



Linkages between aggregate formation, porosity and soil chemical properties



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ABSTRACT

Linkages between soil structure and physical–chemical soil properties are still poorly understood due to the wide size-range at which aggregation occurs and the variety of aggregation factors involved. To improve understanding of these processes, we collected data on aggregate fractions, soil porosity, texture and chemical soil properties of 127 soil samples from three European Critical Zone Observatories. First, we assessed mechanistic linkages between porosity and aggregates. There was no correlation between the fractions of dry-sieved aggregates (>1 mm, DSA) and water-stable aggregates (>0.25 μm, WSA). Soil microporosity and micro + mesoporosity increased with increasing abundance of aggregates, though this correlation was only significant for the WSA fraction. The fraction of DSA did not affect the overall porosity of the soil, but affected the ratio between micro- and mesopores ($\theta_{30 \text{ kPa}}/\theta_{0.25 \text{ kPa}}$), suggesting that micropores are dominantly located within DSA whereas mesopores are located in between DSA and loose particles. Second, we studied the relations between the physical and chemical soil properties and soil structure. Soil texture had only a minor effect on the fractions of WSA and DSA whereas Fe-(hydr)oxide content was correlated positively with both WSA fraction and porosity. This may be attributed to Fe-(hydr)oxides providing adsorption sites for organic substances on larger minerals, thereby enabling poorly reactive mineral particles to be taken up in the network of organic substances. The fraction of WSA increased with an increase in the soil organic carbon (SOC) and Fe-(hydr)oxides content and with a decrease in pH. This pH-effect can be explained by the enhanced coagulation of organically-coated particles at a lower pH. Overall, this study indicates that mechanistic linkages exist between soil chemical properties, aggregate formation and soil porosity.

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1. Introduction

Soils provide major ecosystem services such as food production, water retention and sequestration of soil organic carbon (SOC). The physical structure of the soil plays a crucial role in the processes that facilitate these soil functions. Yet, intensive land use and poor agricultural management practices have led to declines in soil structure, leading to a decrease in the agricultural productivity and SOC stabilization capacity of soils worldwide (Banwart, 2011; Montgomery, 2007). Soil structure is the result of multiple physical, chemical and biological interactions in the soil. However, the complex interactions between chemical properties and soil structure are still poorly understood, which

limits our ability to improve soil structural properties through management. Therefore, there is a need for a mechanistic understanding of the physical and chemical key factors controlling the formation of soil aggregates from loose mineral particles.

Soil structure has been defined as ‘the size, shape and characteristics of soil particles, aggregates, and pores across the size-range from nanometers to centimeters’ (Bronick and Lal, 2005; Oades, 1993). This definition implies that soil structure is the product of various structural entities that cover a size range of several orders of magnitude. According to the widely used aggregate hierarchy theory (Oades and Waters, 1991), soil aggregates and soil pores are formed in a hierarchical manner meaning that microaggregates (<0.25 mm) are the building blocks for macro-aggregates (>0.25 mm) and that aggregation is controlled by different aggregation factors at each size scale. In order to verify the aggregate hierarchy theory, we aimed to quantify soil structure at

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the various hierarchical levels by a combination of (i) the size distribution and stability of the aggregates and (ii) the volume and size distribution of the pores. Soil aggregates and soil porosity reflect different aspects of the structure, with micropores reflecting much smaller structural entities than mesopores, and water-stable aggregates (WSA) reflecting much stronger and smaller aggregates than dry-sieved aggregates (DSA). Because pores are the voids within and between aggregates, the number of aggregates and the pore volume are related. However, experimental evidence for the relation between aggregate fractions and porosity for various soil types is still scarce and limited to a small number of soil samples collected for a narrow range of soil types (Boix Fayos, 1997; Nyamangara et al., 2001). Insight into these relationships can help to understand how the various hierarchical levels of aggregate formation are related.

The interplay between the various physical and chemical processes involved in aggregate formation is highly complex and affected by a range of soil properties. Various factors have been reported in literature to affect or control aggregation (Bronick and Lal, 2005), including (i) soil texture, (ii) the amount of metal-(hydr)oxides, (iii) the amount and characteristics of organic binding agents and (iv) pH and the concentration of multivalent cations. Clay minerals affect aggregation because of their interactions with organic matter (OM), leading to the formation of organo-mineral assemblages (Angers, 1998; Fernández-Ugalde et al., 2013; Six et al., 2000a). Similar, Fe-(hydr)oxides play an important role in the formation of organic-mineral assemblages due to their strong interactions with humic acids (HA) (Asano and Wagai, 2014; Lehtinen et al., 2014; Pronk et al., 2011). However, the role of Fe-(hydr)oxides is mainly studied in oxide-rich soils (Barthès et al., 2008; Duiker et al., 2003; Igwe et al., 2009); their role in soils with low Fe-(hydr)oxide contents is still poorly understood. OM plays a crucial role in the aggregation of mineral particles and is by far the most intensively studied aggregation factor in literature (Angers, 1998; Bronick and Lal, 2005; Dilkova et al., 1998; Pulleman and Marinissen, 2004; Six et al., 2004). OM is a complex mixture of multiple organic substances such as humic-like substances, polysaccharides, proteins, lignin and poorly soluble aliphatic compounds (Calabi-Floody et al., 2011; Wattel-Koekkoek et al., 2001) and this range of properties enables OM to affect aggregation via different mechanisms and at different scales (Tisdall and Oades, 1982). Another factor that affects aggregation is the amount of exchangeable multi-valent cations, such as Ca^{2+} , Al^{3+} and Fe^{3+} , since the coagulation behaviour of particles and the strength of the interactions between OM and mineral particles strongly depends on the concentrations of multi-valent cations (Curtin et al., 1994; Kaiser et al., 2012). Soil pH has a strong effect on the concentration of multivalent cations in the soil solution because, in acid soils, the concentration of metal ions is often controlled by the solubility of their respective metal-(hydr)oxides and in calcareous soils, the solubility of CaCO_3 controls the concentration of Ca (Weng et al., 2011).

