Management of Lignite Fly Ash for Improving Soil Fertility and Crop Productivity

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Received: 12 April 2006 / Accepted: 19 March 2007 Springer Science+Business Media, LLC 2007

Abstract Lignite fly ash (LFA), being alkaline and endowed with excellent pozzolanic properties, a silt loam texture, and plant nutrients, has the potential to improve soil quality and productivity. Long-term field trials with groundnut, maize, and sun hemp were carried out to study the effect of LFA on growth and yield. Before crop I was sown, LFA was applied at various doses with and without press mud (an organic waste from the sugar industry, used as an amendment and source of nutrients). LFA with and without press mud was also applied before crops III and V were cultivated. Chemical fertilizer, along with gypsum, humic acid, and biofertilizer, was applied in all treatments, including the control. With one-time and repeat applications of LFA (with and without press mud), yield increased significantly (7.0–89.0%) in relation to the control crop. The press mud enhanced the yield (3.0–15.0%) with different LFA applications. The highest yield LFA dose was 200 t/ha for one-time and repeat applications, the maximum yield being with crop III (combination treatment). One-time and repeat application of LFA (alone and in combination with press mud) improved soil quality and the nutrient content of the produce. The highest dose of LFA (200 t/ha) with and without press mud showed the best residual effects (eco-friendly increases in the yield of succeeding crops). Some increase in trace- and heavymetal contents and in the level of γ -emitters in soil and crop produce, but well within permissible limits, was observed. Thus, LFA can be used on a large scale to boost soil fertility and productivity with no adverse effects on the soil or crops, which may solve the problem of bulk disposal of fly ash in an eco-friendly manner.

Keywords Lignite fly ash \cdot Press mud \cdot Groundnut \cdot Maize \cdot Sun hemp \cdot Productivity \cdot Heavy metals \cdot Radioactivity

Introduction

At present, about 70% of the total energy requirement of India is met from coal. This situation will persist for several decades, until alternative sources of energy are developed and exploited on a commercial scale. The 85 thermal power plants (TPPs) in India produce about $110 \times$ $10⁶$ t of fly ash per annum. This huge generation of fly ash poses handling, storage, and disposal problems, apart from the possible contamination of soil, crops, and surface and ground water with toxic trace and heavy metals (Carlson and Adriano 1993, Rubenstein and Segal 1993) and radionuclides (Ramachandran and others 1990). In India, fly ash is utilized as (i) a raw material in cement, cellular concrete, lime bricks, lime gypsum blocks, and building tiles; (ii) admixtures in cement concrete and in products made of timber substitutes; (iii) aggregate in concrete, roads, and building blocks; (iv) pozzolana in lime mortars and plasters and Portland cement; (v) a stabilizer for soil and for road construction; (vi) a filler in consolidation of ground, land, and mine-filling; and (vii) a soil conditioner and source of plant nutrients in agriculture and forestry. The other applications of fly ash include metals extraction, creation of cenospheres, and wastewater treatment (Asokan and others 2005). All these uses account for only 38.4% of the fly ash that is produced. This is insufficient in view of the ever-increasing generation of fly ash. In particular,

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barely 15.0% of the $\sim 1.0 \times 10^6$ t per annum of lignite fly ash (LFA) currently generated by two thermal power stations (TPS I and II, capacity 2070 MW) of the Neyveli Lignite Corporation (NLC) is utilized—a distressing situation. Apart from this, there is an urgent need to cope with the growing demand for food in India. A suitable ecofriendly technology, capable of sustainably using waste fly ash on a bulk scale, is needed to help meet this evergrowing demand for food and solve the environmental problem of disposing of huge volumes of fly ash in a single stride.

Fly ash has been examined as a resource material and has been found to comprise most of the elements, except nitrogen and humus, required for growth of agricultural crops. It can be used to correct nutrient deficiencies and to prevent metal toxicity by neutralizing soil acidity (Adriano and others 1980). Much research has been carried out on the utilization of fly ash in agriculture (Bhumbla and others 1991). The uptake of various nutrients and some toxic trace elements by crops after fly ash amendment has been studied, and the produce has been found safe for consumption (Sen and others 1997). The Central Fuel Research Institute (CFRI), in Dhanbad, has investigated the application of fly ash and pond ash in agriculture under a variety of soil types and agro-climatic conditions. These studies have shown that fly ash has positive effects as a liming agent, a soil conditioner, and a source of essential plant nutrients and is also effective in the reclamation of waste, degraded land, and mine spoil (Singh and others 1997, 1998; Ram and others 1999, 2006). Although fly ash contains several essential plant nutrients, it is devoid of humus and nitrogen (Menon and others 1990), but these can be supplemented through organic amendments (Adriano and others 1978), and the leaching of metals can be effectively reduced by chelation (Logan and Traina 1993). Fulvic and humic acid also play a major role in the migration of metals (Banuelos and Ajwa 1999).

The LFA of NLC has been reported to be alkaline (pH, 10–12; Ca, 8–12%), and its cementitious property is useful in improving the water storage capacity of sandy soils while providing a source of plant nutrients (CARD 1997). LFA, being dominated by silt-sized particles, can improve the texture, bulk density (BD), water-holding capacity (WHC), and fertility status of soil. The improvement in fertility status can be ascribed to the amelioration of the texture and other physical properties of the soil, leading to enhanced biological activity with a corresponding impact on nutrient cycling.

Few long-term, detailed studies on the bulk use of LFA in the agricultural sector, particularly on cultivable land, are available. Hence, we carried out a long-term (1997– 2000) study in which the soil at the Centre for Applied Research and Development (CARD)–NLC was amended with different doses of LFA with and without press mud (an organic waste from the sugar industry) for cultivation of groundnut and maize crops. The objective of the study was to determine the effects of LFA on soil fertility, crop yield and nutrient status, trace- and heavy-metal contents, and level of radioactivity in the soil and produce.

