



Geochemistry of barium in soils supplied with carbonatite rock powder and barium uptake to plants[☆]



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ABSTRACT

Apatite-biotite-carbonatite (short form: carbonatite) rock powder originating from Lillebukt alkaline complex, N-Norway is a potential liming and multi-nutrient fertilizer. However, the elevated contents of potentially bio-toxic barium (Ba) (up to 5.5 g kg⁻¹) is a matter of concern. A pot experiment was conducted in order to investigate the geochemistry of Ba in a sandy peat-containing growth media treated with carbonatite, with special focus on Ba-speciation in the pore water. These results were correlated with Ba uptake in *Trifolium repens* L. (white clover) and *Festuca arundinacea* L. (tall fescue). The Ba-speciation and bioavailability as affected by treatment and plant growth was conducted using WHAM/Model VI version 7.0.4 and the diffusive gradients in thin films (DGT) technique, respectively. The increasing cation exchange capacity (CEC) in the growth media, and colloidal fulvic acids (FA) decreased the Ba²⁺ concentration in pore water. Both the Ba detected in DGT (Ba_{dgt}), and exchangeable Ba (Ba_{exc}) were higher in pots containing rock powder. A strong relationship between Ba_{exc} and diffusively bound Ba to FA (Ba_{dfa}) with plant uptake was evident. The latter was likely initiated by organic material mobilization of Ba from carbonatite. The mobilized Ba bound to FA (Ba_{fa}) in pore water was mainly bound diffusively, and the Ba_{dfa} fraction was thus made available to plants. Finally, the Ba_{dgt} was increased in pots grown with white clover compared to tall fescue. Further investigation is required to determine if this is related to increased rhizosphere activity by plants or to the nitrogen fixating plant-microbe relationship.

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1. Introduction

In recent years the use of crushed rocks and minerals as soil amendment in farming practices has received increased attention (Van Straaten, 2006). In addition to its liming potential, the minerals in rock powder weather over time, sustaining steady supply of macro- and micro-nutrients to plants. The carbonatite rock from the Lillebukt alkaline complex, located at Stjernøy in the Northern part of Norway (70° 17.75' N, 22° 34.5' E), contains substantial amounts of calcite (~40% CaCO₃), K (2.7% K, mainly in biotite), phosphorus (2.0% P in apatite) and magnesium (2.7% Mg, mainly in biotite) (Heim et al., 2012). Bakken et al. (1997, 2000), reported K to be easily weathered from the biotite fraction, which in turn was available to plants grown in carbonatite-rock treated soils. However, the potentially hazardous element barium (Ba) can substitute

K in the sheet interlayers of biotite (Shaw and Penczak, 1996) and hence pose undesired Ba-uptake when applying this mineral in agroecosystems. In the carbonatite rock from Stjernøy, Ba replaces up to 10% of K sites in biotite. Since Ba follows the geochemical fate of K due to similar ionic radius (Kabata-Pendias, 2011), Ba may be released to soil in considerable amounts. The fate of K in the soil system and its availability to plants under different conditions has been examined closely (e.g. Marschner, 1993). The exchangeable and free K ions in soil and pore water are easily absorbed by plant roots. Barium on the other hand, may exchange other cations and precipitate in soil and pore water as oxides and hydroxides (Kabata-Pendias, 2011), or in the presence of sulfates and carbonates (Choudhury and Cary, 2001). The use of barium chloride (BaCl₂) method for determination of soil CEC has been adopted by the American Society of Agronomy (Gillman and Sumpter, 1986), due to the Ba's ability to exchange other cations in soil. A field study at Stjernøy comprising 23 native plant species grown in mineral soil developed on carbonatite bedrock, showed that all species were able to take up Ba from the soil. Exceptionally high concentrations of Ba (up to 2 g Ba kg⁻¹ dry weight plant material) in *Vicia cracca* L.

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(bird vetch) were reported (Myrvang et al., 2016a). Studies on Ba uptake by plants show that some species are able to accumulate high concentrations of Ba (Choudhury and Cary, 2001; Kabata-Pendias, 2011). These findings suggest that Ba can be highly mobile and available to plants grown in Ba-rich soils. The literature reports considerable progress in recent years on speciation of trace elements, their mobility and transport as well as the associated ecological risks (Almås et al., 2007; Almås et al., 2006; Degryse et al., 2009; Ivezic et al., 2013; Lofts et al., 2004; Mason et al., 2013; Nolan et al., 2005; Okkenhaug et al., 2015; Zhang and Davison, 2015), still the biogeochemistry of Ba in soil is not equally well investigated. A study on a barite contaminated area reported indications of increased Ba mobility when adsorbed to organic substances in water extracts (Lagas et al., 1984). Lamb et al. (2013) suggested that Ba in pore water was supplied from cation exchange sites associated with surface of soil colloids and thus supplied Ba to plant roots.

Based on earlier findings connected to this study (not yet published), we hypothesize that (i) Ba bioavailability is controlled by the free ion activity of Ba in pore water and lability species on mineral surfaces; (ii) Ba adsorbed to dissolved humic substances will be mobile in soil, and therefore more available to plants; (iii) the release of Ba from carbonatite to soil will increase by supply of peat organic matter. To test the three hypothesis, we conducted a pot experiment with test crops (*Festuca arundinacea* L. and *Trifolium repens* L.) grown in a sand mixed with carbonatite rock powder and added different amounts of peat. The WHAM/Model VI (version 7.0.4) was used to calculate the Ba^{2+} concentration and other Ba species in pore water before sowing, and DGT technique was used to estimate the *in situ* bioavailable metal concentration in pots containing the same soils as affected by rhizosphere activity.

