

# Historical advances in the study of global terrestrial soil organic carbon sequestration

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## Abstract

This paper serves two purposes: it provides a summarized scientific history of carbon sequestration in relation to the soil-plant system and gives a commentary on organic wastes and SOC sequestration.

The concept of soil organic carbon (SOC) sequestration has its roots in: (i) the experimental work of Lundegårdh, particularly his *in situ* measurements of CO<sub>2</sub> fluxes at the soil-plant interface (1924, 1927, 1930); (ii) the first estimates of SOC stocks at the global level made by Waksman [Waksman, S.A., 1938. *Humus. Origin, Chemical Composition and Importance in Nature*, second ed. revised. Williams and Wilkins, Baltimore, p. 526] and Rubey [Rubey, W.W., 1951. *Geologic history of sea water*. *Bulletin of the Geological Society of America* 62, 1111–1148]; (iii) the need for models dealing with soil organic matter (SOM) or SOC dynamics beginning with a conceptual SOM model by De Saussure (1780–1796) followed by the mathematical models of Jenny [Jenny, H., 1941. *Factors of Soil Formation: a System of Quantitative Pedology*. Dover Publications, New York, p. 288], Hénin and Dupuis [Hénin, S., Dupuis, M., 1945. *Essai de bilan de la matière organique*. *Annales d'Agronomie* 15, 17–29] and more recently the RothC [Jenkinson, D.S., Rayner, J.H., 1977. *The turnover of soil organic matter in some of the Rothamsted classical experiments*. *Soil Science* 123 (5), 298–305] and Century [Parton, W.J., Schimel, D.S., Cole, C.V., Ojima, D.S., 1987. *Analysis of factors controlling soil organic matter levels in great plains grasslands*. *Soil Science Society of America Journal* 51 (5), 1173–1179] models.

The establishment of a soil C sequestration balance is not straightforward and depends greatly on the origin and the composition of organic matter that is to be returned to the system. Wastes, which are important sources of organic carbon for soils, are taken as an example. For these organic materials the following factors have to be considered: the presence or absence of fossil C, the potential of direct and indirect emissions of non-CO<sub>2</sub> greenhouse gases (CH<sub>4</sub> and N<sub>2</sub>O) following application and the agro-system which is being used as a comparative reference.

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## 1. Introduction

In recent years, increasing attention has been given to soil organic matter (SOM) in relation to carbon (C) sequestration. Concerns about increasing atmospheric greenhouse gas (GHG) concentrations (carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O)), global warming and climate change have raised questions about the potential role of soils as sources or sinks of C (Houghton, 2003).

In this paper, the authors briefly define the term “soil C sequestration” before (i) discussing the historical scientific roots of the present study of soil C sequestration, and (ii) highlighting the difficulties encountered when estimating soil C sequestration balances for systems using organic wastes (henceforth referred to as ‘wastes’).

## 2. A summarized scientific history of C sequestration for the soil-plant system

The terms “sequestration” and “C sequestration” were first proposed to define the aptitude of terrestrial ecosystems to act as sinks for GHGs. The appearance, use and

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significance of the term “C sequestration” are discussed below. In addition, the methods used to estimate C sequestration in soil at different temporal and spatial scales and the methods used to measure CO<sub>2</sub> fluxes in the soil-plant system are considered.

### 2.1. Appearance of the terms ‘C sequestration’ and ‘soil C sequestration’

A bibliometric search of the ISI-Web of Science database covering the period 1945–2005 suggests that the first incidence of the terms “soil”, “carbon” and “sequestration” being used together to present a concept, occurred in 1991. The number of references using these terms increased rapidly during the 15 years that followed (Bernoux et al., 2006) (Table 1). The concept of soil C sequestration is, therefore, a relatively new one.