Materials and Methods

Field Experiment

The selected CARD–NLC agricultural land (6000 m^2) was leveled and ploughed repeatedly, and the required quantity of fly ash, collected from the NLC ash pond, was uniformly broadcast on the surface of the respective plots and mixed thoroughly by ploughing. The experimental setup was a randomized block design having 18 treatments (T1–T18), each in quadruplicate; plots were 15 m \times 4 m and were treated with various dosages (0, 5, 10, 20, 50, 100, and 200 t/ha) of LFA, with and without press mud (see Table 1), where treatment T1 was the control (with neither LFA nor press mud); and T2 was the application of press mud only. In treatments T3, T5, T7, T9, T11, and T13, LFA alone (5, 10, 20, 50, 100, and 200 t/ha, respectively) was applied. In treatments T4, T6, T8, T10, T12, and T14, LFA (5, 10, 20, 50, 100, and 200 t/ha, respectively) was applied in combination with press mud; and in T15–T18, LFA (5, 10, 20, and 50 t/ha, respectively) was also applied in combination with press mud. For all applications involving press mud, the dose was 10 t/ha. A one-time application of LFA was carried out in T11–T18; repeat applications of LFA were carried out before the cultivation of crops III and V in T3–T10, at the same rate as before the cultivation of crop I.

The cultivation sequence was as follows: crop I, groundnut (August–November 1997); crop II, sun hemp (December 1997–January 1998); crop III, maize (May– August 1998); crop IV, sun hemp (September–November 1998); crop V, groundnut (December 1998–April 1999); and crop VI, maize (November 1999–March 2000). The variety of groundnut planted was VRI-2, and that of the maize was Ganga-5; for sun hemp a local cultivar was used. The recommended NPK (kg/ha) fertilizers for groundnut (17:34:54) and maize (62.5:62.5:50) were applied via urea, super phosphate $(Ca(H_2PO_4)_2)$, and potash muriate (KCl), respectively. Only 50% N (no PK) was applied to crop VI (maize) in T9 (50 t/ha of LFA). No NPK fertilizer was added to sun hemp.

Other amendments, such as gypsum (325 kg/ha), humic acid (20 kg/ha), and biofertilizer ($Rhizobium + a$ phosphobacterium–Bacillus megatherium) (8 kg/ha each), were applied along with a basal dosage of chemical fertilizers in all treatments, including the control. These amendments

Table 1 Treatment details

Treatment	Dose (t/ha)
T1	Control
T ₂	Press mud (10)
T3	LFA(5)
T ₄	LFA (5) + PM (10)
T ₅	LFA(10)
T ₆	LFA $(10) + PM (10)$
T7	LFA(20)
T8	LFA $(20) + PM (10)$
T9 ^a	LFA(50)
T ₁₀	LFA $(50) + PM (10)$
T11	LFA (100)
T ₁₂	LFA $(100) + PM (10)$
T ₁₃	LFA (200)
T ₁₄	LFA $(200) + PM (10)$
T ₁₅	LFA (5) + PM (10)
T ₁₆	LFA $(10) + PM (10)$
T ₁₇	LFA $(20) + PM (10)$
T ₁₈	LFA $(50) + PM (10)$

LFA lignite fly ash, PM press mud. One-time application of LFA in T11–T18; repeat applications in T3–T10 (before cultivation of crops III and V and at the same rate as before the cultivation of crop I) ^a Only 50% of N (no PK) was applied to crop VI (maize)

were made as supplementing agents in view of the poor soil fertility and the normal practice of local farmers. No other amendments were made for growing sun hemp. Press mud, a lightweight waste product from the sugar industry that is abundantly available in the vicinity of the NLC, was used as a source of organic matter (OM) and plant nutrients (Table 2) to improve the texture and fertility of the soil. Gypsum, locally available and of a commercial-grade composition, including 14% sulphur, was applied to reduce crust formation in the soil. Potassium salt of humic acid (20% humic acid content), chemically prepared from lignite at CARD–NLC, was applied to the soil to stimulate microbial growth and to provide some essential elements, such as N and K, as well as to enhance the availability of P and micronutrients, such as Zn, Fe, Cu, and Mn. Biofertilizer, a composition of Rhizobium and a phosphobacterium that was developed in the CARD microbiology laboratory, was mixed with the soil to act as an inoculum for N-fixing and P-solubilizing microbes.

Soil, Fly Ash, and Crop Produce Analyses

Various samples of (i) the dry LFA used for field trials, (ii) the composite soil and press mud before the start of the field experiments (cultivation of the first groundnut crop), (iii) the crop produce (grain or kernel and straw), and Table 2 Physicochemical characteristics of press mud

EC electrical conductivity, WHC water-holding capacity

(iv) the soil from plots after each harvest were collected, processed, and analyzed as discussed below.

For the LFA and the soil, physical properties, such as the mechanical composition, BD, porosity, and WHC, were determined by standard smethods (Piper 1950). Chemical properties, namely, pH, electrical conductivity (EC), organic carbon (OC), and total major secondary nutrients, were also determined by standard methods (Jackson 1967). The available major secondary nutrient (N, P, K, S, Ca, and Mg) contents were measured by following the procedures given in a standard textbook (Tandon 1995).

The samples of soil, LFA, and crop produce were digested in an analytical microwave system (Prolabo microwave oven), and various Suprapur® acids (Merck Germany) were used as reagents for analyzing the total contents of micronutrients (Cu, Zn, Mn, and Fe) and trace and heavy metals (Pb, Ni, Co, Cd, Cr, As, and Hg). Diethylenetriaminepentaacetic acid (DTPA) was used to extract samples for determination of available trace- and heavy-metal contents in the LFA and soil, following the prescribed method (Tandon 1995). The DTPA-extractable fraction of the total content has been found to be relatively bioavailable to roots (Adriano and others 2002). Total trace- and heavy-metal contents in various digested soil, LFA, grain, and straw samples and the available content in DTPA extracts of soils and LFA were estimated by liquid ion chromatography (Waters®). The determination of Cu, Zn, Mn, Fe, Pb, Ni, Cd, and Co contents was made with a C18 column, with sodium octane sulphonate, tartaric acid, and acetonitrile as eluents; a postcolumn reagent; and an ultraviolet light (UV) detector (520 nm). The IC Pak-A HR column, borate–gluconate eluent, and another UV detector (365 nm) were used for determination of Cr (as $CrO₄²$). The calibration standard sample for various trace and heavy metals was ICP multielement standard solution IV (Cat. No. 1.11355.0100), procured from Merck. The detection limit for Cu, Co, Mn, and Zn was 0.005 mg/L; and for Cd, Cr, Fe, Ni, and Pb, 0.015 mg/L. The elements As and Hg were analyzed with a Unicom SP-2900 atomic absorption spectrometer, with the hydride cold vapor generation method. The detection limit for As and Hg was 0.005 mg/L.

