



Black carbon and black nitrogen storage under long-term paddy and non-paddy management in major reference soil groups



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ARTICLE INFO

Article history:

Received 21 March 2016

Received in revised form 21 August 2016

Accepted 29 August 2016

Available online 24 September 2016

Keywords:

Clay-size fraction

Iron oxides

Aluminum oxides

Black carbon

Black nitrogen

C sequestration

ABSTRACT

Crop-residue burning and frequent paddy-soil flooding can lead to a substantial accumulation of black carbon (BC), thus contributing to long-term C sequestration. There is evidence that the turnover of BC in soils also depends on the soil mineral assembly. We studied the effects of paddy and non-paddy soil management and different major reference soil groups on BC storage. We hypothesized that overall BC storage in soil relates to paddy management and the abundance of reactive mineral phases such as Fe and Al oxides, and clay-sized minerals. Parallel to BC, black nitrogen (BN) should accumulate in soil.

Paddy and non-paddy soils were sampled in three different climate zones (tropical, subtropical, and temperate). The soil profiles comprised six replicates of Andosols, Alisols and Vertisols from Java (Indonesia), and Alisols and Cambisols from China, as well as one Fluvisol and Gleysol from Northern Italy.

Samples were taken by horizon down to >1 m depth and analyzed for soil organic carbon (SOC) and BC. The latter was analyzed by oxidation to benzene polycarboxylic acids. Abundance of BN (as aromatic N) was estimated by X-ray photoelectron spectroscopic analyses of selected topsoil horizons.

In topsoils BC vs. SOC accumulation was affected by management for Andosol, Alisols in China, and Vertisols. However, both flooding and crop-residue management seemed to control this. BC contents relative to SOC also differed between the reference soil groups, independent of management ($p < 0.0001$), yet were surprisingly constant within replicates. We conclude that BC co-accumulated with SOC in all soils. However, the overall storage of BC (1 m depth) was affected by a combination of soil group and management. Vertisols contained the largest BC stocks (17–19 t ha⁻¹ in non-paddy and paddy fields), followed by Andosols and Alisols (6–10 t BC ha⁻¹ under paddy management; 3–8 t ha⁻¹ under non-paddy management). The Gleysol and Fluvisol had the smallest BC stocks, independent of soil use (3–4 t ha⁻¹).

Aromatic N proportions increased to >50% of total N after combustion of rice straw. However, aromatic N was barely, or not detectable in soil, and there was no correlation to BC. We conclude that burned crop residues were not a major source for aromatic N in soil. BC and aromatic N showed no distinct relations to soil properties, such as the abundance of clay-sized minerals, and Al and Fe oxides. Differences in BC stocks between the soils were most pronounced in the subsoils, likely caused by physical processes, such as swelling and shrinking of clays and/or translocation by leaching. Climate and regional soil-adjusted management also affected BC accumulation, but this first snapshot indicates that global BC maps may be linked to global soil maps.

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1. Introduction

Rice cultivation feeds about half of the human population (Maclean et al., 2002). Burning rice straw on the fields might significantly affect the carbon cycle. Gaddé et al. (2009) calculated for Asian countries (where most rice cultivation takes place) that 23 to 95% of rice straw

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residues are burned. This may contribute to atmospheric CO₂ and global warming, but residues of biomass combustion (black carbon, BC) may also contribute to relatively persistent C in soil (e.g., Seiler and Crutzen, 1980).

We previously showed that long-term paddy management on marsh sediments in China resulted in equilibrium BC accumulation and degradation. These Cambisols stored ~13 t BC ha⁻¹ after 300 years of paddy management. Adjacent non-paddy systems had only 7 t BC ha⁻¹ (Lehtndorff et al., 2014). However, little is known about BC storage in other paddy soils, particularly if BC storage might systematically change in well-developed soils of different origins and mineral compositions.

Black carbon is recalcitrant in nature (e.g., Seiler and Crutzen, 1980). It comprises a range of incompletely burned organic matter (OM) characterized by fuel and combustion conditions (temperature and oxygen supply) (Czimeczik et al., 2002; Keiluweit et al., 2010). Mean residence time (MRT) estimates in soil ranges from decades to millennia (e.g., Fang et al., 2014; Kuzyakov et al., 2014; Singh et al., 2012a). Differences in BC stability have been mainly related to the degree of aromatic condensation, which increases as burning temperature increases and depends on the fuel used (e.g., Singh et al., 2012b). On-field burning of rice straw or other crop residues yields BC equivalent to that of grass burned at about 300–400 °C (Lehtndorff et al., 2014; Wolf et al., 2013).

Once incorporated into soil, the turnover of soil BC might additionally be controlled by interactions with minerals (Cheng et al., 2006; Cusack et al., 2012). For example, BC stocks in a weathering sequence of volcanic soils were closely related to the contents of reactive, short-range order minerals (Cusack et al., 2012). Incubation experiments also hinted that minerals may have a dominant role in BC turnover: BC in Ferralsols with high (crystalline) Fe oxide concentration had longer MRT (235–106 years) than BC in clay mineral-rich Vertisols (218–44 years for incubation temperatures of 20° and 40 °C, respectively) (Fang et al., 2014). The authors concluded that variably charged oxide minerals stabilized BC better than the permanently charged smectites in the Vertisol. However, the authors also admitted that MRTs might differ dramatically under field conditions, due to other factors such as variations in climate conditions.

