

Weathered Fly Ash Does Not Affect Soil And Biosolid Carbon Mineralization

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Abstract: Fly ash and biosolid wastes can be mixed and applied to soil as a means of disposal. A significant decline in soil respiration following waste application indicates restricted activities of functional microbial populations. Weathering decreases salinity and neutralizes alkalinity in fly ash, but there is little information on the effects of unweathered fly ash and biosolid mixtures on soil carbon (C) mineralization. The objective of this study was to determine the effects of a weathered fly ash–limestone scrubber residue (LSR) mixed with an aerobically digested biosolid on soil respiration in a laboratory incubation study. Biosolids significantly increased carbon dioxide (CO₂) production ($p < 0.05$), but up to 6.75% (w/w) fly ash did not. Mean total C mineralization was 770 mg CO₂-C kg⁻¹ soil in the control and 3,810 mg CO₂-C kg⁻¹ soil in the 6.75% (w/w) biosolid treatment. Fly ash with neutral pH and low salinity appears unlikely to affect soil and biosolid C mineralization.

Keywords: Biosolid, carbon mineralization, fly ash, incubation, respiration

INTRODUCTION

Coal-burning power plants in the United States are required to outfit their stacks with a sulfur dioxide (SO₂) scrubbing unit, which captures SO₂ into a solid waste before it exits to the atmosphere. A mixture of

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captured sulfur-containing particles and fly ash (coal combustion residue), referred to here as fly ash–limestone scrubber residue (LSR), accumulates adjacent to the boiler. The U.S. electrical utility industry generated about 71.2×10^6 Mg of coal fly ash in 2001 (USEPA 2003).

Approximately 35% of fly ash–LSR is used for construction-related applications; the remaining requires disposal. Soil application has been shown to improve soil physical properties such as texture (Chang et al. 1977), aeration (Aitken and Bell 1985; Chang et al. 1977), and water-holding capacity (Chang et al. 1977). Fly ash has also been used to increase the pH of acidic soils (Elsewi and Page 1984; Petruzzelli, Lubrano, and Cervelli 1987; Plank, Martens, and Hallock 1975) and to relieve deficiencies of plant nutrients such as boron (B), calcium (Ca), molybdenum (Mo), and sulfur (S) (Mulford and Martens 1971; Plank and Martens 1974; Wallace et al. 1980; Wallace and Wallace 1986).

Biosolids, semisolid, nutrient-rich by-products from wastewater treatment facilities, are also routinely applied to land. The U.S. biosolid production in 1998 was 6.9×10^6 Mg (dry), of which 41% was applied to land (USEPA 1999). Fly ash and biosolids can be mixed and applied to soil to reduce pathogens and heavy-metal availability in biosolids (Fang, Wang, and Wong 1999; Jiang et al. 1999; Su and Wong 2003; Valls and Vasquez 2000; Wang and Viraraghavan 1998; Wong 1995; Wong, Jiang, and Su 1996) and to enhance the availability of biosolid plant nutrients (Sajwan et al. 2003; Wong and Su 1997).

Fly ash/biosolid mixtures may change the respiration rates in soil depending on the application rate and nature of each waste. A decline in carbon dioxide (CO₂) release (or an increase in O₂ consumption) indicates that substances (e.g., heavy metals) contained in the fly ash/biosolid mixture inhibit microbial growth or that the mixture changes soil chemistry [e.g., pH, electrical conductivity (EC)] to conditions unfavorable for metabolically active microorganisms. Heavy metals and elevated EC can inhibit microbial activities in soil (Baath 1989; Eary et al. 1990; Pichtel 1990). Many soil enzymes are pH dependent (McCarty et al. 1994). Carbon mineralization studies with fly ash–LSR–treated soils have reported a decline in respiration rates (Arthur et al. 1984; Pati and Sahu 2004), although at very low application rates (<2.5%), fly ash may be noninhibiting (Pati and Sahu 2004).

Biosolid co-application raises microbial respiration in fly ash–treated soils to levels close to those of control soil (Pichtel 1990; Pichtel and Hayes 1990). Unlike fly ash, some biosolids contain readily available organic C, which increases microbial numbers and enzyme activities in soil (Coppola 1983; Hattori 1988; Varanka, Zablocki, and Hinesly 1976). Large heterotrophic populations maximize C mineralization within a few days following biosolid amendment. As soil microorganisms utilize readily available biosolid organic C, respiration rates decline (Hsieh,

Douglas, and Motto 1981). Aerobically digested biosolids tend to yield more mineralized C than anaerobically digested biosolids (Hsieh, Douglas, and Motto 1981).

