



Changes in soil properties in a fluvisol (calcaric) amended with coal fly ash

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ABSTRACT

Fluidized bed combustion ash (FBC) is a by-product from coal-fired power stations used for many decades in concrete, cement and brick manufacturing and more recently for trace metal immobilization and pesticide retention in soils. Moreover FBC ash has been recommended by some authors as a soil amendment in agriculture for its nutrient supply. In this study silico-aluminous (SiAl) and sulfo-calcic (SCa) fly ashes have been added to a fluvisol (calcaric) sampled in the Rhine River plain (Germany) in order to investigate the physical, chemical and physico-chemical possible modifications of the amended soil. Earthworms were introduced into microcosms hydrated with a solution of water and CaCl_2 ($3.7 \cdot 10^{-4}$ mg/L; pH = 6.2) and stabilized during 7 and 14 weeks. Batch leaching tests were performed during 6 weeks on the amended soil in order to obtain data on the mobilization potential of major elements (NH_4^+ , Na^+ , K^+ , Mg^{2+} , Cl^- , Ca^{2+} , NO_3^- , SO_4^{2-} , PO_4^{3-} , H_4SiO_4) and trace elements (Sr, Ba, V, Zn, Cu, Co, As, Ni, Mo, Cr, Cd, Pb) during ash–soil–water interactions. The main modification is an increase in pH (≈ 12) of soil water in soil amended with sulfo-calcic ash, whereas soil pH only slightly changes in the other soil as compared to control soil. This alkaline pH causes a strong mortality of the macrofauna and a higher mobility of Co, Ni and V on the contrary to Cr, Cu, Pb and Zn which are less mobile and more strongly retained by soil. Soil physico-chemical properties remain generally similar to control soil in soil amended with silico-aluminous ash (SiAl) but show major differences in the case of soil amended with sulfo-calcic ash (SCa). The sulfo-calcic ash therefore seems less adapted to integration in soils from an agricultural and farming point of view, contrary to the silico-aluminous ash which increases the cation exchange capacity, thus improving the nutrient and water reserve for plants.

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1. Introduction

In a world context where the management of waste, the efficiency of agriculture as well as the quality of the environment become priorities, the fate of industrial ashes is of great concern. Fluidized bed combustion ash (FBC), a by-product from lignite-fired power stations, has been used in concrete, cement and brick manufacturing for many decades (Saint-Ame, 1998). Recently authors have reported about their efficiency in trace metal immobilization in soils and mine tailings and in pesticide retention in soil (Bertocchi et al., 2006; Majumdar and Singh, 2007; Shaobin Wang and Terdkiatburana, 2008).

Some authors studied the properties of soils after amendment with ash (Aitken et al., 1984; Sikka and Kansal, 1994; Desmukh et al., 2000; Grewal et al., 2001; Kalra et al., 2003). When alkaline, fly ash can be added to increase the pH of acidic soils; it can also act as a soil modifier to upgrade the physical properties and improve the texture, upgrade the chemical and biological quality of soil (increase in cation

exchange capacity, capacity of water retention, nutrient availability) and thus participate in the optimization of plant growth (Chang et al., 1977; Phung et al., 1979; El-Mogazi et al., 2001).

Douay et al. (2002) have tested the effect of fly ash integration on the bioavailability of toxic elements in calcium carbonate rich soils but noticed a reduced emergence of the seedlings and delayed growth with respect to the control soil. They explained it by a rise in pH and a poor integration of ashes with the soil. Grumiaux et al. (2007) evidenced toxicity on earthworms after integration of ash in soil and attributed it to soil pH. However, we think that not only pH but also soil texture and structure, and water retention may have played a role in growth delay of seedlings and difficulty for worms to integrate ashes in soil.

