

Review Article

Soil Carbon Sequestration Resulting from Biosolids Application

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Carbon (C) sequestration in soils through the increase of the soil organic carbon (SOC) pool has generated broad interest to mitigate the effects of climate change. Biosolids soil application may represent a persistent increase in the SOC pool. While a vast literature is available on the value of biosolids as a soil conditioner or nutrient source in agricultural systems, there is still limited knowledge on soil sequestration mechanisms of biosolids-borne C or the main factors influencing this capacity. The emerging challenges posed by global environmental changes and the stringent needs to enhance C storage call for more research on the potential of soil biosolids incorporation as a sustainable C storage practice. This review addresses the potential of C sequestration of agricultural soils and opencast mines amended with biosolids and its biological regulation.

1. Introduction

Increasing concern about global climate change has led to growing interest in developing feasible methods to reduce the atmospheric levels of greenhouse gases (GHGs) [1]. Among GHGs, atmospheric carbon dioxide (CO₂) accounts for 60% of the global warming [2]. The atmospheric concentration of CO₂ increased from 280 parts per million (ppm) in the preindustrial era to the present 395 ppm [3]. This increase is attributed to combustion of carbon based fuels, cement manufacturing, deforestation, burning of biomass, and land-use conversion, including soil tillage and animal husbandry. Soil C capture and storage is gaining global attention because of its role as a long-term C reservoir, low cost, and environmentally friendly means to minimize climate change.

Soil C pool is the largest terrestrial C pool, constituting approximately two-thirds of the total C in ecosystems [4]. It is estimated at 2500 Pg to 1-m depth, with the biotic pool estimated at 560 Pg and characterized by fast turnover rates as compared to other natural compartments. The oceanic and geological pools are estimated at 39000 Pg and 5000–10000 Pg, respectively, whereas the atmospheric pool is estimated at 780 Pg [5]. Therefore, total soil C pool is about 4, 1 times the biotic pool and 3 times the atmospheric pool.

Soil organic carbon (SOC) includes plant, animal, and microbial residues in all stages of decomposition. SOC represents a balance between inputs, mostly via primary productivity or organic amendments, and outputs via decomposition [6]. Land application of organic amendments is a management practice that enhances SOC in the short term [7, 8]. In the last decades, the production of organic urban wastes such as biosolids has worldwide increased, and its accumulation, storage, and disposal pose growing environmental and socioeconomical problems. The safe disposal of biosolids has been a challenge for municipal wastewater treatment companies [9, 10]. In most countries, landfilling and land application have been the main disposal practices since they are the most economic outlet [11, 12]. However, sludge is a biodegradable material and releases CO₂, CH₄, and other gases when disposed in landfills [13]. Biosolids are a valuable source of organic matter and plant nutrients, especially nitrogen (N) and phosphorus (P) [14]. Recycling biosolids to agricultural land completes natural nutrient cycles. Besides, repeated land application of biosolids provides long-term benefits by increasing soil organic matter which, in turn, improves soil chemical and biological fertility [15–17]. Land application of biosolids may also result in

a decrease in bulk density, increase in pore size, soil aeration, root penetrability, soil water holding capacity, and biological activity, all of which may be reflected in an increase in crop yields [18]. One of the main disadvantages of land application of biosolids is the fact that it may contain human pathogens and trace elements, including arsenic, cadmium, zinc, copper, chromium, lead, mercury, nickel, and selenium [19–21]. The concentration of these compounds and concerns about nitrate leaching or losses to the atmosphere may limit biosolids land application [22, 23].