Despite the extensive knowledge on aggregation mechanisms (Bronick and Lal, 2005; Six et al., 2004), a quantitative understanding of the relationships between physical/chemical properties and soil structure is still lacking. Therefore, our aim was to assess the contribution of the different aggregation factors on formation of soil structure at the various hierarchical levels using porosity, water-stable aggregates (WSA) and dry-sieved aggregates (DSA) as indicators for aggregation at these different levels. We hypothesize that the volume of micropores is related to the abundance of small aggregates and that the volume of mesopores is related to the abundance of larger aggregates. We furthermore hypothesized soil structure to be the product of multiple physical and chemical interactions, and thus that aggregation is best predicted by a combination of texture, Fe-(hydr)oxide content, organic matter content and pH. Finally, we hypothesized that the dominant aggregation factor differs for the different hierarchical levels of aggregate formation as are in this study reflected by the various soil physical indicators, in which we expect that Fe-(hydr)oxides and clay particles are the dominant aggregation mechanisms at the smallest size scales,

and that SOC content and pH are dominant at the larger size scales. These hypotheses are tested with a dataset of physical and chemical soil properties collected from 12 different sites in three diverse European Critical Zone Observatories (CZO's).

2. Material and methods

2.1. Sites

The soil samples for this study were collected from 12 sampling sites at three European Critical Zone Observatories (CZO's), which are described in detail by Banwart et al. (2012). The CZO's are characterized by different parent material, land use, and climate and are located in Austria (Marchfeld CZO), in the Czech Republic (Slavkov Forest CZO), and on the island of Crete, Greece (Koiliaris CZO):

The *Marchfeld CZO* is located on the former floodplains of the River Danube downstream of Vienna in Austria. This CZO includes six sites that are part of a chronosequence (20–20,000 years old) (Lair et al., 2009a). Climate is temperate with dry and hot summers, and soils are covered by deciduous forestry and annual crops. The soils, classified as Fluvisols and Chernozems, are calcareous and the clay mineralogy is dominated by illite and chlorite (Banwart et al., 2012). This CZO has been extensively studied, in particular in terms of soil age (Lair et al., 2009a), phosphorus speciation and adsorption (Lair et al., 2009b; Zehetner et al., 2008) and carbon sequestration (Zehetner et al., 2009).

The *Koiliaris CZO* is located on Crete, east of the city of Chania. Climate is Mediterranean and soils are covered by natural vegetation (shrubs and herbaceous species). The three sites have been formed at different sedimentary deposits: fluvial, unconsolidated and consolidated bedrock. Soils are classified as Fluvisols (site K1,2) and Cambisols (site K3), respectively, and are degraded due to deforestation for cropping and grazing. Organic matter contents and aggregate stability are therefore low (Stamati et al., 2013) and soils are susceptible to erosion and desertification. Further information about this CZO can be found in Stamati et al. (2013a) and Moraetis et al. (2014).

The Slavkov Forest CZO, in western Czech Republic, is located in a national protected area. Climate is temperate and the soils are used for forestry (mainly Norway spruce). This CZO includes three sites, which are located on the slopes of three first-order catchments (Lysina, Pluhuv Bor, Na Zelenem). The catchments are characterized by different parent material (granite, serpentinite and amphibolite) and therefore by three distinct soil types (Podzol, Stagnosol and Cambisol, respectively). They therefore show strong differences with respect to their mineralogy and chemistry (Table 1). Detailed information about the chemistry and hydrology of the three catchments can be found in Kram et al. (2012) and Banwart et al. (2012). Site coordinates are given in Table 1.

2.2. Soil sampling

Soils samples were taken from all 12 sampling sites in spring 2010. Three soil profile pits were dug at each site, yielding a total of 36 pits. For the first soil pit, soil samples were taken from each distinct soil horizon down to the C-horizon, which resulted in 3–6 soil samples per pit. The C-horizon was always within 1.2 m of the surface. For the second and third pit, only the most prominent soil horizons were sampled which resulted in 1–3 soil samples per pit. Soil samples taken from the upper 10 cm were considered as topsoils whereas samples from deeper horizons were considered as subsoils. For the 36 pits together, this sampling design yielded a total of 126 samples: 39 top- and 87 subsoil samples. Because of the heterogeneity of the field sites and the differences in sampling depth of the various samples taken from the different pits, all soil samples are regarded as individual samples in the data analyses. Bulk soil samples were taken for the analysis of soil texture, aggregates, and soil chemical properties. The field-moist samples were gently broken up by hand so that the larger clods broke along natural fissures, and roots and any rock fragments were removed by hand. Undisturbed

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