BC might also stabilize by occlusion by soil minerals (e.g., Brodowski et al., 2006). In paddy soils, changing redox conditions may lead to BC stabilization by reaction with, or occlusion in, frequently precipitated short-range order Fe oxides. Effects may vanish by reductive dissolution of Fe oxide-OM associations (e.g., Kirk, 2004; Winkler et al., 2016). In clay-rich soils, BC may be physically protected in deeper soil horizons upon swelling and shrinking (self-plowing due to deep cracks that develop in dry seasons, which allow fresh OM to directly enter the subsoil). These effects may alter the residence time of BC to an unknown extent. In contrast, Fang et al.'s (2014) incubation study implied that Vertisols contain less BC than Ferralsols, and at a more advanced stage of degradation. In any case, it seems reasonable to assume that the storage of BC in soil relates to its mineral assembly.

Amounts and composition of BC in soil can be estimated by a variety of methods, but each has limitations. These limitations are mainly due to large differences in the BC materials. Previous studies showed that oxidation of BC to benzene polycarboxylic acids (BPCAs) recovered at least 70% of BC as BPCA in soil (Hammes et al., 2007; Roth et al., 2012). The relative amounts of five- to six-times carboxylated BPCAs relate to the degree of aromatic condensation of BC in soil (McBeath et al., 2011; Schneider et al., 2010). This is a result of the variation in BC surface/interior ratios. In other words, the less the proportion of six-times carboxylated BPCA (mellitic acid; B6CA), the larger the surface of BC (Glaser et al., 1998). The composition of BPCAs may be related to the source of BC, indicating if it stems from biomass, such as rice straw burning, or industrial combustion processes (Lehtndorff et al., 2015; Wolf et al., 2013). Additionally, it was assumed that BC degradation leads to large, oxidized, negatively charged surfaces that react with the soil matrix, thereby conserving BC (Brodowski et al., 2005a; Cheng et al., 2006). This

effect may differ for aerobic (non-paddy) and anaerobic (paddy) management conditions. BC accumulation, especially in subsoil, may be accompanied by a relative loss of mellitic acid due to decondensation (Rodionov et al., 2010). Singh et al. (2012a) reported that BC characterized by smaller proportions of mellitic acid was more prone to stabilization processes than condensed counterparts. In this line, both preferential stabilization of decondensed BC and intrinsic stability may contribute to its long residence time in soil (e.g., Singh et al., 2012a).

Burning OM also produces BN, but its role in N storage and cycling is not yet understood (de la Rosa and Knicker, 2011; Knicker, 2007). In previous studies, we found that at least 50% of paddy soil N was abundant in unidentified forms, or not bound to microbial residues (Roth et al., 2011) and proteinaceous amino acid N (unpublished data). X-ray photoelectron spectroscopy (XPS) may reveal if straw burning in paddy management leads to accumulation of N in heterocyclic, aromatic forms as previously detected in other soils (Abe et al., 2005; Mikutta et al., 2009). For example, Ding et al. (2014) related the aromatic N peak in the XPS N 1s spectra to BN in water samples and found a coupling between dissolved BN and BC. We tested if the method is applicable to rice char and soil, and if there is a correlation of BC to aromatic N.

The main objectives of this study were to elucidate the possible effects of paddy versus non-paddy management, and different major reference soil groups on the storage of BC. We sampled pairs of paddy and adjacent non-paddy soil depth profiles from different major soil groups of the world (IUSS Working Group, 2014), analyzed them for BC, and compared this to pedogenic oxide contents and soil texture (see also Winkler et al., 2016). This inevitably included a sampling from different climatic and land-use regions, since soil mineralogical settings were specific to geographic regions and parent material. Since the accumulation of aromatic N in soil due to inputs of BN is not yet well understood, we aimed to detect parallels in soil BC and BN contents. Sampling was done for well-developed soils that were under permanent paddy and non-paddy management during at least the last century.

2. Materials and methods

2.1. Sampling

Three tropical paddy and corresponding non-paddy soils formed on volcanic and clay-rich geological substrates were sampled in Java, Indonesia. To account for regional factors relevant to soil formation, we compared their properties to two subtropical paddy/non-paddy soils from China, and a temperate paddy/non-paddy soil in Mediterranean Italy, where there was no on-field rice straw burning (Fig. 1). Soil classification was done according to the World Reference Base for Soil Resources (IUSS Working Group, 2014) (Table 1). At each site, six profiles were dug into three independent paddy and three non-paddy fields (one main and two subsites per soil and management type). However, only one profile per management was sampled in Italy (Table 1). The profiles were sampled according to horizons to at least a 100 cm soil depth. BC and BN analyses were done for selected horizons, and only the main sites were investigated in full-depth resolution (see Tables 2a, b, and 3).

The sampled soils under non-paddy management were classified as Andosols (Indonesia), Alisols (Indonesia and China), Vertisols (Indonesia), Cambisols (China), and Fluvisol (Italy). Upon long-term paddy management, Anthrosols formed from these soils (except for Italian paddy soil, classified as Gleysol). All sites had been continuously cropped for generations, according to local farmers. To account for the specific properties of the soils under study, we simplified the nomenclature and referred to the parent Andosol, Alisol, Cambisol, and Vertisol for both paddy and non-paddy managed sites (except for the Fluvisol/Gleysol pair at the Italian site). Soil details are described next.

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