Soil C sequestration is defined as any persistent increase in SOC originated from removing CO₂ from the atmosphere. Increases in soil carbon storage may be accomplished by the production of more biomass. In this way, there is a net transfer of atmospheric CO₂ into the soil C pool through the humification of crop residues, resulting in net carbon sequestration [24]. Powlson et al. [25] postulated that whether SOC increases produced by applying organic residues constitute C sequestration entirely depend on the alternative fate of this material. For example, biosolids soil application represents additional carbon retention in soil if the alternative disposal method is landfilling. On the contrary, if organic wastes are regularly land applied, increases in SOC resulting from biosolids application cannot be regarded as a waste management practice to mitigate climate change. Although many studies have examined the potential value of biosolids to increase soil productivity, relatively limited work has examined its effect on soil C sequestration [8, 26, 27].

2. Biosolids-Borne C Sequestration Capacity of Soils

2.1. Agricultural Soils. Soil C sequestration capacity reflects soil aptitude to retain and stabilize C. In the light of global change scenario, soil humification mechanisms have acquired renewed interest. Traditionally, the stability of organic compounds has been regarded as the main process controlling SOC retention, but recent analytical and experimental advances have demonstrated that molecular structure alone does not explain SOM stability [28]. The amount of SOC accumulated does not only depend on the quantity and quality of the organic inputs, but on the physicochemical and biological influence of the environment as well [28, 29]. Various mechanisms of SOM stabilization have been proposed [27]. Physical protection by incorporation into soil aggregates or by occlusion in micropores inaccessible to soil microorganisms or chemical protection through interaction with mineral surfaces is considered important mechanisms to reduce the bioavailability and accessibility of SOM for soil microorganisms or hydrolytic soil enzymes [30]. Another mechanism of SOC stabilization is assumed to be biochemical protection. Biotic organic C is biologically transformed by the action of microbial populations (i.e., bacteria, fungi) and invertebrates, resulting in acquired chemical stability [31]. Therefore, the residence time of organic compounds in soils is directly related to intrinsic or developed resistance to further microbiological degradation.

Biosolids are typically made up of 40–70% organic matter, with organic C content ranging from 20–50%, total nitrogen

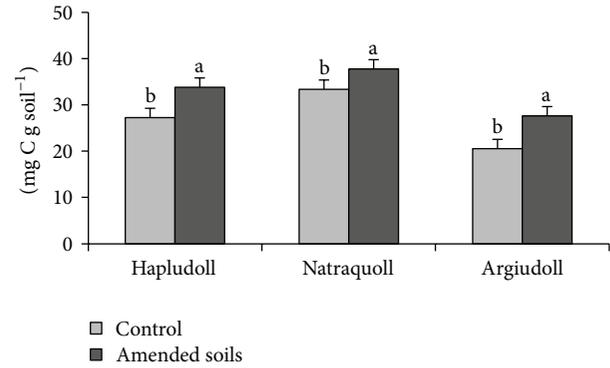


FIGURE 1: Storage capacity of biosolids-borne C of typical soils of the Pampas region, Argentina, 360 days after biosolids application (application rate of 150 dry t/ha). Different letters for the same soil indicate significant differences at the 0.05 probability level ($n = 4$). (Adapted from Torri et al. [34]).

(N) ranging from 2–5%, and a C/N ratio of about 10–20 [32, 33]. Biosolids' organic matter has usually undergone some degree of stabilization through anaerobic or aerobic digestion before being land applied. Long- and short-term observations have demonstrated that biosolids amended soils accumulate a significantly higher amount of organic C compared to mineral fertilized soils. Over a 34-year reclamation experiment, Tian et al. [8] reported that soil C sequestration capacity in agricultural soils under conventional tillage was significantly correlated with biosolids application rate. In the short term, Torri et al. [34] reported soil C accumulation in three pristine representative soils of the Pampas region, Argentina, amended with high biosolids rate. These soils had different particle size distribution, although the clay fraction had the same origin and mineralogical composition. During the 30 days following biosolids incorporation, decomposition mechanisms associated with an intense biological activity originated a rapid CO₂-C release [35]. After 150 days, the three pristine amended soils appeared to reach a new equilibrium, with C contents being significantly higher than the respective controls. Total C content did not change significantly between days 150 and 360 in the three studied soils, showing the potential of these soils to store biosolids-borne C (Figure 1).