Most definitions of C sequestration (whether soil specific or not) refer simply to CO<sub>2</sub> removal from the atmosphere and storage in an organic form in the soil or plants. However, CH<sub>4</sub> and N<sub>2</sub>O are also involved in exchanges between the soil-plant system and the atmosphere. The United Nations Framework Convention on Climate Change (UNFCCC) provides an overall framework for intergovernmental efforts to tackle the challenges posed by climate change. Under the UNFCCC, governments have to provide national inventories of anthropogenic emissions (by sources) and removals (by sinks) of all GHGs. To facilitate this, the Intergovernmental Panel on Climate Change (IPCC) published guidelines for the production of GHG

inventories (IPCC/UNEP/OECD/IEA, 1997). Fluxes for all gases are expressed as equivalents of CO<sub>2</sub> after application of a conversion factor that reflects the global warming potential (GWP) of each gas in relation to CO<sub>2</sub>. Current conventions yield a 100 year-GWP value of 23 for CH<sub>4</sub> and 296 for N<sub>2</sub>O. A recent review by Six et al. (2002) highlights the importance of accounting for all GHGs when considering “C sequestration”. The authors found that in both tropical and temperate soils, C levels increased in no-tillage (NT) systems as compared to those under conventional tillage (CT), but that in temperate soils average N<sub>2</sub>O emissions increased substantially under NT as compared to CT and the increase in N<sub>2</sub>O emissions (when expressed on a C–CO<sub>2</sub> equivalent basis) lead to a negative total GWP, even if a positive C storage was observed in the soil. Even the case of systems that use organic forms of N fertilizer can be a hazard when considered in terms of N<sub>2</sub>O emissions (Flessa et al., 2002; Giller et al., 2002; Millar et al., 2004).

When the above issues are considered, it becomes apparent that “soil C sequestration”, as a concept, should not be restricted to a mere quantification of C storage or CO<sub>2</sub> balance. All GHG fluxes must be computed at the plot level in C–CO<sub>2</sub> or CO<sub>2</sub> equivalents, incorporating as many emission sources and sinks as possible across the entire soil-plant system. Therefore, Bernoux et al. (2006), proposed a new definition of C sequestration applied to the soil or soil-plant system:

“Soil C sequestration” or “soil-plant C sequestration”, for a specific agro-ecosystem, in comparison with a reference one, should be considered as the result – for a given period of time and portion of space – of the net balance of all GHGs, expressed in C–CO<sub>2</sub> equivalents or CO<sub>2</sub> equivalents, computing all emissions sources at the soil-plant-atmosphere interface.

The confusion (as is often the case) between the notion of “SOC storage” (C stored in the soil irrespective of its origin) and “soil C sequestration” (GHGs, expressed in equivalent C–CO<sub>2</sub>, stored in the soil and originating from the atmosphere) can thus be avoided.

### 2.2. Early measurements of soil CO<sub>2</sub> concentration and fluxes

#### 2.2.1. The first *in situ* and *in vitro* measurements of soil CO<sub>2</sub> concentrations

The first *in situ* measurements of soil CO<sub>2</sub> concentrations were made by Boussingault and Levy (1852, 1853) at depths ranging from 40 to 240 cm. Using sophisticated equipment, to avoid contamination of soil CO<sub>2</sub> by atmospheric CO<sub>2</sub>, they showed that concentrations of CO<sub>2</sub> in soils without farmyard manure (FYM) application were 22–23 times higher than those found in the atmosphere, and that applying FYM, could increase this concentration by a factor of up to 245.

According to Waksman (1938), the first measurements of soil CO<sub>2</sub> emissions under laboratory controlled condi-

Table 1

Number of references indexed in the ISI-Web of Science (1945–2006) for the word queries “soil,” “carbon,” and “sequestration” (query 1) and “soil” and “carbon” (query 2) in the topics and in the title (in parentheses), updated from Bernoux et al. (2006)

Years	Number of references returned by the Queries		Query 1/Query 2
	Query 1	Query 2	
1945–1990	0	719	0
1991	1 <sup>a</sup>	643	1.6
1992	5 (1 <sup>b</sup> )	694	7.2
1993	14 (1)	816	17.2
1994	7	908	7.7
1995	21 (1)	985	21.3
1996	24	1220	19.7
1997	36 (2)	1398	25.7
1998	47 (3)	1520	30.9
1999	42 (3)	1568	26.8
2000	78 (8)	1618	48.2
2001	107 (14)	1727	62.0
2002	148 (15)	1851	80.0
2003	174 (17)	2142	81.2
2004	229 (33)	2136	107.2
2005	255 (27)	2611	97.6
2006 (till December 6th)	265 (21)	2740	96.7

Queries performed on December 6, 2006.