Only few studies were devoted to the physico-chemical modifications such as water retention and stability, induced by fly ash amendment into calcium carbonate rich soil. The aim of the present study is to investigate these possible changes of a fluvisol (calcaric) from the Rhine River plain improved by two types of fly ashes (silico-aluminous and sulfo-calcic ashes) considered as decontaminating media.

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2. Material and methods

2.1. Soil sampling and analyses

The soil used in the present study was taken from a periurban zone in the Rhine River plain, close to Strasbourg city (France). According to the World Reference Base for Soil Resources (IUSS Working Group WRB, 2007) the soil is a haplic fluvisol (calcaric) developed over a carbonated sandy loam formation representative of the river alluvia deposited in the Rhine Graben. It comprises a silty loam layer (0–30 cm) overlying more sandy layers (30–100 cm) corresponding to quaternary Rhine alluvia deposited on a deep clayey layer (100–140 cm) interpreted as a Rhine overflow deposit. The soil has already been studied by Lahd Geagea (2007) who also traced the environmental pollution in dust, barks and lichen around the site by Pb, Sr, Nd, and C isotopic ratios, showing a slight contamination in lead in the 0–18 cm A0 horizon, as do most of soils in urban and perinatal areas (Elsass et al., 2006). It was therefore interesting to investigate the influence of fly ash amendment on the physico-chemical properties and trace element retention capacity of this type of soil. The site is located in Germany (UTM 32 coordinates: 570413597N 5384604E; Riehl, 2008). After digging in a soil profile under the native forest, samples were collected from the surface 0–30 cm depth which corresponds to the A horizon.

The physico-chemical characteristics of the fine earth (sieved at 2 mm) were determined using the following standard analytical procedures: for pH and Eh, the standard procedure NF EN 12176; total organic carbon (TOC) by the volume of lost gas during the total dissolution of carbonates by HCl (volumetric calcimetry, NF ISO 10693); organic matter content (OM) by calcination at 375 °C during 16 h after predrying at 150 °C (NF EN 12875); cation exchange capacity (CEC) by the Metson method (NF X31-130, 1993); structural stability using standard procedure NF X 31-515 (June 2005); bulk mineralogy and clay mineralogy by XRD (Brücker D5000, bulk analysis: 3–65° 2 θ scanning angle, clay analysis: 2–30° 2 θ scanning angle, 1 s–0.02° upward step, Cu anticathode, wave length $\lambda_{\text{CuK}\alpha 1}$ = 1.54056 Å, 30 mA current, 40 kV voltage); C, H, S, N analyses were performed by the SCA CNRS laboratory in Lyon by thermal conductimetry or IR detection after combustion at 1050 °C for C, H and N, and 1350 °C for S, with a precision of 0.3%; major and trace elements in soil were analysed by ICP-AES and ICP-MS (Jobin Yvon 124 and VG PQ 2+).

2.2. Fly ash selection and analysis

Fly ash is a residue of combustion of carbon fossil material in coal-fired thermal power plants. Ashes are captured by various filtration processes before gases escape to the atmosphere.

These residues are classified in wastes from power stations and other combustion plants and assigned code number 10 01 02 in the waste list of the European community (decision 2001/118/CE, January 16th, 2001 modifying decision 2000/532/CE as regards the list of waste). They are considered as non hazardous waste contrary to wastes of incineration. Considering French legislation, they are neither classified as special industrial waste (Decree 97–517, May 15th, 1997), nor as dangerous waste (MEDD, 1998, 2002).

Fluidized bed combustion ashes were supplied by Surschiste S.A. and collected from the Carling (SiAl) and Gardanne (SCa) coal thermal power plants, France. They result from the combustion of pulverized coal at low temperature (850–880 °C) and are mainly alkaline.

The chemical composition of SiAl and SCa fly ashes was obtained by ICP-AES analyses; CEC measures were performed by the Metson method. Scanning electron microscopy (SEM) observations and chemical micro analyses were done on a TESCAN Vega 2 XMU Low vacuum, coupled with a micro analytical device (EDAX PEGASUS).