Many mathematical models have been proposed to describe the decomposition process of land applied organic materials, ranging from one to multicompartiment models [29, 36, 37]. In the study made by Torri et al. [34], a first-order exponential model provided the best fit to C mineralization data for the three soils (1):

$$\%C_B [t] = C_{RB} + C_{LB}e^{-kt}, \quad (1)$$

where %C_B is the percent residual C from biosolids in soil; %C_{LB} the initial percent of biosolids borne-C in the labile pool; %C_{RB} the percent of biosolids borne-C in the resistant pool; k is the first order rate constant (day⁻¹); t is the time after biosolids application (days).

According to this model, organic C added through biosolids consisted of two fractions of different degree of biodegradability: a labile fraction (53–71%) that was quickly

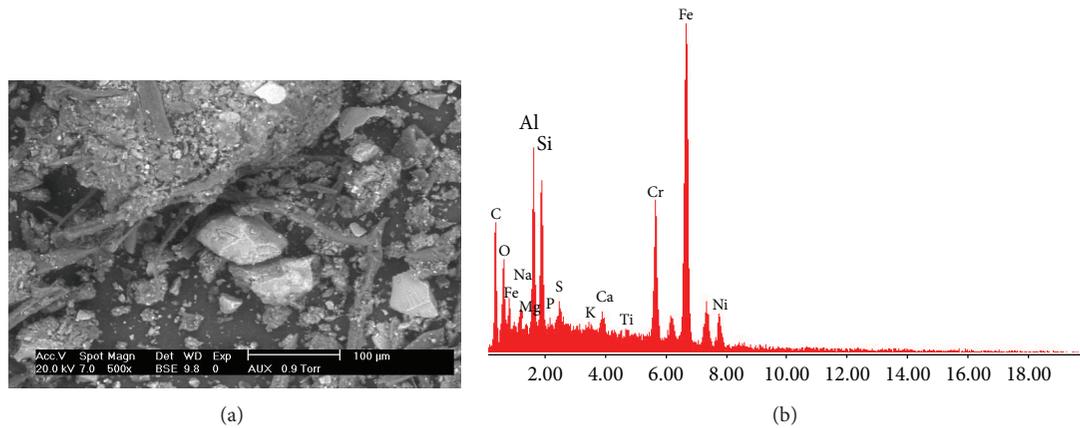


FIGURE 2: Scanning electron microscopy with X-ray microanalysis (SEM-EDS) image of biosolids (a) and X-ray diffraction (XRD) patterns of biosolids (b).

mineralized at a constant rate (k) and a recalcitrant fraction (28.5–45.4%), not available or resistant to soil microorganisms that remained in the soils one year after biosolids application [34]. Organic compounds of the recalcitrant fraction are mainly stable cholestane-based sterols [38] which may have a turnover rate in the order of hundreds of years [39]. The labile organic fraction has often been found to be largely dependent on the degree of biosolids stabilization [40].

In general, C mineralization rates of freshly added organic sources have been found to be more rapid in soils with low than with high clay content [41]. An explanation to this could be that residual substrate and decomposition products become stabilized by sorption onto mineral clay particles, by incorporation into pores that are too small for bacteria or fungi to penetrate. Consequently, they are physically inaccessible to microbial turnover [42, 43]. Furthermore, organic matter may be protected against decomposition as a result of being inside large aggregates that become partially anaerobic because of slow O_2 diffusion through the small intra-aggregate pores [44].

