



# Fire-induced formation and biodegradation of humic substances in Andosols of Japan



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## ABSTRACT

The aim of this article was to elucidate the possible mechanisms of the formation and degradation of humic substances in Japanese Andosols. Charred plant residues (CPRs) produced as a result of human activity and wildfires are incorporated into soils and degraded into smaller particles, i.e. charred plant fragments (CPFs). During this process, some CPRs are transformed into fulvic acids and, especially, humic acids with graphite-like structures. CPR-derived CPFs and humic substances are widely distributed in Japanese Andosols and contribute to their high carbon accumulation. The biostabilities of Andosol humic substances are higher than that of non-Andosols. Our results show that plant charring is a new concept related to the formation and storage of humic substances in Japanese Andosols.

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## 1. Introduction

Soil organic matter (SOM) is an integral part of the soil that affects the fertility and the physical, chemical, and biological properties of soils, thereby influencing crop productivity. SOM is the largest carbon

(C) pool (1500 Gt) in terrestrial ecosystems and environments. The soil C pool constitutes about two-thirds of the global terrestrial C pool, and is twice the size of the atmospheric C pool (Batjes, 1996). Furthermore, since soil organic C (SOC) is generally refractory, it is closely connected with global C cycling and global warming (Lal, 2004; Swift, 2001). The SOC stock results from the balance between the input and output of C in soils. Thus, the mechanisms of the formation and decomposition of SOM must be studied in detail.

Traditionally, SOM has been classified into humic and non-humic substances. Furthermore, humic substances have been divided into

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humic acid (HA, alkali-soluble and acid-insoluble), fulvic acid (FA, alkali- and acid-soluble), and humin (alkali-insoluble) fractions. The mechanisms that underlie the formation of humic substances, which comprise the majority of SOM, still need to be clarified because these very complex processes may involve numerous organic components as starting materials, as well as many reactions such as oxidation, polymerization, condensation, and hydrolysis. Furthermore, the synthesis of humic substances can proceed abiotically (e.g. Shindo and Huang, 1982) and biotically (e.g. Ahn et al., 2006), with the mediation of inorganic components, microorganisms, and enzymes. Previous studies (e.g. Stevenson, 1982) have suggested several hypotheses, known as the lignin, polyphenol, and sugar-amine condensation theories, for the formation of humic substances. Recently, an integrated polyphenol–Maillard reaction pathway was proposed (Huang and Hardie, 2009).

On the other hand, in the case of Japanese volcanic soils (Andosols or Andisols), which display a thick black or blackish-brown A horizon, it is thought that grassland plants such as susuki (Japanese pampas grass or *Eulalia*, *Miscanthus sinensis*, a C<sub>4</sub> plant) are the primary sources of the abundant humus, which may reach 300 g kg<sup>-1</sup> (Dahlgren et al., 2004; Ministry of Agriculture and Forestry Japanese Government [MAFJG], 1964; Shoji et al., 1990, 1994; Yamane, 1973; Wada, 1986). Furthermore, the burning of vegetation is necessary to maintain grasslands for a long period of time, because the climax vegetation under the meteorological conditions that prevail in Japan is forest (MAFJG, 1964; Yamane, 1973). Nevertheless, very little information was available on the relationships between charred plant residues (CPRs) and the quality and quantity of SOM until we initiated a series of studies on CPRs in Japanese Andosols, mainly from the perspective of soil analysis.

Kumada et al. (1967) proposed that soil HAs could be classified into Types A, B, P, and Rp based on their optical properties, using  $\Delta \log K$  and  $RF$  values ( $\Delta \log K$ : the logarithm of the ratio of the absorbance of the HA at 400 nm to that at 600 nm;  $RF$ : the absorbance of HA at 600 nm multiplied by 1000, divided by the volume in mL of 0.02 mol L<sup>-1</sup> KMnO<sub>4</sub> consumed per 30 mL HA solution), where the degree of humification (as reflected by the darkness of the color) of Type A is the highest of all soil HAs. An important feature of SOM in surface and buried A horizons of Japanese Andosols is the predominance of Type A HAs with a graphite-like (turbostratic) structure, which exhibits two absorption peaks at  $d = 0.35$  and  $0.21$  nm, corresponding to the (002)- and (10)-bands of graphite, respectively (Kumada, 1987; Matsui et al., 1984). The intense black color of soil humus is attributed to the accumulation of Type A HAs (e.g. Kumada, 1987). Thus, in our studies, we have referred to Type A HAs as black HAs (Shindo et al., 2004a, 2005).

Although the chemical structures of black HAs have not yet been determined, Japanese researchers have shown that black HAs are distinguishable from other types of HAs, based on their higher absorbance of 1% HA solution at 600 nm (Kumada, 1987), their higher aromaticity (Maie et al., 2002), the larger size of their C-layer planes with condensed aromatic structures (Ikeya et al., 2011), their higher content of oxygen-containing functional groups such as carboxylic groups (e.g. Tsutsuki and Kuwatsuka, 1978), and their lower content of identifiable components such as fatty and phenolic acids (Ikeya et al., 2004).