There is limited information on the effects of unweathered fly ash/biosolid mixtures on C mineralization in soil, and there is almost no information regarding weathered fly ash/biosolid mixtures. Weathering processes decrease salinity in fly ash-LSR by carbonation, which forms Ca and Mg carbonates. These processes also neutralize alkalinity in fly ash-LSR. The objective of this study was to determine the combined effects of a weathered fly ash-LSR (EC, 2.45 dS m⁻¹; pH, 7.8) and an aerobically digested biosolid on C mineralization in soil.

MATERIALS AND METHODS

Soil, Biosolid, and Fly Ash-LSR Characterization

The samples were analyzed at the Regulatory Services Soil Testing Laboratory of the University of Kentucky by standard methods (Soil Survey Staff, 1992). Chemical and physical properties of the soil, biosolids, and fly ash-LSR are given in Table 1. The soil used in the incubations was Pembroke silt loam (a fine silty, mixed, mesic Mollic

Table 1. Properties of soil, biosolids, and fly ash-LSR after drying

Constituent	Soil	Biosolids	Fly ash-LSR
Total C (%)	1.8	24.8	0.58
Total N (%)	0.22	2.89	BD
pH	7.5	7.1	7.8
CEC (meq 100 g ⁻¹)	5	ND	ND
EC (dS m ⁻¹)	1.25	7.35	2.45
Metals (mg kg ⁻¹)			
Cd	0.4	0.3	0.6
Cr	BD	BD	2.0
Ni	0.7	14.5	0.4
Pb	8.0	8.0	4.0
Cu	4.0	6.0	7.0
Mo	0.1	0.1	8.1
Zn	8.0	22.0	18.0
B	< 0.05	< 0.02	50.0
Texture (%)			
Sand	6.8	ND	ND
Silt	67.9	ND	ND
Clay	25.2	ND	ND

Notes. ND, not determined; BD, below detection.

Paleudalf) from Bowling Green, Ky. Bulk soil samples were collected to a depth of 15 cm, placed into plastic containers, sealed with plastic lids, and transported to a greenhouse, where they were stored for approximately 1 year. The soil was sieved through a 1-cm metal sieve and air-dried for 1 week. Dried soil samples were returned to their original containers and stored three additional months in the greenhouse, then sieved through a 2-mm metal sieve before use.

Aerobically digested biosolids were obtained from Bowling Green Municipal Utilities, Bowling Green, Ky. They were transported to the field and stored as a mass beneath a plastic cover for 4 months before sample collection. The samples were transported to a greenhouse in plastic containers and stored for 9 months. Before use, biosolids were air dried for 1 week, ground, and passed through a 2-mm metal sieve.

The fly ash-LSR was collected from a pond at the Paradise Power Generation Plant, Muhlenburg, Ky. It was transported to the field, where it was stored as a mass beneath a plastic cover for 2 months before sample collection. The fly ash-LSR was transported to a greenhouse in plastic containers and stored there for 13 months. The fly ash-LSR was air dried for a week and sieved through a 2-mm metal sieve before use.

Incubation Design

The experiment was a complete factorial design with three replications, in which 200 g of dry soil was combined with dried biosolid and fly ash-LSR at the selected rates (0, 0.25, 0.75, 2.25, and 6.75%; w/w) and mixed for 5 min in a mechanical mixer. The total number of treatments was 25 (5 biosolid rates \times 5 fly ash-LSR rates). A 50-g mixture from each treatment was placed into a plastic bag. Distilled water was added to achieve 20% gravimetric water content in the samples, which corresponded to 60% water-filled pore space in untreated soil. Each treated and moistened soil was placed into a 500-mL mason jar, sealed with a gas-tight metal lid penetrated by a butyl rubber stopper, and incubated at 35 °C for 28 days. Distilled water was periodically sprayed onto the incubating soils to maintain the initial water content. Because the available time for CO₂ measurement on a given sampling day was limited, we performed three separate incubations with 14-day intervals between them.

