



Effect of industrial residue combinations on availability of elements



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HIGHLIGHTS

- Beneficial combination of fly ash and biosolids.
- Nutrient availability increase.
- Potentially toxic element availability decrease.
- Measured element availability was differed from the calculated leaching potential.

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ABSTRACT

Industrial residues, such as fly ashes and biosolids, contain elements (e.g., N, P, K, S, Ca and Zn) that make them a viable alternative for synthetic fertilizers in forestry and agriculture. However, the use of these materials is often limited due to the presence of potentially toxic substances. It is therefore necessary to assess and, when warranted, modify the chemical and physical form of these and similar waste materials before any advantages are taken of their beneficial properties. Biofuel fly ash, municipal solid waste incineration (MSWI) fly ash, biosolids, peat, peat residues and gypsum board waste were combined in various proportions, and this resulted in increased leaching of N, P, S, Cu and Mn, but decreased leaching of Ca, K, Mg, Cr, Fe, Ni, Zn, Al, As and Pb. Chemical fractionation revealed that elements Ca, K, Mg, S and Mn were predominantly exchangeable, while the rest of the elements were less mobile. Cadmium was mostly exchangeable in MSWI fly ash, but less mobile in biofuel fly ash mixtures. Recycling of MSWI fly ash in the mixtures with fertilizers is considerably less attractive, due to the high levels of salts and exchangeable Cd.

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1. Introduction

The recycling of industrial residues such as biofuel fly ash (BFA), an air pollution control residue from combustion of wood-based fuel, and municipal sewage sludge (biosolids) on soil as the source of nutrients is encouraged [1,2]. In contrast to coal fly ash [3–5], municipal solid waste incineration (MSWI) fly ash is not commonly considered as a source of nutrients, due to the elevated concentrations of undesirable substances. As it was shown in the previous study, application of MSWI fly ash as soil amendment can induce plant toxicity [6]. Since powdered fresh fly ash had a very high alkalinity (pH 12), a direct application to soil was not recommended [6]. Therefore, taking into account recommendations from a

previous study [6], materials treated in form of granules/pellets could be used instead. Granulation/pelletizing is frequently used in order to transform the raw ash into a product that has a lower pH (due to carbonation reactions) and is thus easier to handle. Granulated ash has a smaller surface area compared to ash in a powder form and this leads to a reduced reactivity and a slower release of ash constituents [7–10]. This, in turn, might increase the possibility for plants to take up nutrients over an extended period of time.

In order to further impede the release of various elements and to improve the mechanical properties of granulated fly ash, other materials, such as peat and gypsum [11,12], may be used together with biosolids (BioS). Application of such biosolids to soil increases the content of organic matter, improves the water-holding capacity and, most importantly, supplies N and P. Thus, by combining ash with biosolids, a better balance between the macronutrients (N, P, K) in the material mixtures can be achieved. At the same time, such mixing may lead to chemical reactions which can be expected to reduce the solubility of potentially toxic elements (e.g., Cd and

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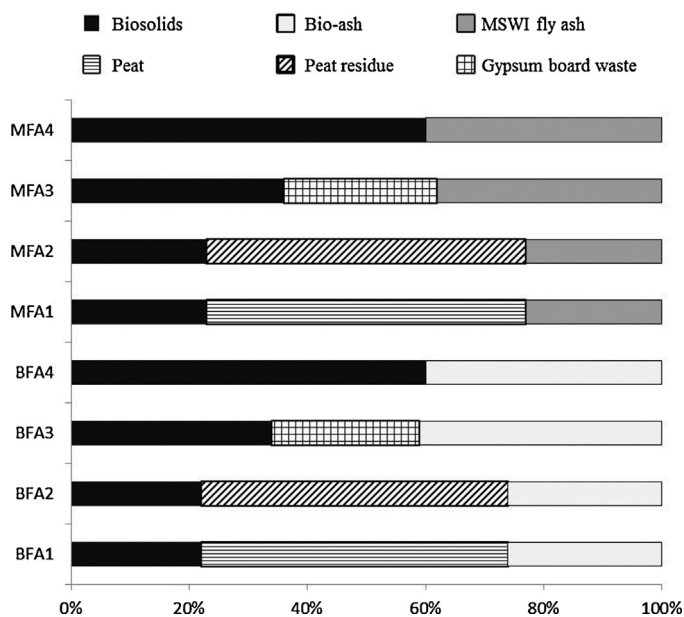


Fig. 1. Proportions of industrial residues and materials used in combinations.

Pb) in ash and biosolids by immobilizing them within the mixture matrix.

The aim of this study was to evaluate potential environmental impacts of composite industrial residues as well as their suitability for use as soil fertilizers by analyzing the element availability in biosolids and ashes, with and without addition of other materials and including modeling of the solubility controlling mineral phases.

Both types of fly ashes, biofuel and MSWI were selected, in order to identify differences between these two contrasting residues in terms of solubility of potentially toxic substances.