<sup>a</sup> Thornley et al. (1991).

<sup>b</sup> Dewar and Cannell (1992).

tions were made by Ingen-Housz (1794–1796), who demonstrated the effect of organic inputs and the importance of oxygenation, temperature and humidity. As early as 1855, Corenwinder (1855, 1856) was using equipment which was very similar to today's respirometry apparatus.

### 2.2.2. Measurements of CO<sub>2</sub> fluxes at the soil-plant-atmosphere interface

2.2.2.1. *Lundegårdh's studies at the plot scale.* The main forerunner of modern plot scale measurements was the Danish ecophysiologicalist, Henrik Lundegårdh (1888–1969), whose abridged biography was recently published by Larkum (2003). Between 1924 and 1930, Lundegårdh published considerable data on CO<sub>2</sub> fluxes at the soil-plant interface in two books (1924, 1930) and a large paper (1927).

In these three publications, Lundegårdh reported an impressive number of quantitative data on *in situ* CO<sub>2</sub> fluxes between atmospheric, plant and soil components. Data were collected using instruments for the sampling of soil atmosphere (equivalent to our present day static chambers) or continuous monitoring of CO<sub>2</sub> fluxes at the plant or atmosphere level. In his 1927 publication, he even describes field equipment and experimental designs which are analogous to those used in the present-day "Free Air CO<sub>2</sub> Experiments (FACE)," which are probably the most sophisticated experiments we have today for the study of CO<sub>2</sub> fluxes at the field level. FACE experimenters, however, seldom refer to Lundegårdh's remarkable forerunning work.

2.2.2.2. *From the square meter scale to the hectare scale.* The "eddy covariance" (or "eddy correlation") technique is commonly used to estimate CO<sub>2</sub> fluxes at the plot ( $\geq 1$  ha) scale in continuous natural or cultivated agroecosystems. A recent and exhaustive historical review of the results obtained by using this approach is given in Baldocchi (2003). The technique can also be used at the scale of the cultivated plot (100 m<sup>2</sup>) and indeed, has been used by Reicosky et al. (1997) to study the effect of tillage on soil CO<sub>2</sub> fluxes.

### 2.3. Assessment of soil C stocks and dynamics at different scales

The importance of the soils component of an ecosystem, in terms of its influence on atmospheric GHG budgets, becomes apparent when land use, land use change and forestry (LULUCF) are considered. The published guidelines for the estimation and reporting of GHG inventories (IPCC/UNEP/OECD/IEA, 1997) recommend calculation of the net fluxes of CO<sub>2</sub> from the various C stocks in the different ecosystem compartments. The rationale of the IPCC for making this choice was that "there are large uncertainties in all current methods for estimating fluxes of CO<sub>2</sub> from forestry and land-use change. Direct measurements of changes in C stocks are extremely difficult since

one must confront the difficulty of determining small differences in large numbers as well as the inherent heterogeneity of terrestrial systems." The soil may act as a sink (by SOC accretion and CH<sub>4</sub> absorption) or a source for C-CO<sub>2</sub> in the medium term (0–50 year). There has thus emerged a growing need to: (i) quantify present SOC stocks at different spatial scales (from the plot to the continental), and (ii) predict SOC dynamics in response to LULUCF by the use of simple and robust mathematical models.

#### 2.3.1. Evaluation of SOC stocks

The content of OC, OM or humus in soil was determined as early as the beginning of the 19th century, as evidenced by Thaër's *Humus Theory* (1809). The emergence of soil C sequestration as an issue has resulted in a large effort to compile databases of SOC stocks at scales ranging from the plot to the globe. Table 2 summarizes historical data on the evaluation of SOC stocks at the global scale. The first publication was probably that of Waksman (1938) who evaluated SOC for topsoil. Later, Rubey (1951), a geologist, calculated the soil C stock for deeper soil profiles using SOC contents for nine main soil types published by Twenhofel (1926), which were based on selected values reported by Lyon et al. (1915). Rubey's estimate (710 Gt C for the 0–100 cm layer) was reasonably close to Batjes' modern (1996) result (based on 4353 soil profiles) of 1500 Gt C for same depth. Similarly, the global estimates of Waksman (1938) of 400 Gt C, for the upper 30 cm of the soils, is also close to that of Batjes' (1996) estimate of 684–724 Gt C for the same soil layer.