The dehydrogenase activity in soil was estimated by the triphenyltetrazolium chloride method (Klein and others 1971). Total bacteria, ectomycorrhiza, and P-solubilizing bacteria counts were estimated by following standard procedures (Linderman 1992, Chhonkar and others 2002). The levels of γ -emitting radionuclides (²²⁶Ra, ²²⁸Ac, ⁴⁰K) in the soil, LFA, and crop produce were measured using a γ -ray spectrometer with a high-purity germanium (HPGe) detector with a resolution of 1.95 kV at 1.33 MeV, a volume of 77 cm^3 , and lead shielding lined with a 1-mm-thick layer of aluminum. The detector was kept at 2π geometry for counting. The γ -rays emitted from the sample were detected by the HPGe detector coupled with a PC-based multichannel analyzer. The detection limits of the radionuclides in the soil and LFA samples was 5 Bq/kg; and for grain and straw samples, 0.05 Bq/kg.

Statistical Analysis

To assess the significance ($p < 0.05$) of the yield data, sample analyses, and various biological parameters, we determined the critical difference value by one-way analysis of variance, using an MSTAT package (Freed and Eisensmith 1991). For other cases, wherever applicable, the standard error is shown in tables and figures.

Results and Discussion

Physicochemical Properties of Experimental Soil and LFA

The field soil was classified as an Alfisol (USDA Soil Taxonomy). From Table 3, it can be seen that this was texturally sandy loam (sand, 65.7%; silt, 19.3%, clay 15.0%). The WHC (26.7%) and porosity (35.6%) values were lower than for fertile soil (ICAR 1996). Conversely, LFA had silt as the predominant fraction (56.0%), and it had lesser contents of sand (34.8%) and clay (9.2%). The WHC and porosity values were higher and the BD lower for LFA than for the field soil.

The field soil (Table 3) was acidic (pH, 5.49). Its EC (0.612 dS/m) was in the normal range, but its OC content (1.32%) was higher than is normal in Indian soil $(0.4-0.6\%)$ (ICAR 1996). This was probably due to the deposition of fine, unburned coal particles on the surface of the field in the vicinity of a TPP. Similarly, the suspended coal particles near the lignite mine might also have enhanced the OC content of field soil. The fertility of the soil was poor, as indicated by the low concentrations of total and available major and secondary nutrients (mg/kg): for N, 660 and 100; P, 82 and 4.2; K, 8900 and 140; S, 5700 and 39.6; Ca, 10,500 and 62.7; and Mg, 5600 and 48.4. Similarly, low concentrations of total and available micronutrients were observed (mg/kg): for Cu, 82.59 and 2.44; Zn, 92.47 and 1.23; Mn, 145.65 and 15.89; and Fe, 20,600 and 31.95. The total contents of the trace and heavy metals were as follows: for Pb, Ni, Co, and Cr, within the range 12.68–41.77 mg/kg; for Cd, 2.04 mg/kg; and for As and Hg, below detection limit (BDL). The available Ni and Cd contents were 1.96 and 0.04 mg/kg, respectively, and those of Pb, Co, Cr, As, and Hg were BDL. The radioactivity of the y-emitters ²²⁰Ra, ²²⁸Ac, and ⁴⁰K was 19.6, 37.4, and 212.2 Bq/kg, respectively.

The LFA was alkaline (pH, 10.23–10.54), with higher EC (3.98–4.29 dS/m) and lower OC contents (0.13–0.17%) than the experimental soil. The contents of the secondary nutrients, S, Ca, and Mg, were also higher, but the values for P and K were less than the corresponding values in the soil. Among the micronutrients, the concentrations of total Zn, Mn, and Fe were appreciably higher and that of Cu was lower. LFA also had higher concentrations of total and available Pb, Ni, Cd, Co, Cr, and As than the soil. The concentration of Hg in both LFA and soil was BDL. The radioactivities of the y-emitters ²²⁰Ra (79.6 Bq/kg), ²²⁸Ac (80.4 Bq/kg), and 40 K (391.8 Bq/kg) were higher than were found in the soil.

Biometric Observations

During the cultivation of the groundnut and maize, regular biometric observations of growth and yield of the standing crops were made at different growth stages. One-time application (T11–T18) and repeat applications (T3–T10) of LFA ranging from 5 to 200 t/ha with and without press mud (10 t/ha) and other amendments significantly ($p < 0.05$) increased plant height, the number of root nodules, and the number of cobs, the maximum being with treatment T14 (200 t LFA/ha with press mud) (data not included). The overall growth of both crops was luxuriant, with uniform and early maturity and intensely green leaves and with bigger cobs and fewer incidences of pests, disease, and weeds in the LFA-treated plots. Such improvements were somewhat better in combination treatments than in treatments with LFA alone. A significant residual effect on plant growth parameters from a one-time application of LFA in treatments T11 (100 t/ha), T12 (100 t/ha + press mud), T13 $(200 \t{t/ha})$, and T14 $(200 \t{t/ha} + \text{press mud})$ was observed

Parameter	Original soil (CARD)	Lignite fly ash		
Sand $(\%)$	65.7 ± 0.58	$32.4 - 35.3$		
Silt $(\%)$	19.3 ± 0.16	$54.7 - 57.8$		
Clay $(\%)$	15.0 ± 0.13	$9.2 - 10.0$		
BD (g/cm^3)	1.60 ± 0.01	$0.893 - 0.971$		
WHC $(\%)$	26.7 ± 0.23	44.5–47.2		
Porosity $(\%)$	35.6 ± 0.31	$52.3 - 54.6$		
EC (dS/m)	0.612 ± 0.005	3.98-4.29		
Dehydrogenase activity (mg $kg^{-1} h^{-1}$)	0.29 ± 0.002			
pH	5.49 ± 0.048	$10.23 - 10.54$		