2. Materials and methods

2.1. Nutrient rich residues

Fresh and dry (without added water) biofuel fly ash (BFA) from combustion of tree bark was collected from a fluidized bed incinerator. Fresh fly ash from a municipal solid waste incineration was collected and contained about 50% of added lime. The sampling of this ash was done during the two separate occasions. The ash (MFAG) samples during the first occasion were used for the preparation of granules (MFA1–MFA3) and ash (MFAP) sampled later was used for the preparation of pellets (MFA4). The fuel comprised household waste, animal by-products, recycled wood chips, sleepers containing creosote and certain other hazardous wastes. Dewatered, anaerobically digested biosolids were collected from the storage pile at a sewage water treatment plant. All residues originate from northern Sweden.

2.2. Other materials

Commercially available peat (P) and gypsum boards (Gbw) were obtained from a local retailer. Peat residue (Pr) is a by-product, a fine fraction remaining after the production of a sorbent for clean-up applications.

2.3. Preparation of mixtures

Residues were combined in various proportions (Fig. 1). For smaller volumes, this was carried out manually, and for larger volumes a cement blender was used. Biofuel fly ash was mixed with

peat and biosolids (BFA1), biofuel fly ash mixed with peat residues and biosolids (BFA2), biofuel fly ash mixed with gypsum board and biosolids (BFA3) and biofuel ash mixed with biosolids (BFA4). Also, the mixtures were prepared using MSWI fly ash in the same manner as biofuel fly ash and were named as follows: MFA1, MFA2, MFA3 and MFA4.

Gypsum board was milled prior to the mixing. The proportions of the different materials were chosen based on the results of a screening experiment in which a large number of residue combinations were tested. The selection of the proportions in this study was based on the pH values of the mixtures. The material combinations with the pH values closest to neutral were selected in order to avoid N volatilization.

The mixtures MFA1, MFA2, MFA3 and BFA1 were granulated with 30% of water using a shaking plate (all of them contained approximately 50%, w/w biosolids). The mixtures BFA2 and BFA3 did not require additional water due to the high amount of biosolids (65%, w/w). They were granulated manually. All granules were dried at 50 °C for 48 h.

Preparation of large quantities of BFA4 and MFA4 mixtures (intended for a field experiment) was not possible by using the shaking plate. Therefore, pelletizing was chosen instead, and this was done using a pelletizing equipment made of a vibrating perforated plate (Van Aarsen type CT 20, No 88732, The Netherlands). Mixtures MFA4 and BFA4 were dried at 50 °C for 48 h prior to pelletizing to reduce the excess water.

2.4. Element extractions

Batch leaching test. A standard one-stage compliance batch leaching test [13] at a liquid to solid ratio of 10 l/kg (L/S 10) was applied to estimate the concentrations of leachable constituents, electrical conductivity (EC) and pH in materials.

Sequential extraction test. Six steps were applied for element fractionation in 1 g of the material according to the following procedure which has been adjusted to this study. After each step, residues were centrifugation at 10 000 (rpm) for 15 min, washing with 10 ml deionized water (except Fraction III), centrifuged and eluate discarded, while the residues were used in the subsequent steps:

Fraction (I): Exchangeable metals (the most soluble). Extraction was done with 40 ml of 1 mol/l NH_4OAc (adjusted to pH 6.5 acetic acid (HOAc)) by shaking 16 h [14].

Fraction (II): Bound to carbonates. Extraction was done with 8 ml of 1 M NaOAc adjusted to pH 5.0 by agitating for 5 h [15].

Fraction (III): Poorly crystalline Fe(III) oxyhydroxide fraction. Extraction was done with 25 ml of 0.2 M NH_4 -oxalate adjusted to pH 3.0 for 2 h at ambient temperature in darkness, then additional 12.5 ml of solution was added again [16].

Fraction (IV): Crystalline Fe–Mn oxide fraction. Extraction was done with 20 ml 0.04 M $\text{NH}_2\text{OH}\text{--HCl}$ in 25% (v/v) HOAc adjusted to pH 2 in a water bath at 96 °C for 6 h [15].

Fraction (V): Organic matter and secondary sulphide fraction. Extraction was done with 25 ml of 35% H_2O_2 at 85 °C for 1 h [15].

Fraction (VI): Residual fraction. Extraction was done with 15 ml aqua regia ($\text{HNO}_3\text{:HCl}$, 1:3, v/v), in a microwave digester (CEM Microwave Sample Preparation System, Model MARS 5) at 195 °C for 10 min.

AL-extraction. 5.0 ± 0.1 g of the air-dried material was extracted with 100 ml of AL solution containing 0.1 M/l ammonium lactate and 0.4 M/l acetic acid at the pH 3.75 [17]. The extract was immediately agitated for 90 min at room temperature and filtered. The concentration of P was measured in the extracts by inductively coupled plasma optical emission spectroscopy (ICP-OES, Perkin Elmer Optima 2000 DV).

Leaching potential calculation. The theoretical leaching of each element from the mixtures (C_t) was calculated as a sum of the

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