#### 2.3.2. The need to model SOM/SOC dynamics

The first qualitative approach for modeling SOM dynamics was by H.B. de Saussure in his famous "Voyages dans les Alpes" (1780–1796). Extracts were re-published by his son, N.T. de Saussure, in his book "Recherches chimiques sur la végétation" (1804). They were based on observations made by his father during a journey through the plain between Turin and Milan, a region that has been cultivated since antiquity. His observations and reflections can be summarized as follows:

- since no continuous accumulation of SOM occurs even with continuous organic inputs, some of these inputs must be destroyed,
- the amount which is destroyed must, to a certain extent, be proportional to the absolute existing amount,
- limits to SOM accretion must vary depending on climate, nature of mother bedrock, vegetation, cropping system and fertility of the land,
- even if all conditions are favorable to SOM accumulation, there must be a maximum for the thickness of the humus layer beyond which destructive causes equal productive ones.

H.B. de Saussure's conclusions (completely ignored by historians of soil science) thus convey the basic equilibrium

Table 2  
Publications including an evaluation of SOC stocks at the global level

Authors and Year	Number of profiles	Results for soil profile (GtC)			
		0–100 cm soil layer		Other soil layer	
		Data	Litter included: (yes/no)	Data	Depth in cm
Waksman (1938)	(n.d.)	–	–	400	(0–30)
Rubey (1951) <sup>a</sup>	9	–	–	709	(0–?)
Bohn (1976)	~200	3000	(n.d.)	–	–
Bohn (1982)	187	2220	(y)	–	–
Post et al. (1982)	2696	1395	(n)	–	–
Eswaran et al. (1993)	1000 (World) + 15000 (USA)	1576	(n)	–	–
Sombroek et al. (1993)	400	1220	(n)	–	–
Eswaran et al. (1995)	1000 ?	1576	(n)	652	(0–25)
	–	–	–	927	(0–50)
Batjes (1996)	4353	1462–1548	(n)	684–724	(0–30)
	–	–	–	2376–2456	(0–200)
Jobbágy and Jackson (2000)	2721	1502	(n)	1993	(0–200)
	–	–	–	2344	(0–300)

n.d.: Not determined.

<sup>a</sup> Rubey (1951) used SOC contents for 9 main soil types published by Twenhofel (1926) based on values reported by Lyon et al. (1915).

concepts utilized by modern mathematical models of SOM dynamics, yet it was not until 137 years later that a mathematical formulation of SOM (C or N) dynamics for the decrease in organic N content with cultivation was expressed by Jenny (1941). This was followed by the more general model on SOM dynamics of Hénin and Dupuis (1945). Many models have now been published and are in use (Smith et al., 1997). The most famous ones are probably the RothC model of Jenkinson and Rayner (1977) and the Century model of Parton et al. (1987). These models were designed to run at the plot level. Coupling them with geographical information systems (GIS) in order to simulate changes in SOC storage at scales from plot to global is an ongoing challenge faced by investigators of global change. More recently, the Global Environment Facility Soil Organic Carbon (GEFSOC) system linked the models Roth-C and Century, and the IPCC computational method, to spatial databases via a GIS (Milne et al., 2006). A first step in system development was the evaluation of the performance of the two SOC models in soil, climate and land use conditions in four case study countries (Easter et al., 2007).

### 3. Wastes and soil carbon sequestration

In this section of the paper, the authors discuss wastes and their use in the LULUCF sector in relation to soil C sequestration.

A typology of wastes useable in agriculture can be made based on their origin:

- (i) wastes from the LULUCF sector, e.g., crop residues, forestry and agroforestry residues;
- (ii) wastes from industry and/or urban activities, for example municipal solid wastes, sewage sludge and diverse industrial wastes (textiles, plastic, metals, etc.).

In the first case (wastes originating from LULUCF), the C in these wastes has a biogenic character, the initial source being atmospheric CO<sub>2</sub>. In the second case, C can be of both biogenic and fossil origin. The relative contribution of waste during a complete “C sequestration” assessment will, therefore, have to account not only for all GHGs (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O) produced during decomposition, but also for any fossil C that may be present.