Carbon Dioxide (CO₂) and Oxygen (O₂) Measurements

Headspace was sampled with a 1-mL gas-tight tuberculin syringe at 1 day intervals for the first 6 days and at 2- to 5-day intervals for the remainder

of the incubation. The mason jars were vented for approximately 5 min following each sampling to maintain O₂ concentrations greater than 10% for the incubation. Gas samples were analyzed on a gas chromatograph equipped with a thermal conductivity detector (TCD) operated at 120 °C and a filament temperature of 140 °C. Helium (He) was the carrier gas ($V_{\text{flow}} = 20 \text{ mL min}^{-1}$). A 2-m Porapak Q column at a temperature of 60 °C was used for CO₂ separation and analysis. A 2-m molecular sieve 5A column in line with the Porapak Q column at 60 °C was used for O₂ analysis. Although CO₂ concentrations were measured in all incubations, the O₂ concentrations were measured in the first incubation only.

Statistical Analysis

An exponential function, $y = a \ln(x) + t$, best described the relationship between C mineralization rate (y) and time (t) in the current study. After this function was applied, coefficient a and intercept c for each treatment were determined. An analysis of variance (ANOVA) was performed on these coefficients and total C mineralization values to determine significant ($p \leq 0.05$) treatment effects.

RESULTS AND DISCUSSION

Total C Mineralization

Soil, biosolid, and fly ash–LSR storage can be regarded as a treatment effect uniformly applied across all treatments. The blocking effect (incubations) was also significant ($p \leq 0.05$), which can be attributed to temperature fluctuations during the incubations. Respiration was not limited by oxygen, because the O₂ concentrations in the mason jars remained greater than 10% with the exception of day 1 in the 2.25 and 6.75% biosolid treatments (Figure 1). The mean total C mineralization values for the 30-day incubation are given in Figure 2. As biosolid rate increased, total C mineralization significantly ($p < 0.05$) increased, rising from 770 mg CO₂-C kg⁻¹ soil in the control to 3810 mg CO₂-C kg⁻¹ soil in the 6.75% biosolid treatment.

Total C mineralization values were compared with reported values from other studies in Figure 3. The total C mineralization values in the current work are similar to the results from a study that also used an aerobically digested biosolid (Hsieh, Douglas, and Motto 1981). However, this does not always hold true, because some wastewater treatment plants add chemicals to the biosolids, including lime [Ca(OH)₂], alum [Al₂(SO)₄], and ferric chloride [FeCl₃] to flocculate

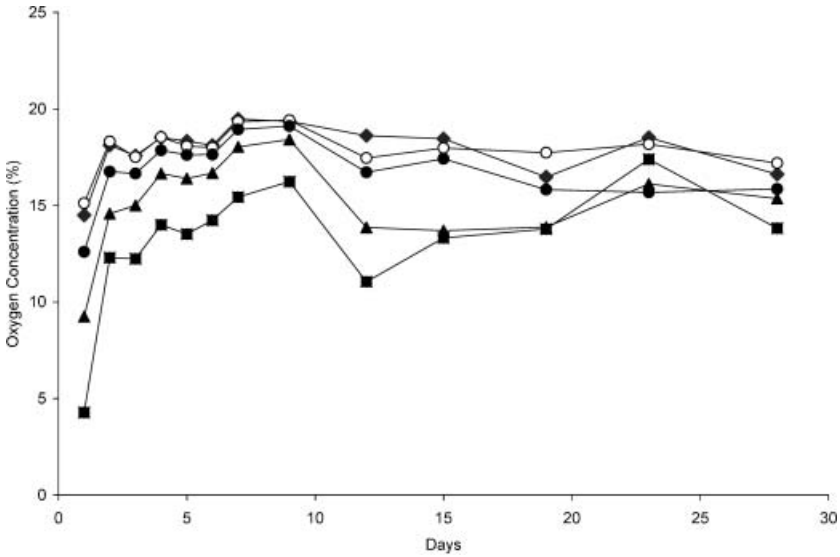


Figure 1. Oxygen (O₂) concentrations in the mason jars, which contained 0% (◆), 0.25% (○), 0.75% (●), 2.25% (▲), and 6.75% (■) biosolids during the first incubation. Data are averaged across all fly ash-LSR rates.