Table 3 Physicochemical characteristics of experimental soil before the experiment and of LFA applied at different stages of the field trials $(mean \pm SE)$

Major secondary nutrients, trace elements, and heavy metals (mg/kg)

BD bulk density, BDL below detection limit, CARD Centre for Applied Research and Development, EC electrical conductivity, LFA lignite fly ash, WHC water-holding capacity

^a Available nutrient is the portion of the total nutrient content in the soil that can be absorbed most readily and easily assimilated by the growing plants

during cultivation of successive crops, including those treated with repeat application of LFA at 20 and 50 t/ha. The residual effect was not significant at lower doses (5–50 t/ha) in one-time applications with crop V (groundnut), but significant effects were observed in the case of crops III and VI (maize). The growth performance and root nodulation of crops II and IV (sun hemp), grown as green manure, were luxuriant, even up to 200 t/ha of LFA with and without press mud (T13 and T14). Thus, the overall growth of crops was greatly influenced by one-time application of 100 and 200 t

Fig. 1 Effect of one-time and repeat application of different doses of lignite fly ash (LFA) with and without press mud (PM) on the pod or grain yield of groundnut and maize crops grown at the Centre for Applied Research and Development, Neyveli Lignite Corporation. Note: Treatments followed by the same letter are not significantly different at LSD $(p < 0.05)$

LFA/ha alone, in comparison to the control; however, such effects were slightly more pronounced in amendments with press mud.

Crop Yield

The kernel yield (Figure 1) of crop I (groundnut) significantly ($p < 0.05$) increased by 26.0–64.0% in relation to the control (T1) after application of LFA alone at 5–200 t/ha (T3, T5, T7, T9, T11, T13). In combination treatments (LFA with press mud; i.e., T4, T6, T8, T10, T12, T14), the increase was 28.3–66.7%, with the maximum increase being with treatment T14 (200 t/ha LFA + 10 t/ha press mud). An additional, significant ($p < 0.05$) increase of 3.0– 5.0% was observed as a result of the addition of press mud at 10 t/ha. Similarly, in crops III (maize), V (groundnut), and VI (maize), a significant ($p < 0.05$) increase (13.0– 88.5%) in yield over that of the corresponding control was noticed for treatments with LFA alone and for combination treatments. It is pertinent to mention that the ranges of difference for crop V (groundnut) were smaller than those for crops III and VI (maize). To illustrate the point, for crop V the range of increase was 13.0–77.4%, whereas for crops III and VI it was 23.6–88.4% and 26.6–79.7%, respectively. It seems that the application of fly ash alone and in combination with press mud is more effective for maize than for groundnut. Furthermore, one-time application of LFA alone at 100 t/ha (T11) and 200 t/ha (T13) and repeat applications at 50 t/ha LFA alone (T9) before the sowing of crops III and V showed a highly significant $(p < 0.01)$ increase (55.0–85.0%) in yield over that of the corresponding control. Thus, the application of LFA alone and in combination with press mud, particularly at higher doses of 50–200 t/ha, was more effective in increasing the yield of groundnut and maize crops; the highest yield dose was 200 t/ha. In general, crop yields significantly increased with applications of LFA up to 200 t/ha with and without press mud; however, for crop I (groundnut) the difference between the kernel yield at 200 t/ha LFA and that at 100 t/ha LFA was insignificant (Figure 1). However, in subsequent crops the yield differences between 100 and 200 t/ha were significant, with appreciably higher values at 200 t/ha, thereby evincing a better residual effect at the higher dose.

The scatter plot of the yield data (Figure 2) revealed that for both groundnut and maize, the increase in yield was linear with LFA doses up to 50 t/ha, after which the yield increased at a slower rate up to the highest dose, 200 t/ha. However, the response to LFA was higher in groundnut than in maize. The yield response curve (Figure 2) fit better with a logarithmic model ($R^2 = 0.941 - 0.99$) than with a linear model ($R^2 = 0.726 - 0.772$). An important finding was that the grain yield of crop VI (maize) grown with 50% of the recommended dose of N-only fertilizer (without P or K) was almost the same as that of the crop grown with

Fig. 2 Scatter plot of crop yield response to dose of lignite fly ash (LFA)

100% of the recommended fertilizer dose, thereby suggesting a substantial saving. A similar result was recently observed by Mittra and others (2005).

Like the yield of edible produce from various crops, the yield of straw showed significant ($p < 0.05$) increases (7.1–88.7%) over the corresponding control when LFA doses from 5 to 200 t LFA/ha were applied alone (T3–T13) and in combination with press mud (T4–T14); 5–15% of the increase was contributed by press mud (data not included). Also, there was less of an increase in the yield of a crop grown after sun hemp (which was used as green manure) than in the preceding similar crop, showing that the combination treatments were more effective than green manuring alone.

The significantly increased yield for groundnut and maize, even up to the final crop—that is, the overall beneficial effect of LFA application with other amendments was an indirect indicator of improvement in soil fertility. As was found in another investigation (Narayanaswamy and Nambirajan 2000), there were fewer pests in LFA-amended plots, particularly polyphagus pests such as Helicoverpa armigera and Spodoptera litura, a circumstance that also enhanced crop yield.

Effect of LFA Application on Soil Characteristics After Harvest of Crop VI

Physicochemical Characteristics

Figure 3 shows that in the physical properties of the soil (sand, silt, and clay contents, BD, available water, WHC, porosity, pH, and EC) after the harvesting of crop VI (final crop), significant improvements were observed up to a dose of 200 t/ha of LFA alone (T3–T13) and with press mud (T4–T14).