Despite the protection provided by clay particles, many studies reported that mineralization rates of biosolids-borne C were not related to soil texture [34]. Strong et al. [45] indicated that clay protects the organic matter which had time to become entangled in the soil matrix, although this may not happen if a great volume of fresh material is added. Moreover, the turnover of organic matter in soils of different texture was better explained by other soil parameters, such as temperature, moisture, pH, redox condition, and nutrient availability [46–50]. In the study of Torri et al. [34], the water content of the three studied soils was periodically adjusted according to water holding capacity and did not limit microbial activity. In this case, soil pH was the dominant variable on soil decomposition of biosolids. Most studies reported that acidic pH delayed the decomposition of soil organic matter because of its profound influence on biomass, activity, and composition of the soil microbial community [51, 52]. The higher pH of the Natraquoll stimulated microbial activity, increasing carbon mineralization of incorporated biosolids, whereas slightly acidic soils retained more biosolids-borne C (Figure 1).

In turn, the effects of microbes on composition and recalcitrance of SOC have been shown to be as important as climatic and edaphic characteristics [53]. Soil aggregates are held together by mucilages and by fungal hyphae, roots, and polysaccharides. Increased amounts of any of these agents would promote the formation of macroaggregates, which, after further decomposition, form the stable and more resistant core of microaggregates [54, 55].

X-ray fluorescence analysis of dried sludge indicated that oxides of Si, Al, and Fe were the three main inorganic constituents of biosolids [7, 56]. X-ray diffraction analysis of biosolids performed by a number of researchers also indicated the presence of quartz, feldspars, kaolinite, mica, and expandable clays [57, 58]. In Figure 2(a), the scanning electron microscopy with X-ray microanalysis (SEM-EDS) image of a biosolids sample showed the presence of both organic and inorganic phases [7]. The inorganic phase included silica, Fe, Al, and calcium compounds (Figure 2(b)). Taking into account that soil organic matter carries negative charges, whereas Fe oxides have positive ones, under certain conditions both of them may be intimately associated [59]. Biosolids-borne amorphous iron and aluminum oxides would promote SOM stabilization after biosolids application [60, 61]. Amorphous Fe and Al oxides are very reactive due to their small size and high surface area. These reactive surfaces are often presumed to account for sorption and stabilization of OM [62]. More recent research has suggested that stabilization mechanisms include ternary C - Fe oxide - clay associations [63] and the formation of unidentified chemical bonds [64]. Lines of evidences that the association of organic matter with secondary hydrous Fe and Al phases would prevent SOC degradation have been published [65, 66]. Taking into account that biosolids are rich in amorphous Fe/Al oxides [60, 67], long-term biosolids application could increase soil amorphous Fe/Al, leading to increased SOM stability.

Some authors indicated that “the capacity of terrestrial ecosystems to store carbon is finite and the current sequestration potential primarily reflects depletion due to past land use” [68]. However, recalcitrant organic carbon and amorphous Fe/Al oxides are land applied through biosolids amendments. The presence of Fe/Al oxides has been shown

to stabilize soil organic matter and, likely, biosolids borne organic matter [66]. Moreover, Kögel-Knabner et al. [69] suggested that one possible reason for soil C immobilization in these soil amendments could be its adsorption or complexation by Fe and Al cations. These results may indicate that, with time, biosolids amended soils would be able to store more carbon than the one depleted due to past land use. Nevertheless, this hypothesis requires further investigation.

2.2. Opencast Mine Sites. In recent years, there has been growing evidence that biosolids may be used to restore mine spoils or tailings. Rehabilitation of rocky materials exposed by mining typically involves physical amelioration and organic matter incorporation. The use of biosolids in such operations represents an opportunity to couple biosolids application with soil C sequestration. In several cases, this practice has led to high soil C accumulation, edaphic improvement, and an effective vegetation cover establishment [70, 71]. Furthermore, organic matter inputs and revegetation lead to humus accumulation and pedogenesis, including soil profile development [70, 72–74]. As the system evolved, grasses progressively dominated and represented the most important source of SOC in biosolids amended soils [72]. After a decade of biosolids incorporation, SOC reached concentrations comparable to those found in hydromorphic soils and about twice those of undisturbed mineral soils under primary forests [70].