2. Methods and materials

2.1. Characteristics of the growth media used in the pot experiment

The primary growth medium was an aeolian sand (0.06–0.6 mm) with a low content of total carbon (0.015 wt%). The sand composition is mainly quartz and feldspar originating from weathering of arkosic and granitic bedrock, where Ba is supposed to replace K and Ca in the non-exchangeable silica lattice of feldspars (Heier, 1962). Total Ba concentration (X-ray Fluorescence-analysis) in the sand and carbonatite was $500 \pm 5 \text{ mg kg}^{-1}$, and $2500 \pm 110 \text{ mg kg}^{-1}$, respectively. The concentrations of Ba in peat ($3.8 \pm 1 \text{ mg kg}^{-1}$) and dolomite ($9.4 \pm 0.51 \text{ mg kg}^{-1}$) was determined by means of ICP-OES (Perkin Elmer Optima 5300DV) after digestion in concentrated nitric acid (65 wt% sub-boiled ultra-pure HNO_3) using microwave technique (Milestone UltraClave 3; hold time 25 min at the maximum temperature 250 °C). The content of organic C was determined using a LECO TruSpec analyzer (Table 1). The pH in a soil-water suspension (1:2.5) was determined by means of a Thermo Scientific Orion2Star pH benchtop pH meter and a combined glass membrane electrode. Cation exchange capacity (CEC) and exchangeable Ba (Ba_{exc}) were measured by

extracting 3.00 g dry soil in 25 mL 1 M ammonium acetate pH 7.0 for 24 h followed by filtration through blue ribbon filter. Metal concentrations were determined by means of ICP-OES, and CEC was calculated as the sum of protons, K, sodium (Na), Ca, and Mg (Tables 1 and 2). Reagents of analytical grade or better were used throughout, and all water used was de-ionized ($>18 \text{ M}\Omega \text{ cm}^{-1}$).

2.2. Pot experiment

Trifolium repens L. (white clover) and *Festuca arundacea* L. (tall fescue) (Lid and Lid, 1998) were cultivated in a greenhouse experiment. Initially 2.1 kg dry matter (DM) of sand were mixed with 50, 200, and 500 g, respectively, of unfertilized peat DM (equal to about 2, 10, and 20 wt% peat per pot) in 3 dm^{-3} non-perforated pots. Porosity was improved in pots of 2 and 10 wt% peat by adding inert polystyrene balls adjusting to a total soil volume of 3 dm^3 . The fourth growth medium consisted of 1000 g peat DM equivalent to 3 dm^3 . The pH was optimized to range within 5–7 in all the pots. Sand and peat mixes that were not supplied with carbonatite, were supplied with dolomite to an equal lime effect (Table 2). In the control pots (Table 2), 50 mL of 0.075 M NaOH was supplied to increase pH. Sand mixed with 2 wt% was not given 150 g carbonatite, as the pH would increase beyond 7.5. Thereafter liquid fertilizers were applied to treatments grown with tall fescue, corresponding to 300, 75, and 300 mg pot^{-1} of N, P, and K, respectively. The white clover cultivated pots were added liquid fertilizers corresponding to 180, 75, and 300 mg pot^{-1} of N, P, and K, respectively. Initially seeds of white clover were germinated in a 10 mL soil naturally containing rhizobium bacteria for inoculation, and seedlings were pricked to their respective pots. Per pot, 12 seeds of tall fescue and 4 seedlings of white clover were applied. Each treatment was conducted with four replicates. Plants were grown under environmentally controlled conditions with a temperature at $19 \pm 1 \text{ }^\circ\text{C}$ during day (16 h) and $15 \pm 1 \text{ }^\circ\text{C}$ at night (8 h). In order to simulate day light, 500 W lamps emitting yellow and white light in equal intensity, with an approximate photon flux of $200 \mu\text{mol m}^{-2} \text{ s}^{-1}$ at day time, were used. The soil humidity was kept near 60% of the potential water holding capacity by measuring total weight every second day. Weight of each pot was balanced by deionized water throughout the experimental period.

2.3. Plant harvest and analysis

Leaves and stem of tall fescue and trifoliolate leaves of white clover were harvested at 6, 10, and 14 weeks after sowing. Plant samples were dried at 50 °C for 72 h before recording the DM yield. Subsequently, plant samples were milled, and approximately 0.2 g accurately weighed samples were digested at 250 °C (25 min at maximum temperature) with 65 wt% HNO_3 (sub-boiled ultrapure), using microwave technique. The plant concentrations of Ba in addition to aluminium (Al), Ca, K, Mg, sodium (Na), sulfur (S), and iron (Fe) were determined using ICP-MS (Agilent 8800 Triple Quadrupole).

Table 1
Selected properties (mean and standard deviation, $n = 3$) of carbonatite, dolomite, aeolian sand and unfertilized peat before application.

	pH _{H2O}	CEC (cmol+ kg ⁻¹)	BS (%)	Total C (%)	Exchangeable Ba (cmol+ kg ⁻¹)
Carbonatite	9.3 ^a	65 ± 5.4	100	4.5 ± 0.23	0.13 ± 0.011
Dolomite	9.5 ^a	38.7 ± 0.95	100	12.56 ± 0.070	0.0014 ± 0.00011
Aeolian sand	4.7 ^a	0.51 ± 0.024	100	0.015 ± 0.0028	0.0310 ± 0.00045
Unfertilized peat	3.8 ^a	126 ± 1.8	9.5 ± 0.35	50.8 ± 0.37	0.0093 ± 0.00075

^a $n = 1$.

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