The application of LFA modified the texture of soil from sandy loam to silt loam by reducing the sand content (to 60.2% from 64.2%), clay content (to 15.3% from 15.7%), and BD (to 1.50 Mg/m^3 from 1.58) and increasing the silt content (to 24.5% from 20.1%), WHC (to 30.3% from 27.4%), porosity (to 40.5% from 37.1%), and available water (to 4.85% from 2.86%). In fact, the moisture retention capacity of the soil after these amendments increased to a great extent, and moisture was clearly visible for a longer time than in control plots. After irrigation, the water completely infiltrated the soil profile of the control plot within 3–4 h of irrigation because of the sandy loam texture of the soil; in contrast, the duration of moisture retention was observed to have been enhanced in LFA-amended plots. In addition, cultivation between the rows was easier in LFA-amended plots than in control plots because there was less crust formation and the soil was properly conditioned. This is important for agricultural purposes, because it allows for easy penetration and better growth of plant roots. Fly ash consists of hollow spheres, and it is possible that the small LFA particles, containing a larger proportion of silt $(54.7–57.8%)$ than the soil $(19.3%)$, accumulate in voids by releasing bigger particles of soil. This helps to modify the texture of soil (Khan and others 1996, Ram and others 2006). The increase in WHC is attributable to the higher silt content in LFA than in soil, which is in agreement with the observation of Furr and others (1977). The decrease in BD was obviously due to the change in total porosity as well as to modification of the macro- and micropore size distribution (Chang and others 1977,

Fig. 3 Effect of application of lignite fly ash on final status of different physicochemical properties of soil after harvest of crop VI (final) maize. AW, available water; BD, bulk density; EC, electrical conductivity; WHC, water-holding capacity; LFA, lignite fly ash; PM,

Adriano and others 1980). Furthermore, the presence of Ca–Si minerals, which became pozzolanic (zeolite-forming) upon being added in the form of fly ash to the moist soil (Fulekar and Dave 1986), improved various physicochemical properties of the soil, such as BD, porosity, WHC, and available water (Elseewi and others 1980). The pH and EC values during the complete span of amendment with LFA alone and with press mud and other amendments increased to 6.62 from 5.51 and to 0.870 dS/m from 0.608, respectively.

The OC content of soil with LFA alone and with LFA in combination with press mud treatments increased from

press mud. T1–Control, T2–Press mud (10 t/ha), T9–LFA (50 t/ha), T10–LFA (50 t/ha) + PM (10 t/ha), T11–LFA (100 t/ha), T12–LFA (100 t/ha) + PM (10 t/ha), T13–LFA (200 t/ha), T14–LFA (200 t/ha) + PM (10 t/ha)

1.32% (control) to 1.52% (200 t/ha LFA + press mud). This is probably ascribable to the application of OM, such as press mud, biofertilizer, and humic acid, apart from the contribution of root biomass and exudates. There was significant ($p < 0.05$) improvement in the content of available major and secondary nutrients (Figure 3): N increased to 125 mg/kg from 110; P, to 5.6 mg/kg from 4.5; K, to 147.6 mg/kg from 145.8; S, to 50.3 mg/kg from 42.9; Ca, to 74.5 mg/kg from 64.1; and Mg, to 50.6 mg/kg from 49.2. This increase in Ca and Mg content reflected the gradual increase in pH of the soil with increasing dose of LFA.

	μ auntin (t LPA/na)	KauluaCitvity (Dy/Kg)							
		Soil	Groundnut (crop I)	Maize (crop III)	Groundnut (crop V)	Maize (crop VI)			
226 Ra	Control	18.1 ± 0.47	0.65 ± 0.02	0.57 ± 0.012	0.67 ± 0.02	0.61 ± 0.014			
	100	27.3 ± 0.72	0.71 ± 0.05	0.63 ± 0.03	0.80 ± 0.05	0.66 ± 0.04			
	200	32.5 ± 0.93	0.82 ± 0.07	0.69 ± 0.04	0.86 ± 0.05	0.72 ± 0.02			
228 Ac	Control	35.8 ± 0.98	0.72 ± 0.04	0.76 ± 0.03	0.77 ± 0.03	0.79 ± 0.02			
	100	41.9 ± 1.62	0.81 ± 0.07	0.81 ± 0.06	0.84 ± 0.05	0.84 ± 0.05			
	200	47.2 ± 1.69	0.99 ± 0.08	0.84 ± 0.05	0.97 ± 0.07	0.87 ± 0.05			
$^{40}{\rm K}$	Control	208.7 ± 9.2	97.8 ± 3.2	57.5 ± 2.9	98.2 ± 3.8	58.2 ± 3.1			
	100	219.4 ± 11.7	106.9 ± 4.3	61.7 ± 3.6	107.6 ± 4.7	62.2 ± 4.1			
	200	224.1 ± 13.6	120.3 ± 4.8	62.1 ± 3.4	122.5 ± 6.1	63.5 ± 4.7			
	Permissible limits								
	Soil ^a (Bq/kg)		Edible crop produce ^b						
			Permissible daily intake limit (Bq)		Calculated maximum daily intake (at 600 g/day) (Bq)				
226 Ra	370	0.61		0.516					
228 Ac	259	2.2		0.594					

Table 4 Effect of application of LFA on γ -radioactivity of soil after the harvest of crop VI (final) maize crop and on produce (mean \pm SE)

LFA lignite fly ash

 a UNSCEAR (1982)

 b Bowen (1966) and Eisenbud and Thomas (1977)

The available micronutrients and trace and heavy metals also increased considerably. In particular, Cu increased to 2.48 mg/kg from 2.31; Zn, to 1.42 mg/kg from 1.26; Mn, to 16.35 mg/kg from 16.05; Fe, to 35.98 mg/kg from 32.85; and Ni, to 2.30 mg/kg from 1.99. For the other trace and heavy metals, such as Cr, Pb, Co, Cd, and As, the content was BDL.