Similar to Andosols, chernozemic soils (Chernozems or Mollisols) also display a black or blackish-brown A horizon that contains black HAs (Kumada, 1987; Nishimura et al., 2009), black C (BC; Schmidt et al., 1999), or charred plant fragments (CPFs, the so-called degradation products of CPRs, as described later; Nishimura et al., 2009). However, the soil-forming conditions, such as parent material, climate, and vegetation, differ completely between these soil groups (Food and Agriculture Organization of the United Nations, 2014). The reasons why black HAs are formed in Andosols have not been explained scientifically. In terrestrial ecosystems and environments, like Japanese Andosols, CPRs are produced during the burning of vegetation as a result of human activity and wildfires, and they are frequently found in soils (e.g. Kuhlbusch, 1998). Furthermore, carbonaceous materials such as charcoal and charred wood residues have been assumed to be

possible sources of soil humus (Kumada, 1983). Thus, we favored CPRs as the possible origin of black HAs, and initiated extensive studies on the role of *plant charring* in the formation and accumulation of black HAs in soils, with special reference to Japanese Andosols (Shindo et al., 1986a, 1986b, 1986c).

The fate, dynamics, and function of humic substances in soils are largely controlled by their resistance against abiotic (involving weathering) and biotic decomposition (mineralization to CO<sub>2</sub>). Thus, we also investigated the biodegradation of humic substances.

The formation and decomposition of humic substances in soil environments, both of which are mediated by abiotic and biotic agents under the influence of oxygen and moisture, may be slow transformation processes. An experimental model (experimental pedology) may provide an effective approach for clarifying the mechanisms by which they operate. We therefore employed experimental pedology, in addition to soil analysis, to investigate the formation and subsequent degradation of humic substances.

The aim of this article was to elucidate the possible mechanisms of the formation and degradation of humic substances in Japanese Andosols, mainly by integrating previous studies on pyrogenic organic matter, which were performed from the perspectives of experimental pedology and soil analysis.

Black C, which is produced by incomplete combustion of fossil fuels and vegetation (Goldberg, 1985), and exists as a continuum from char to charcoal to soot, is ubiquitous and can be found in soils, sediments, ice, and the atmosphere (e.g. Forbes et al., 2006; Masiello, 2004; Schmidt and Noack, 2000). Recently, biochar, which is formed as a result of the pyrolysis (thermochemical decomposition in a low/zero-oxygen environment) of plant biomass and then applied to agricultural soils as a soil supplement, has received increased attention because of its potential agricultural, environmental, and economic benefits (e.g. Kookana et al., 2011; Lehmann, 2007; Sohi et al., 2010). Since both BC and biochar contain either CPRs or CPFs, the present article provides valuable information for a better understanding of the structural transformation of BC and biochar in soil environments.

## 2. Charring processes of herbaceous and woody plants

Since the 1990s, numerous studies have been conducted using different biomass types and charring conditions to elucidate the formation process of BC and to make useful biochar products. The results obtained in these studies clearly show that the quality and quantity of charred biomass are influenced by biomass factors (e.g. type, volume, and condition of the plant) as well as environmental factors (e.g. temperature and duration of burning or pyrolysis; Baldock and Smernik, 2002; Bergeron et al., 2013; Masiello, 2004; McBeath et al., 2013; Preston and Schmidt, 2006; Schmidt and Noack, 2000). In addition, studies of the physicochemical and spectroscopic characteristics of BC and biochar have indicated that they usually possess condensed aromatic structures, although the degree of condensation depends on biomass factors as well as environmental factors (Baldock and Smernik, 2002; Knicker, 2007, 2011; Masiello, 2004; Preston and Schmidt, 2006; Schmidt and Noack, 2000; Soucémariadin et al., 2013). Furthermore, new condensed aromatic structures are formed from plant components such as cellulose, lignin, and lipids during burning or pyrolysis (Almendros et al., 1997, 2003; Baldock and Smernik, 2002; Knicker et al., 2008).

We investigated the physicochemical properties (elemental composition and X-ray diffraction [XRD] pattern) and spectroscopic properties (ultra violet [UV], visible, Fourier transform-infrared [FT-IR], and <sup>13</sup>C nuclear magnetic resonance [NMR] spectra) of CPRs prepared in the laboratory (at up to 450 °C in an oven) and those resulting from prescribed grassland fires. The results can be summarized as follows: (i) the charring process in susuki, which is a representative grassland plant in Japan, was characterized mainly by the disappearance of the cellulose structure and the formation of a black HA-like substance (HA-LS) with polycondensed aromatic structures and an FA-like substance (FA-LS;

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