2.3. Microcosm settings

Terra-cotta flowerpots 17 cm deep were used as microcosm settings. Soil samples from the A horizon (0–30 cm) were homogenized and placed in the microcosms and supplemented by FBC moistened ashes (6% in mass of the total mass contained in the microcosms). FBC ash was mixed with the soil by hand.

Five media were tested: artificial soil considered as control (T), artificial soil with silico-aluminous ash (TSiAl (7)) or with sulfo-calcic ash (TSCa (7)) incubated for seven weeks in order to stabilize pH according to the experiments of Grumiaux et al. (2007) and fourteen weeks (TSiAl (14) and TSCa (14) step at which worms may have gained weight. Control soil (T) as well as soils TSiAl (7) and TSCa (7) were duplicated to examine the analytical reproducibility and heterogeneity of the samples.

Earthworms taken from an untreated biological garden were introduced into each microcosm. As they are qualified as structure engineers of soil (Jongmans et al., 2003) they enable reproducing a natural soil structure in microcosms and form aggregates by mixing ash and soil. The first period of microcosm setting procedure was needed as the introduction of ash in soil caused earthworm mortality in TSCa mixtures, probably due to the increase in pH (Grumiaux et al., 2007). In general even in the best conditions of earthworm survival in microcosms, the mortality was higher in TSCa mixtures than in TSiAl.

Microcosms were regularly hydrated with a solution of water and CaCl₂ (3.7 10⁻⁴ mg L⁻¹, pH = 6.2). This choice was made to simulate the pH and ionic strength of natural rainwater, knowing that the rainwater pH in the region ranges between 6.3 and 6.9 (Elsass et al., 2006) and that the ionic strength is nearly 1.10⁻⁴ mol L⁻¹ (Elsass, personal communication, unpublished). This choice is justified by the fact that the pH of water is a key parameter affecting the mobility of trace elements in soil but also in ashes (Jankowski et al., 2006). The choice of the addition of CaCl₂ to pure water, instead of ammonia as chosen by Jankowski et al. (2006) or of MgCl₂ (Rousseau, 2003), was justified considering that an important part of the watering of land comes from the water table or from rivers and has a conductivity of 414 $\mu\text{S cm}^{-1}$. On the other hand the presence of calcium induces flocculation, but as the soil is already saturated in calcium, the addition of CaCl₂ will have no side effect (Tessier et al., 1999). Two samples of the watering solution were analysed by spectrometry (ICP-AES and ICP-MS).

After 7 and 14 weeks, the soils were dried and dry sieved through 2000, 1000, 500, 100 and 50 μm mesh sieves and by hand selection with tweezers of 2 to 5 mm aggregates for structural stability measures. Water adsorption experiments were carried out on less than 500 μm aggregates, considering that they are the first aggregates formed by worms after mixing of soil and ash (Marinissen and Didden, 1997; Jongmans et al., 2003), and thus contain the macro and micro-aggregates the most rich in organo-clay assemblages (Christensen, 1992; Puget, 1997).

2.4. Isothermal sorption of water vapour

The method of relative humidity fixed points (HR) was used for the measurements of humidity established at constant ambient temperature over-saturated salt solutions. Relative humidities were chosen according to the scale of relative humidity of air, certified against saturated salt solutions. They are prescribed by the "Organisation Internationale de Métrologie Légale" (1996) according to the description in the standard ISO 483 and in the national standards [NF X 15-014 and 15-119].

Water adsorption experiments were performed at increasing relative humidity (8.8, 14.2, 22.5, 32.8, 45.9, 64.4, 75.2, 84.7, 92.3 and 93%). An amount of 0.5 g of sample previously dried for 24 h at 110 °C was deposited in a glass cup of known mass and placed in a plastic desiccator (2 L) where the relative humidity was kept constant and fixed by an over-saturated salt solution. The sample lies close to

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