 40 K 925 104 73.5

 $T_{\text{recommant}}$ (t LR/h_0) Radioactivity (B_0/h_0)

It should be noted that there was a greater increase in the content of major and secondary nutrients and micronutrients from combination treatments than from LFA treatments alone. The improvement in the physicochemical properties of amended soil indicates the possibly beneficial impact on crop yield from the decomposition and N-demineralization of press mud and other OM (Adriano and others 1982). The acidifying tendency of protons from decomposing OM from press mud, biofertilizer, humic acid, and even gypsum would offset the alkalizing tendency of the CaO and MgO present in LFA, keeping the pH of the soil near neutral (6.62) after the harvest of the final crop (crop VI). As a result, the wide gap between the pH values of soil (5.47) and those of LFA (10.2–10.5) and the increase in buffering capacity do not matter much and result in no toxicity due to controlled carryover of toxic trace and heavy metals by crop produce. Here, the hypothesis can also be considered that fly ash mixed with biosolids produces a material with more balanced properties (e.g., neutral pH and the presence of essential elements) and less potential for environmental contamination than either material applied alone (Stevenson 1982, Sims and others 1993, Wong 1995). The addition of sewage sludge and other amendments, such as lime and gypsum, to coal ash has been effective in maintaining pH of the soil, changing the water-soluble OC and EC, and releasing N, P, and K for assimilation by plants during revegetation of soil (Martens and Beahm 1976, Pietz and others 1989). Thus, after application of various dosages of LFA up to 200 t/ha, alone and in combination with press mud and other amendments as supplementing agents, the physicochemical properties and fertility of the soil improved. The probable mechanism for improving soil health by LFA treatment is discussed in detail elsewhere (Ram and others 2006).

Radioactivity Level

After application of the maximum dose of LFA (200 t/ha), the radioactivity of γ -emitters increased above the control values (Table 4): that of 226 Ra rose to 32.5 Bq/kg from 18.1; ²²⁸Ac, to 47.2 Bq/kg from 35.8; and ⁴⁰K, to 224.1 Bq/kg from 208.7. However, the radionuclide contents were well within the permissible limits (UNSCEAR 1982) and were in good agreement with other findings (Ramachandran and others 1990; McMurphy and Rayburn 1993, Ram and others 2006).

Table 5 Effect of application of LFA on the biological activities in soil after the harvest of each crop

	Crop	T1	T ₂	T ₉	T ₁₀	T ₁₁	T ₁₂	T ₁₃	T ₁₄	LSD $(p = 0.05)$
Ectomycorrhiza (spores/g)	Groundnut (crop I)	34	46	65	70	86	90	95	98	3.26
	Maize (crop III)	65	70	90	98	97	103	106	117	3.47
	Groundnut (crop V)	70	78	110	120	126	130	136	140	4.04
	Maize (crop VI)	59	67	85	93	95	101	105	112	3.47
P-solubilizing bacteria (\times 10 ⁴ CFU/g)	Groundnut (crop I)	1.2	2.4	3.2	3.8	4.2	4.7	5.2	5.8	0.28
	Maize (crop III)	1.8	3.4	6.9	7.0	7.9	8.5	9.2	9.4	0.44
	Groundnut (crop V)	2.3	4.4	6.8	7.5	8.2	9.1	10.5	12.0	0.40
	Maize (crop VI)	2.2	2.4	2.9	3.6	4.6	6.2	7.0	8.0	0.45
Total bacterial count (\times 10 ⁴ CFU/g)	Groundnut (crop I)	2.0	4.0	5.2	5.7	7.1	7.9	8.5	9.2	0.41
	Maize (crop III)	2.5	4.1	8.0	9.2	9.7	10.5	12.0	13.4	0.54
	Groundnut (crop V)	3.0	4.6	8.6	9.5	16.0	19.0	22.0	26.0	0.53
	Maize (crop VI)	2.6	4.0	5.9	6.2	7.5	8.2	9.2	9.7	0.46
Dehydrogenase activity (mg $kg^{-1}h^{-1}$)	Groundnut (crop I)	0.30	0.34	0.34	0.36	0.35	0.38	0.37	0.39	0.03
	Maize (crop III)	0.32	0.36	0.33	0.38	0.35	0.40	0.38	0.41	0.03
	Groundnut (crop V)	0.34	0.41	0.38	0.42	0.39	0.41	0.38	0.44	0.04
	Maize (crop VI)	0.31	0.43	0.36	0.44	0.38	0.42	0.39	0.46	0.03

Initial values before the start of experiment: Ectomycorrhiza 32 spores/g, P-solubilizing bacteria 1.2×10^4 CFU/g, total bacterial count 2.0×10^4 CFU/g, dehydrogenase activity 0.30 mg kg⁻¹h⁻¹

CFU colony-forming unit, LFA lignite fly ash, LSD least significant difference

Biological Activity

The biological activity of the soil, which was low initially, improved significantly ($p < 0.05$) as a result of the addition of different doses of LFA alone (T3–T13) and in combination treatments (T4–T14), including green manuring (Table 5). After the harvest of crop I (groundnut), soil that had been amended with LFA at 5–200 t/ha alone and in combination with mud (T3–T14) showed a significant $(p < 0.05)$ enhancement in biological activity in comparison with the control. Up to the dose of 200 t/ha LFA alone and in combination with press mud, ectomycorrhiza increased to 98 spores/g (initial value, 32 spores/g); P-solubilizing bacteria increased to 5.8×10^4 colony-forming units (CFU)/ g (initial value, 0.9×10^4 CFU/g); total bacterial count rose to 9.2×10^4 CFU/g (initial value, 1.8×10^4 CFU/g); and dehydrogenase activity increased to 0.30–0.39 mg $kg^{-1} h^{-1}$ (initial value, $0.27 \text{ mg kg}^{-1} \text{ h}^{-1}$). The same trend also held and was rather proportionately higher for succeeding crops. These enhanced microbial activities in the soil might have played a crucial role in appreciably improving the soil fertility and the yield of different crops. The organic amendment as a supplementing agent might have contributed to improving the soil by increasing the cation exchange capacity (CEC) and the OM content. The increase in soil OM was also attributable to a greater input from root biomass as a result of better crop productivity. The readily metabolizable C and N in press mud and other organic amendments not only led to increased root biomass and root exudates as a result of greater crop growth but also contributed to the increase in soil biological activity (Masto and others 2006). This resulted in better fertility status and enhanced microbial activity, in addition to enhancing the immobilization of toxic elements and inhibiting their effects on the microbes (Chaney and Giardono 1977). Additionally, the role of amended soil texture in enhancing biological activity could not be ruled out (Lyon and others 1952). These observations are in agreement with findings by other workers (Pichtel 1990, Lai and others 1999).