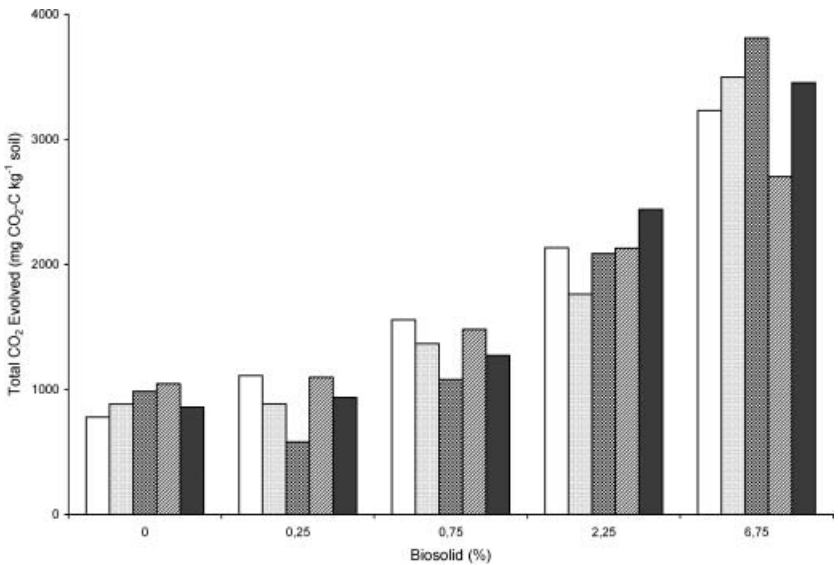


Figure 2. Total C mineralization in the mason jars which contained 0% (blank), 0.25% (dots), 0.75% (squares), 2.25% (slices), and 6.75% (shaded) fly ash-LSR during a 30-day incubation. Data are the mean of three replications.

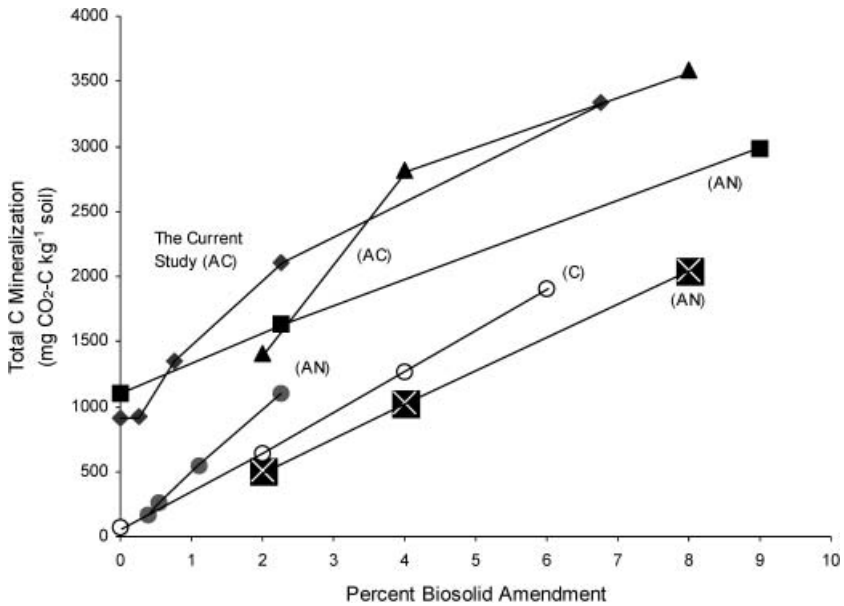


Figure 3. Comparison of total C mineralization data from the current and comparable studies. Data from the current study; Boyle and Paul (1989); Hsieh, Douglas, and Motto (1981); Terry, Nelson, and Sommers (1979); Tester et al. (1977). AC, aerobically digested biosolids; AN, anaerobically digested biosolids; C, composted biosolids.

and condition the biosolids or to remove P from the effluent (Tester and Parr 1983). These chemicals increase the EC in biosolids and soil amended with such biosolids. Carbon mineralization in soil is greater when this salinity is removed from the biosolid (Tester and Parr 1983). The extent and nature of industrialization in the sanitary district and the seasonal variability of wastewater entering the wastewater treatment facility also contribute to the variation in chemical composition of biosolids (Sommers, Nelson, and York 1976).