Mined spoils in tropical regions present factors that can accelerate C flow, such as high temperatures, acidity, and low charges surfaces, and factors that may retard it, like clay texture, overburden materials, and anaerobic conditions due to waterlogging. Carbon accumulation in revegetated mining spoils suggests that retarding factors have prevailed under stripped soil situation. Common inorganic components present in mining spoils include aluminosilicates (allophane and imogolite) and Fe-(oxy) hydroxides (like ferrihydrite). Both of them have been reported to stabilize soil C [75, 76]. Since vegetation coverage is usually established on low-permeable rocky material in mining sites, roots are usually subjected to waterlogging, leading to SOC accumulation [77]. Several hypotheses about deleterious effects on soil microbial activity by compounds formed under anaerobiosis, such as volatile fatty acids, H_2S , and toxic concentrations of NH_3 , Al, Fe, and other elements, have been postulated [78].

Although mining spoils are not a suitable environment for plant development, the incorporation of biosolids has promoted plant colonization and unprecedented organic-C accumulation was reported in temperate and tropical regions [70, 79]. Depending on site management, Shrestha and Lal [80] estimated that C sequestration rates for reclaimed mine soils could be in the order of 0.1–3.1 and 0.7–4 $Mg\ ha^{-1}\ y^{-1}$. Scaling up such figures, mining site restoration could account for $\approx 16\ Tg\ CO_2$ per year [80].

3. Biological Regulation of C Storage in Biosolids Amended Soils

Soil organic C dynamic depends on microbial activity, community composition, and soil enzyme activity [81]. While

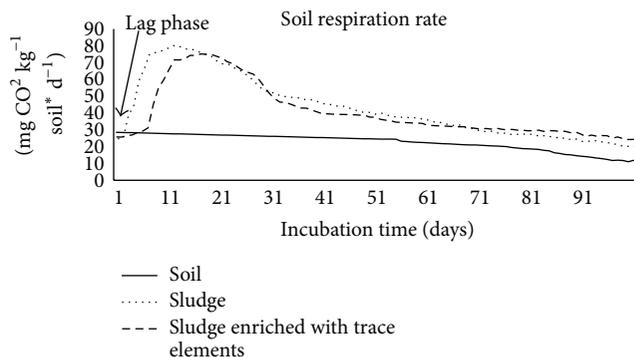


FIGURE 3: Typical respiration trends of soils amended with sewage sludge enriched or not with trace elements ($Cd\ 12\ mg\ kg^{-1}$, $Zn\ 300\ mg\ kg^{-1}$). A lag phase can be observed before the onset of the C flush for metal spiked sludge.

incorporation rates and quality are relevant factors controlling soil organic C inputs, land application of biosolids may change the activity and diversity of soil microbial communities [82, 83]. Consequently, the use of biosolids as a soil amendment may have variable effects on soil C retention.

3.1. Microbial Activity. Most of the existing information on soil microbial responses due to biosolids incorporation is related to the use of sewage sludge [84–86]. Contrasting responses—from stimulation to inhibition—have been reported. These results reflect the large variability of sludge chemical composition and the level of resolution of the biological studies (i.e., biochemical or biomolecular) [87–89].

In most soils, microbial activity and proliferation is typically C-limited whereas N limitation hardly occurs [90]. Biosolids application modifies soil main regulating parameters, particularly the nutrient stoichiometry. Sludge incorporation into soils provides labile C that produces a large initial C- CO_2 flush due to rapid labile organic matter decomposition and microbial growth. This respiratory response is known as the “C dominated phase” (Figure 3) and is invariably bell-shaped. The shape of the curve is generally related to N availability, the quality of biosolids’ organic matter, and the release of trace elements or organic pollutants through biosolids decomposition [87]. On the contrary, soil application of composted or thermally dried sludge may reduce soil microorganisms decomposition rates due to the higher proportion of recalcitrant C added [89].

