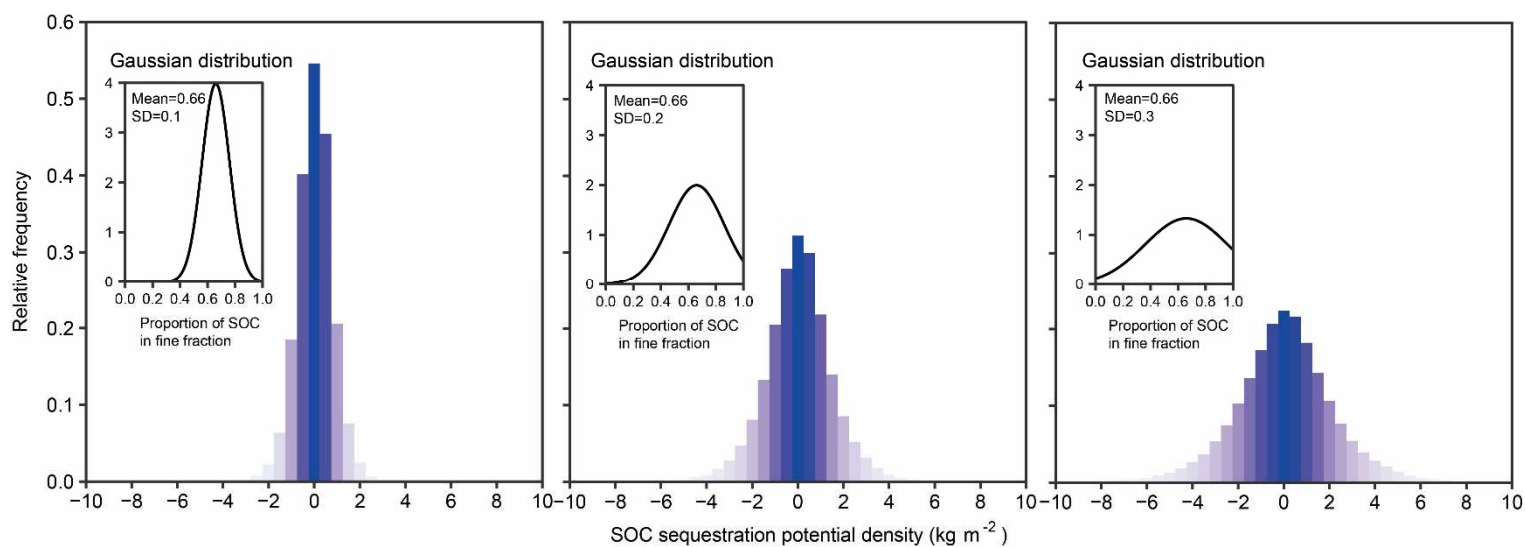


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Fig. 3. The influence of the proportion of SOC in the fine fraction on the national estimates of SOC_{spd} stocks in French topsoil. Solid lines show how SOC_{spd} stocks change with proportion of SOC_{fine} under three land uses and dashed lines are the corresponding minimal and maximal SOC_{spd} stocks for each land use.



474 Figure 4. Sensitivity analysis: influence of the SOC_{fine} proportion distribution on
 475 SOC_{spd}



476

477 Table 1. Proportion of SOC in fine fraction among total SOC for grassland and forest
 478 topsoil (after Chen et al., 2018)

Land use	Country	Sampling depth (cm)	Number of sampling sites	0-20 μm^{a}	0-50 μm^{b}	0-53 μm^{c}	0-63 μm^{d}	Reference
Grassland	USA	0-20	2			0.68		Cambardella and Elliott (1992)
	Canada	0-15	2			0.65		Carter et al. (1998)
	USA	0-20	4			0.80		Conant et al. (2003)
	Belgium	0-20	6		0.58			Accoe et al. (2004)
	France	0-30	2		0.85			Chenu et al. (2004)
	Germany	0-30	1			0.88		John et al. (2005)
	Switzerland	0-20	2				0.60	Leifeld and Fuhrer (2009)
	UK	0-18	1		0.69			Coppin et al. (2009)
	Germany	0-20	11				0.70	Wiesmeier et al. (2014)
Forest	Canada	0-15	1			0.69		Carter et al. (1998)
	France	0-30	1	0.66	0.67			Balesdent et al. (1998)
	France	0-24	5		0.68			Jolivet et al. (2003)
	Germany	0-24(30)	2	0.65				Rumpel et al. (2004)
	Germany	0-25	1			0.56		John et al. (2005)
	Germany	0-20	14				0.66	Wiesmeier et al. (2014)

479 ^a proportion of SOC in 0-20 μm among total SOC content;

480 ^b proportion of SOC in 0-50 μm among total SOC content;

481 ^c proportion of SOC in 0-53 μm among total SOC content;

482 ^d proportion of SOC in 0-63 μm among total SOC content.