The fly ash-LSR did not significantly ($p > 0.05$) affect total C mineralization in this study. Earlier work (Pichtel 1990; Pichtel and Hayes 1990), however, showed that soil C mineralization significantly ($p \leq 0.05$) declined, especially in a poorly buffered soil, with the addition of an unweathered fly ash (5%, w/w) that had an extreme pH (12.2). Biosolid co-application (5%, w/w) increased C mineralization values to levels similar to those of the control soil in these studies, but only for well buffered soil. Arthur et al. (1984) also showed that a fly ash with high heavy-metal content significantly ($p \leq 0.05$) decreased soil C mineralization when the application rates were more than the greatest rate in our study. Soils with high buffering capacities appear to be preferable for

unweathered fly ash-LSR disposal because of the potential adverse effects on C mineralization of fly ash's alkalinity and salinity. Unlike Pichtel (1990) and Arthur et al. (1984), we used a weathered fly ash-LSR with a neutral pH, low heavy-metal contents, and a soil with relatively high buffering capacity (Table 1), so the potential adverse effects of fly ash-LSR on C mineralization were minimal.

Pattern of C Mineralization during the Incubations

The relationship between time and mean C mineralization rates in a replication (the first incubation) is given in Figure 4. Only the results from the first incubation are given because the sampling days differ among the incubations. The C mineralization rates were relatively high at the beginning of the incubation due to the labile biosolid organic C addition (Hattori 1988; Hsieh, Douglas, and Motto 1981; Neilson and Pepper 1990; Terry, Nelson, and Sommers 1979), redistribution of soil organic matter by sieving (Beare et al. 1994; Ross et al. 1985), and suitable water and temperature regimes for microbial activity (Blet-Charaudeau, Muller, and Laudelout 1990). Respiration rates exponentially declined for 2 weeks and stayed relatively constant thereafter. Similar trends have been observed in previous studies (Hattori 1988; Hsieh, Douglas, and Motto 1981; Terry, Nelson, and Sommers 1979).

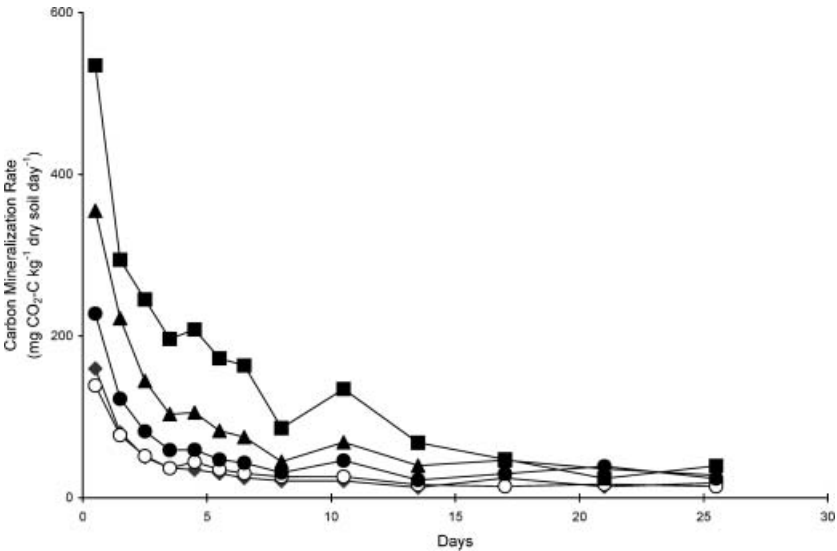


Figure 4. Carbon mineralization rates in the mason jars that contained 0% (◆), 0.25% (○), 0.75% (●), 2.25% (▲), and 6.75% (■) biosolids: first incubation. Data are averaged across all fly ash-LSR rates.

The C mineralization pattern was characterized by exponential function (1):

$$\text{CMR} = a[\ln(t)] + c \quad (1)$$

where CMR is C mineralization rate at time t , and a and c are constants specific to each treatment. This function best described the relationship between time and CMR in the treatments. The R^2 values for these regressions ranged from 0.78 to 0.93. Biosolid amendments significantly increased the absolute values of a and c ($p < 0.01$). Fly ash-LSR did not have a significant effect ($p > 0.05$), nor was there a significant interaction between biosolids and fly ash-LSR ($p > 0.05$).