Before the onset of CO_2 release, an increase in the lag time may be observed in sludge amended soils [91]. The lag phase allows the adaptation required for bacterial cells to begin to exploit new environmental conditions [92] and has been suggested to be a sensitive microbial indicator of soil metal pollution [93]. Sludges with high content of trace elements may result in a longer “lag phase,” as depicted in Figure 3. However, this may not be a universal microbial response [94].

There is still considerable controversy about the effects of biosolids applications on the native SOC pool [95]. Some authors reported that the strong microbial activity induced by the application of biosolids with high contents of labile

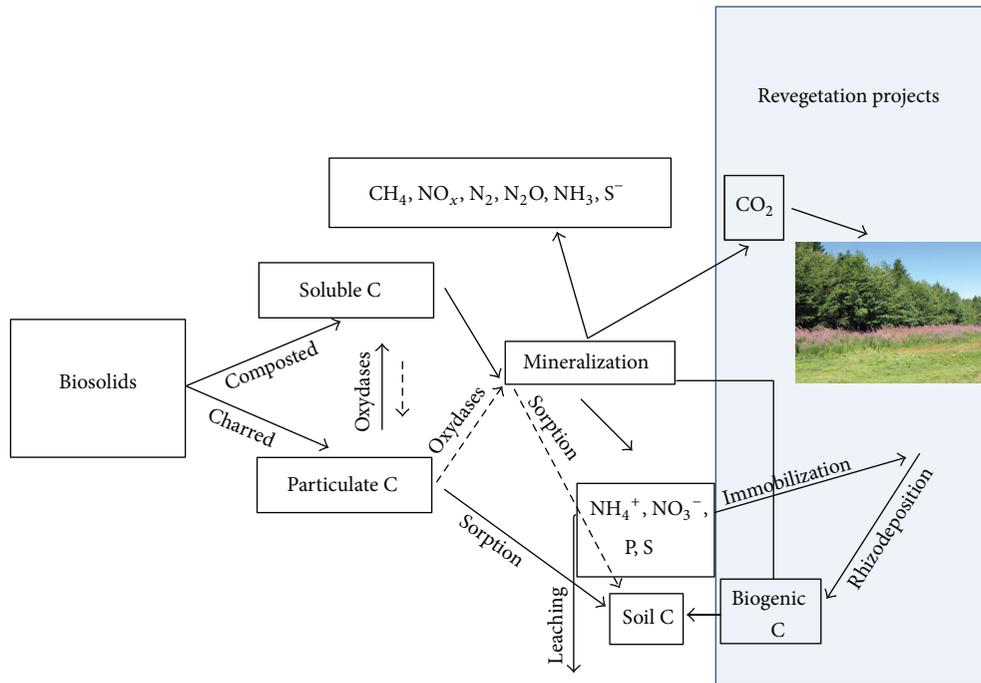


FIGURE 4: Schematic contribution of biosolids-borne C to SOC fractions and to soil nutrient fluxes, in addition to biogenic extra C inputs in mine spoils revegetation projects. The dotted lines indicate the still poorly understood effect of oxydases on particulate C mineralization or the stabilization of mineralized soil C by chemisorption.

organic substances would mineralize native SOC [96]. This effect was mainly noticed when biosolids were applied to soils containing high initial levels of SOC, a phenomenon known as *priming effect* [97]. Conversely, the application of the same amount and type of biosolids in soils with low levels of initial SOC contributed, in general, to an increase in the short-term SOC pool. An explanation to this was the low density of microbial communities in soils with low organic matter content. Although positive and negative priming effects have been reported, the latter does not always result in reduced soil C storage capacity. In general, the amount of added organic amendment entirely compensates the priming-induced C loss [97]. Soil microorganisms can be triggered into activity by the availability of water and nutrients such as those present in the root exudates [98], and therefore these factors are important covariates to be accounted for assessing the biological control of biosolids soil C storage. Theoretically, C storage is achievable when the metabolic energy required to decompose biosolids-borne organic substrates is larger than the energy obtained by catabolism within microbial cells.