Effect of LFA Application on Characteristics of Groundnut Kernels and Maize Grain

Nutrient Status

From Figure 4 it can be seen that the total content of major and secondary nutrients in groundnut kernels from crop I significantly ($p < 0.05$) increased, except for N. After application of LFA alone at 5–200 t/ha (T3–T13) and in combination with press mud (T4–T14), the values for N increased from 4.61% (control, T1) to 4.72%; for P, from 0.50% to 0.58%; for K, from 1.39% to 1.53%; for S, from 0.13% to 0.19%; for Ca, from 0.57% to 0.70%; and for Mg, from 0.19% to 0.29%. These increases in the concentration of major and secondary nutrients were in accord with the yield results.

After application of LFA alone (T3–T13) and in combination with press mud (T4–T14), the total content of

Fig. 4 Effect of application of lignite fly ash on major secondary nutrient content in kernel samples from crop I groundnut. Note: Treatments followed by the same letter are not significantly different $(p < 0.05)$. T1–Control, T2– Press mud (10 t/ha), T9–LFA (50 t/ha), T10–LFA (50 t/ha) + PM (10 t/ha), T11–LFA (100 t/ha), T12–LFA (100 t/ha) + PM (10 t/ha), T13–LFA (200 t/ha), T14–LFA (200 t/ha) + PM (10 t/ha). Treatments followed by the same letter are not significantly different at LSD $(p < 0.05)$

micronutrients in the kernels and grain showed a significant $(p < 0.05)$ increasing trend over that of the corresponding control: for Cu, the increase was from 7.81 to 7.95 mg/kg; Zn, from 47.5 to 49.19 mg/kg; Mn, from 37.42 to 38.08 mg/kg; and Fe, from 66.7 to 68.3 mg/kg (Figure 5). The total content of some trace and heavy metals also increased significantly ($p < 0.05$) over that of the corresponding control (Figure 5): Cr increased from 0.69 to 0.88 mg/kg; Pb, from 0.13 to 0.20 mg/kg; and Ni, from 11.75 to 12.18 mg/kg. The total contents of Co, Cd, As, and Hg were BDL in all treatments with LFA alone, in combination treatments, and in the corresponding control.

The increased concentration of major and secondary nutrients, micronutrients, and trace and heavy metals in the kernel samples of crop I also occurred in crop produce (kernel and maize) of crops III, V, and VI. However, there was little variation in the ranges (i.e., there were slightly higher ranges of difference in the case of kernels and lower ranges of difference in the case of maize grain). As a point of illustration, the trace- and heavy-metal and micronutrient contents in the produce, including the produce of subsequent crops, are given in Table 6. There should not be any apprehension about accumulation of toxic trace and heavy metals in crop produce grown with LFA.

The greater increase in major and secondary nutrients in kernels than in maize grain was probably due to the fact that LFA helped leguminous crops by stimulating activity in the rhizosphere. This caused the secretion of organic molecules that might have enhanced the availability of these nutrients in the LFA-amended field, along with favorable conditions for the growth of N-fixing and P-solubilizing microorganisms. The small increase in traceand heavy-metal contents suggests a low availability of these metals, which gradually decreased because of (1) the increase in the pH of the soil with increasing LFA dose (Fulekar 1993), (2) the presence of metals in oxide form (Page and others 1979), and (3) an increase in the adsorption of metals by soil with increasing pH and the resulting marginal uptake by crop produce (Adriano and others 1980). Besides the presence of metals in oxide form (Page and others 1979) in LFA, the possibility of iron oxyhydroxides (e.g., goethite) forming from ash–water interaction, with elements scavenged through flocculation either as a coating agent or as discrete grains on the surface

Fig. 5 Effect of application of lignite fly ash on micronutrients and trace- and heavy-metal contents in kernel samples from crop 1 groundnut. T1–Control, T2–Press mud (10 t/ha), T9–LFA (50 t/ha), T10–LFA (50 t/ha) + PM (10 t/ha), T11–LFA (100 t/ha), T12–LFA (100 t/ha) + PM (10 t/ha), T13–LFA (200 t/ha), T14–LFA (200 t/ha) + PM (10 t/ha). Treatments followed by the same letter are not significantly different at LSD ($p < 0.05$)

of fly ash particles (Leckie and others 1980), could also be of significance. However, the possibility of metals becoming more soluble after a long time cannot be entirely ruled out, because the liming effect of fly ash application is prone to diminish because of natural or anthropogenic soil acidification (Sims and others 1995). The uptake of toxic trace and heavy metals in kernels depends on several factors, such as soil pH, OC, and CEC (Chaney 1973, Yassoglou and others 1987, Xian 1989, Chambers and Sidle 1991), microorganisms around the root zone (Kisku and others 2000), concentration of the metals and their form of occurrence, and soil depth (Olaniya and others 1992). Other factors, such as mobility of the trace element toward the root, its transport from the root surface into the root, and its translocation from the root to the shoot (Chaney and Giardono 1977), also play an important role. Absorbed Pb and Cr, however, normally remain in roots and are not readily translocated to shoots (Alloway 1995). In contrast, Cd can readily be translocated from roots to shoots, but the high Zn contents and pH of LFA-amended soil reduce the uptake of Cd. Probably for these reasons, the uptake of these toxic trace and heavy metals in the kernel and maize grain is well within the critical toxicity limit (Macnicol and Beckett 1985, Alloway 1995, Srivastava and Gupta 1996). The higher uptake of Ni (from 11.75 to 12.85 mg/kg) was probably due to its mobility in plants, as was its extensive accumulation in seeds (Alloway 1995), which is also within the critical toxicity limit. The concentrations of Co, Cd, and As were BDL, and this can be simply explained in view of their limited original content in both the soil and the LFA. These observations are in agreement with those of earlier studies (Srivastava and Gupta 1996; Saxena and others 1998), and the values are within the critical toxicity concentration limits.