#### 3.1. Estimates of soil CO<sub>2</sub> balance

Soil CO<sub>2</sub> balance can be evaluated either by: (i) direct measurement of CO<sub>2</sub> emissions from the soil (which is difficult and poses a certain number of problems on scales greater than 1 m<sup>2</sup>); or (ii) indirect methods, over different temporal scales, calculating difference in soil C stocks before and after input (surface application or incorporation) of a particular waste being studied. The second case is applicable only if it is possible to neglect additions and/or removals of C by sedimentation and erosion (Bernoux et al., 2006).

Rural waste differs from urban and industrial waste in the following way:

- *Rural waste.* As this waste is entirely biogenic in origin, an accumulation of C in the soil can be regarded as having originated from atmospheric CO<sub>2</sub>. Therefore, C accumulated in the soil as a result of the addition of rural waste can be described as “C sequestration”, provided CH<sub>4</sub> and/or N<sub>2</sub>O are not emitted during decomposition (mineralization + humification).
- *Urban and industrial waste.* For this waste, it is necessary to evaluate the fossil C content compared with biogenic C content (IPCC, 2000). Generally, sludge is considered as being entirely biogenic in origin (0% fossil C); however, this is not the case for other materials, as shown in Table 3 (modified from IPCC, 2000).

When a waste contains a high percentage of fossil C, C in soils originating from this fraction should not be referred to as sequestered C (originating from the atmosphere), but as stored C. This fraction should be omitted from any sequestration balance. It is therefore necessary to be able to estimate the amount of C remaining in the soil that has originated from fossil C. This will probably differ greatly from sequestered C of biogenic origin. Therefore, fundamental studies of the dynamics of decomposition of various types of fossil C need to be undertaken.

### 3.2. Estimates of the N<sub>2</sub>O and CH<sub>4</sub> balances for the “soil-waste” system

Regardless of the type of waste – rural, urban, industrial – with or without fossil C, it is necessary to evaluate how the waste affects the soil in terms of CH<sub>4</sub> and N<sub>2</sub>O emissions. For example, it is known that farming systems that use direct seeding, with the associated recycling of organic matter (either by mulching residues or using live cover crops) increase the risk of additional N<sub>2</sub>O emissions considerably compared to conventionally tilled systems (Six et al., 2002). In the same way, agroforestry practices imply the return of N-rich leaves to the cultivated system and can, therefore, induce large additional N<sub>2</sub>O emissions compared to non-agroforestry systems (Millar et al., 2004). In both of these cases, the amount of N<sub>2</sub>O emitted, when considered as C–CO<sub>2</sub> or CO<sub>2</sub> eq., can cancel out the benefits obtained by gross storage of C in the soil. This should be accounted for in any assessment of C sequestration (Bernoux et al., 2006). However, when establishing the final C sequestration balance, the question also arises of the fate of the waste were it not to be applied to the

soil, e.g., the reference(s) situation against which the balance is being made. For instance, if the waste was destined to be burned if it was not applied to the soil, the reference should include the GHGs that would have been emitted under this scenario. It is, therefore, necessary to take into account the whole “soil-waste” system. In this particular case, the nature of the waste, as well as the technique of incineration, should be considered. As an example, emission factors for N<sub>2</sub>O differ drastically with the type of incineration plant: for municipal waste they are 40–150 and 240–660 kg/Gg humid wastes for hearths or grates and fluidised beds, respectively. For solid sewage sludge, with the same incineration plant type, the values are 400 and 300–1530, respectively (obtained from data of IPCC, 2000).

Andreae and Merlet (2001) estimate the emission factor for the burning of plant residues to be 0.7 g N<sub>2</sub>O/kg dry matter burned. The IPCC/UNEP/OECD/IEA (1997) recommends that all GHG emissions from the burning of agricultural residues and animal waste are estimated in the ‘Energy Section’ in g N<sub>2</sub>O on an energy basis and thus not per weight of material. For the burning of savannah, the same report recommends the use of values proposed by Crutzen and Andreae (1990), which are in the range of 0.005–0.009 (mean = 0.007) kg N–N<sub>2</sub>O/kg dry matter burned.