Soil management practices may influence soil C storage after biosolids amendment. It has been widely reported that intensity of mineralization processes is much higher in soil surface layers than in deepest soil horizons [99]. However, root exudates or decomposition of root litter in subsurface horizons may induce priming effects that may, in turn, enhance the decomposition of biosolids-borne organic C. Low molecular weight organic compounds present in root exudates support large microbial biomass in the rhizosphere and induce SOM mineralization [99, 100]. Therefore, the current information on the priming effects on native SOM

cannot be extended to biosolid amended soils. The reason for this is that the overall effects will not only depend on soil's characteristics and basal microbial activity, but on the nature of nutrient inputs as well. Therefore, the biosolid-borne C storage capacity of soils will depend on the site specific equilibrium between C inputs and outputs, regulated by mineralization processes. Nevertheless, it is undisputable that, on the long term, biosolids amended soils retain more C than soils under conventional agricultural regime in different tillage, fertilization, and rotational schemes [101]. The same applies for marginal and degraded soils [102].

3.2. Soil Enzyme Activity. Soil enzyme activity is responsible for most of the soil functionality, as it promotes the decomposition of organic matter and release nutrients in forms available to plants and microorganisms. Soil enzymes are actively released by proliferating soil microorganisms, plant roots, and soil fauna or passively released by dead microorganisms and root cell sloughing. A large fraction of the enzymes released in the extra- or pericellular space are stabilized by stable soil organic and inorganic phases [103], which protect them from soil proteolytic activity [104]. Biosolids amendments play a dual role on soil enzyme activity: biosolids may increase the stabilization of extracellular enzymes through their solid phase surface properties or they may provide substrates (i.e., proteins and peptides) for soils enzymes, leading to microbial proliferation and the increase of enzymatic activity. This aspect is important for the long term of biosolids-borne C storage in soils because the presence of active enzymes reduces energy barrier to the decomposition of complex organic substrates thus facilitating the

biosolids matrix mineralization without new and metabolically expensive synthesis of extracellular enzymes [105]. This may produce a long-term positive feedback effect on soil microbial activity after amendment (Figure 4).

In any case, the presence of soil stabilized “enzymatic background” makes soil’s decomposition capacity only partially related to the actual levels of microbial activity in biosolids amended soils. As it is currently impossible to estimate the ratio between intra- and extracellular enzyme activity, it is not possible to precisely determine the actual contribution of active microorganisms to biosolids degradation.

4. Conclusions

In the light of global change scenario, increasing soil organic matter stocks has been identified as a feasible way for soil C sequestration. Many experiments indicated that application of biosolids to land or opencast mines resulted in an increase in carbon reserves of soils from different regions and under different management practices. Biosolids are typically made up of 40–70% organic matter, consisting of two fractions of different degree of biodegradability: a labile fraction that is quickly mineralized by soil microorganisms and a recalcitrant fraction, not available or resistant to soil microorganisms, responsible for soil organic carbon accumulation. The amount and proportion of recalcitrant C in biosolids are important attributes to predict the biological control of C storage in soils. It seems to be a direct relationship between C recalcitrance and mineralization: the higher the C recalcitrance in biosolids amended soils, the higher the metabolic energy needed for biosolids mineralization by soil microorganisms. Monitoring microbial communities and soil enzyme activity may be used as ecological indicators of biosolids C stabilization in soil.

Many studies reported that mineralization rates of biosolids-borne C may not depend on soil texture and that slightly acid soils retained more biosolids-borne carbon than soils with a higher pH. Furthermore, amorphous iron and aluminum oxides usually found in biosolids would play an important role in soil organic C accumulation. Therefore, the capacity of soils to sequester biosolids borne C may not be finite. It is important to remark that the benefits associated with the use of biosolids for soil carbon sequestration are in addition to other benefits, like the improvement of soil quality in terms of physical, chemical and biological fertility, although the presence of contaminants may impact soil microbial communities on the long term.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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