Radioactivity Level

Grain and straw samples of groundnut and maize from crops I, III, V, and VI grown in the control treatment as well as in soil amended with higher doses of LFA (100 and 200 t/ha alone) showed an increase in the uptake of the γ -emitters ²²⁶Ra, ²²⁸Ac, and ⁴⁰K in comparison with the produce of the corresponding preceding crops (Table 4). Also, the uptake was higher with LFA treatment than in the control, the maximum being at 200 t/ha (T13). However, this increase in radioactivity level—from 0.65 to 0.86 Bq/kg for 226 Ra, from 0.72 to 0.99 Bq/kg for 228 Ac, and from 97.8 to 122.5 Bq/kg for 40 K in kernels (and lower ranges in maize grain)—would not have any adverse effect, because these values are generally found in produce from crops grown on agricultural land without fly ash or other amendment and are well within the normal range (Bowen 1966, Eisenbud and Thomas 1977). The radioactivity level of these emitters was found to increase only marginally except in the case of 40 K. This was possibly due to the application of LFA with higher radioactivity values for 40 K (392 Bq/kg) than found in the soil (212 Bq/kg). Although the radioactivity of potash muriate was not measured, it is possible that its application as a component of the basal dose of chemical fertilizer (NPK) during cultivation raised the level of $40K$ in the produce (Srivastava and Gupta 1996). In a separate study, concern was expressed about radioactivity in produce from crops where chemical fertilizers including KCl had been applied (Maxino 2001). However, the application of phosphatic fertilizer, liming, and manures has been shown to reduce the uptake of

Crop Concentration (mg/kg) Control PM 10 t/ha LFA 50 t/ha LFA 50 t/ha + PM 10 t/ha LFA 100 t/ha LFA 100 t/ha + PM 10 t/ha LFA 200 t/ha LFA 200 t/ha + PM 10 t/ha Groundnut (crops I and V) Cu 7.81–8.14 7.85–8.19 7.90–8.20 7.93–8.23 7.86–8.25 7.88–8.27 7.92–8.26 7.95–8.33 Zn 47.52–49.72 47.57–49.77 49.01–49.84 49.15–49.92 48.02–50.06 48.09–50.11 49.11–51.15 49.19–51.24 Mn 37.42–38.65 37.55–38.79 37.90–38.83 38.05–38.89 37.72–38.95 37.79–39.04 38.00–39.11 38.08–39.18 Fe 66.72–68.83 66.87–68.98 68.19–69.22 68.20–69.45 67.29–70.06 67.36–70.19 68.21–70.42 68.30–70.61 Cr 0.47–0.69 0.49–0.71 0.67–0.84 0.71–0.85 0.65–0.82 0.69–0.84 0.72–0.85 0.83–0.88 Pb 0.13–0.17 0.11–0.14 0.17–0.16 0.19–0.15 0.14–0.20 0.12–0.17 0.19–0.22 0.20–0.20 Ni 11.75–12.14 11.79–12.19 12.15–12.59 12.08–12.52 12.03–12.78 11.97–12.69 12.26–13.01 12.18–12.87 Maize (crops III and VI) Cu 4.98–5.62 5.02–5.67 5.49–5.92 5.56–5.95 5.37–5.91 5.42–5.95 5.49–6.08 5.63–6.11 Zn 29.76–31.27 29.95–31.32 31.03–31.90 31.27–32.00 30.69–31.89 30.96–32.06 31.14–32.14 31.48–32.22 Mn 39.27–42.51 39.38–42.39 41.67–43.59 42.18–43.61 40.85–43.66 41.07–44.02 42.67–44.45 41.85–44.30 Fe 78.63–88.72 78.95–88.95 81.03–89.90 81.56–89.98 80.79–89.87 81.16–90.03 81.89–90.12 82.06–90.23 Cr 0.47–0.50 0.49–0.62 0.67–094 0.71–0.98 0.65–0.93 0.69–0.99 0.72–1.16 0.83–1.24 Pb 0.09–0.12 0.10–0.14 0.18–0.18 0.17–0.20 0.16–0.17 0.18–0.18 0.19–0.19 0.20–0.21 Ni 0.39–0.46 0.45–0.53 0.61–0.67 0.69–0.63 0.65–0.64 0.66–0.69 0.68–0.71 0.71–0.75

Table 6 Effect of application of LFA on concentration of micronutrients, trace elements, and heavy metals in kernel and grain samples from various crops

LFA lignite fly ash, PM press mud

radionuclides in crops (Skiba 1987) by decreasing the exchangeable fraction of the radionuclides (Butnik and Ischenko 1990).

Conclusions

LFA acted as an excellent soil texture modifier, source of essential plant nutrients, and liming agent for improving soil fertility and crop (groundnut and maize) yield (7–89%). In general, the highest yield dose of LFA was 200 t/ha for both one-time and repeat application. However, appreciable yields were also obtained on application of LFA alone and in combination with press mud at 100 or 50 t/ha (repeat application). The highest dose of LFA (200 t/ha) alone and in combination with press mud showed a better residual effect in increasing the yield of succeeding crops. The organic amendments contributed to the proliferation of microbes in the soil and increased their activities, which played a crucial role in improving the texture and fertility status of the soil, increasing the crop yields, and producing a better residual effect. Besides increasing the OM content of the soil, the organic amendments immobilized toxic trace and heavy metals; hence, there were no adverse conditions. Accordingly, LFA alone and together with press mud and other amendments could be successfully used as a fertility-boosting resource material in the agriculture sector in an eco-friendly manner,

and its use would contribute to solving the fly ash disposal problem.

Acknowledgments The authors are grateful to the Standing Scientific Research Committee, Department of Coal, Ministry of Coal, Government of India, for providing financial assistance and to the director of CFRI and NLC authorities for providing infrastructural facilities. They also thank the director of CFRI for permitting this paper to be published.

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