The biosolid rate that maximized total C mineralization was determined in the following manner. We assumed that coefficient a values indicated how quickly biosolids decompose in soil. An increase in a indicates that the overall rate of biosolid decomposition increases. The percentage biosolids application rate was converted into a field application rate. The mean a values for each biosolid rate were pooled across fly ash-LSR rates and plotted against field biosolid application rates. This plot best fit a second-degree polynomial model (2):

$$y_a = -0.0041x^2 + 1.263x + 30.24 \quad (R^2 = 0.99) \quad (2)$$

where y_a is coefficient (a) and x is biosolid rate (Mg ha^{-1}). The biosolid application rate that maximized CMR was determined by taking the derivative of y_a and calculating x in the derivative formula where $y_a' = 0$. The biosolid rate maximizing the mineralization was 154 Mg ha^{-1} . The greatest biosolid application rate in our study corresponds to 135 Mg ha^{-1} , so speculation for rates more than 154 Mg ha^{-1} using Eq. (2) could be erroneous.

Percentage of C Mineralization in Biosolid

Approximately 2, 26, 24, and 17% of the organic C in biosolids evolved as $\text{CO}_2\text{-C}$ in the 0.25, 0.75, 2.25, and 6.75% biosolid treatments, respectively. We expected the greatest percentage of C mineralization at the smallest biosolid level because the percentage of C mineralization has declined as the biosolid application rate increased in earlier works (Figure 5). In the 5 Mg ha^{-1} biosolid treatment (0.25% biosolids, w/w), organic C in soil was approximately three times higher than that added by biosolids. Consequently, the CO_2 analysis probably was not sensitive enough to differentiate between the CO_2 attributable to soil and biosolids in this treatment.

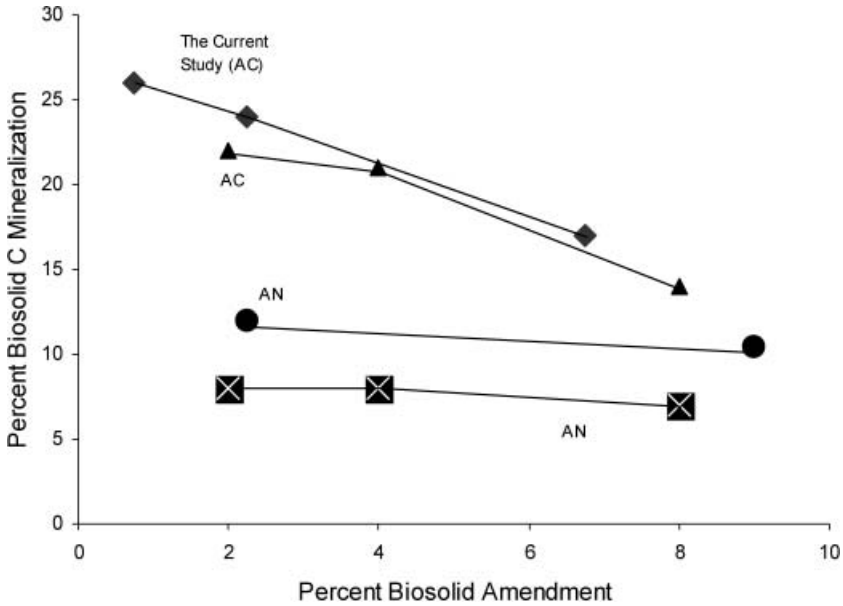


Figure 5. Comparison of normalized biosolid C-mineralization data from the current and comparable studies. Data from the current study; Boyle and Paul (1989); Hsieh, Douglas, and Motto (1981). AC, aerobically digested biosolids; AN, anaerobically digested biosolids.

The decrease in the percentage of C mineralization at greater biosolid application rates could be related to elevated salinity in these treatments. Although we did not measure EC, others did during similar incubations. Fine, Mingelgrin, and Feigin (1989) determined that soil EC rose from 0.6 to 0.7 dS m⁻¹ in a 20 Mg ha⁻¹ biosolid-amended soil following a 30-day incubation. The EC increased from 2.0 to 8.0 dS m⁻¹ when the biosolids rate was 200 Mg ha⁻¹.

CONCLUSIONS

We determined that weathered fly ash-LSR with a neutral pH and low salinity did not inhibit C mineralization in either soil or co-applied biosolids. Weathering during long-term storage probably neutralized alkalinity and lowered salinity in the fly ash-LSR. It is most likely safe to conclude that weathered fly ashes could be applied to soil at rates up to 100 Mg ha⁻¹ (5%) without affecting C mineralization in soil amended with or without biosolids. Application of fly ash/biosolid mixtures is more suitable for highly buffered soils than for poorly buffered soils.

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