As an example of a complete balance (Cerri et al., 2004), we will consider the case of an alternative management strategy for sugarcane (*Saccharum officinarum*) production in Brazil over the course of 1 year (Table 4). In Brazil, sugarcane fields cover almost 5 million ha, and the harvest nearly always involves a pre-harvest burn. There is, therefore, a near complete combustion of leaves,

Table 3  
Carbon content (in relation to the dry or wet weight) and % of fossil carbon for urban and industrial wastes (IPCC, 2000)

	Municipal solid wastes	Sewage sludge	Clinical waste	Hazardous waste
C Content of waste (g C/100 g waste)	33–50 (wet)	10–40 (dry)	50–70 (dry)	1–95 (wet)
Fossil carbon as % of total carbon (g C-fossil/100 g C total)	30–50	0	30–50	90–100

Table 4  
Mean annual balance for 1 ha of sugarcane managed with burning (B) and without burning (WB) in kg Ceq (negative value indicate a Ceq sink)

Component	B	WB	$\Delta$ (B-WB) (annual)	Estimated level of uncertainty <sup>d</sup>
	kg Ceq ha <sup>-1</sup> y <sup>-1</sup>			
Soil compartment (0–20 cm)	–	–	–1625	*
Litter stock	Not computed (labile compartment)			*
Annual flux <sup>a</sup> of CH <sub>4</sub>	–39	–18	21	**
Annual flux <sup>a</sup> of N <sub>2</sub> O	323	460	137	***
CH <sub>4</sub> emitted during the burning	230 <sup>b</sup>	–	–230	**
N <sub>2</sub> O emitted during the burning	140 <sup>c</sup>	–	–140	****
Total	–	–	–1837	

<sup>a</sup> Annual flux measured at the soil-litter interface with the atmosphere.

<sup>b</sup> Central value of the range (220–240 kg Ceq) of estimates.

<sup>c</sup> Central value of the range (40–240 kg Ceq) of estimates.

<sup>d</sup> Range of uncertainty (in %): \* <25, \*\* 25–50, \*\*\* 50–100, \*\*\*\* > 100.

and consequently a transformation of plant carbon into CO<sub>2</sub>, accompanied by emissions of N<sub>2</sub>O (transformation of part of the plant into N) and CH<sub>4</sub>. An alternative to this mode of management is not to burn sugarcane before harvesting. This alternative is set to become law in São Paulo state. First results indicate that the adoption of ‘without burning’ (WB) management is accompanied (during the first years) by an increase in soil carbon storage and a decrease in CH<sub>4</sub> emissions. Moreover, adopting harvesting without burning has other positive effects, e.g., an increase in the quantity and biodiversity of soil macrofauna. In addition, a decrease in nutrient losses and a reduction in the risk of erosion are observed. However, a WB management strategy involves a mechanized harvest and can have socio-economic implications. In terms of C sequestration, decomposition of the sugarcane residues, amounting to ~13 t of dry matter per year, was not complete by the end of the year and thus the soil C storage increased (Table 4). The final annual balance of the two systems shows that the WB management option is a win–win option: the soil C increases (1625 g C) and the net emissions of N<sub>2</sub>O and CH<sub>4</sub> on a C–CO<sub>2</sub> eq. basis are reduced, resulting in a benefit of 1837 g C eq. Nevertheless, this study represents a unique evaluation which needs to be confirmed. In addition, this study was carried out within a productive cycle of sugarcane and therefore did not include the effects of the replanting (which occurs every 6 year) on soil carbon dynamics.

#### 4. Conclusion

In conclusion, widespread use of the word “sequestration” seems to be a recent phenomenon (~1992). In contrast, many great experiments conducted in the past, with the concept of “sequestration” as their central hypothesis, appear to have been completely forgotten. It is the duty of modern scientists to give due credit and pay tribute to scientific forerunners who may have been forgotten, as is the case for Lundegårdh and his FACE experiment. It is also important to distinguish the classical notion of SOC storage associated solely with CO<sub>2</sub> fluxes, from the notion of SOC sequestration, which takes into consideration the total balance of the different GHG fluxes (CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, etc.) and is calculated on a CO<sub>2</sub> equivalent basis. In many cases, SOC storage differs greatly from SOC sequestration. Using such a definition, it becomes clear that the estimation of SOC sequestration for systems using waste application is difficult and that more research is needed to enable rules to be established for use in policy.

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