



Invited review

Pedogenic carbonates: Forms and formation processes



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ABSTRACT

Soils comprise the largest terrestrial carbon (C) pool, containing both organic and inorganic C. Soil inorganic carbon (SIC) was frequently disregarded because (1) it is partly heritage from soil parent material, (2) it undergoes slow formation processes and (3) has very slow exchange with atmospheric CO₂. The global importance of SIC, however, is reflected by the fact that SIC links the long-term geological C cycle with the fast biotic C cycle, and this linkage is ongoing in soils. Furthermore, the importance of SIC is at least as high as that of soil organic carbon (SOC) especially in semiarid and arid climates, where SIC comprises the largest C pool. Considering the origin, formation processes and morphology, carbonates in soils are categorized into three groups: geogenic carbonates (GC), biogenic carbonates (BC) and pedogenic carbonates (PC). In this review we summarize the available data and theories on forms and formation processes of PC and relate them to environmental factors. After describing the general formation principles of PC, we present the specific forms and formation processes for PC features and the possibilities to use them to reconstruct soil-forming factors and processes. The following PC are described in detail: earthworm biospheroliths, rhizoliths and calcified roots, hypocoatings, nodules, clast coatings, calcretes and laminar caps.

The second part of the review focuses on the isotopic composition of PC: δ¹³C, Δ¹⁴C and δ¹⁸O, as well as clumped ¹³C and ¹⁸O isotopes known as Δ₄₇. The isotopic signature of PC enables reconstructing the formation environment: the dominating vegetation (δ¹³C), temperature (δ¹⁸O and Δ₄₇), and the age of PC formation (Δ¹⁴C). The uncertainties in reconstructural and dating studies due to PC recrystallization after formation are discussed and simple approaches to consider recrystallization are summarized.

Finally, we suggest the most important future research directions on PC, including the anthropogenic effects of fertilization and soil management. In conclusion, PC are an important part of SIC that reflect the time, periods and formation processes in soils. A mechanistic understanding of PC formation is a prerequisite to predict terrestrial C stocks and changes in the global C cycle, and to link the long-term geological with short-term biological C cycles.

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1. Introduction: inorganic carbon in soil and pedogenic carbonates

1.1. Relevance of soil inorganic carbon

Soils with 2,470 Pg C (Eswaran et al., 2000) are the largest terrestrial C pool and are the third greatest C reservoir in the world after oceans with 38,725 Pg (IPCC, 1990) and fossil fuels with 4000 Pg (Siegenthaler and Sarmiento, 1993) containing organic and inorganic C (Eswaran et al., 2000). Plant litter, rhizodeposits and microbial biomass are the main sources of the soil organic carbon (SOC) pool. The SOC pool comprises 697 Pg C in 0–30 cm and 1500 Pg C in 0–100 cm depths (IPCC, 2007). Intensive exchange of organic C with the atmosphere, especially connected with anthropogenic activities, led to a very broad range of studies related to the organic C cycle in soil and these have been summarized in many reviews (e.g. IPCC, 2007; Kuzyakov, 2006a).

In contrast to organic C, the exchange of soil inorganic carbon (SIC), i.e. various soil carbonate minerals (mostly calcite), with the atmosphere and the involvement of SIC in biotic C cycles is much slower (mean residence time of 78,000 years (Schlesinger, 1985)). Additionally, the distribution depth of SIC is opposite to that of SOC: most stocks are located deeper than one meter (Díaz-Hernández et al., 2003; Wang et al., 2010). These two reasons explain why much fewer studies focused on SIC than on SOC (Drees et al., 2001; Rawlins et al., 2011). Nonetheless, large stocks of SIC – 160 Pg C in 0–30 cm (Nieder and Benbi, 2008), 695–748 Pg C in 0–100 cm depth (Batjes, 1996) and 950 Pg C up to 2 m (Lal, 2012) – reflect its importance especially over the long term. The SIC content in first 2 m of soil in semi-arid regions could be 10 or even up to 17 times higher than SOC (Díaz-Hernández et al., 2003; Emmerich, 2003; Shi et al., 2012). Furthermore, a much longer mean residence time of SIC – millennia (Schlesinger, 1985) – shows its greater role in the global C cycle compared with SOC (few hours to centuries) (Hsieh, 1993; Qualls and Bridgham, 2005). SIC also links SOC with short residence times to the long-term geological C cycle (Liu et al., 2010). Soils of arid and semi-arid regions with usually alkaline pH (>8.5) and richness in Ca and/or Mg (>0.1%) may enhance the SIC content following organic fertilization and increase the respired CO_2 (Bugchio et al., 2016; Wang et al., 2015).

Changes in environmental properties such as soil acidification because of N fertilization, N fixation by legumes or intensification of rewetting cycles due to irrigation could release great amounts of SIC and increase CO_2 emissions (Eswaran et al., 2000; Shi et al., 2012). Such effects, though driven by natural processes, are well known in our planet's history, e.g. between the Pleistocene and Holocene, when around 400–500 Pg C were released from SIC and strongly intensified global warming over a short period (Adams and Post, 1999). The formation and accumulation of carbonate minerals in soils, in contrast, can directly mitigate the increase of atmospheric CO_2 (Landi et al., 2003; Xie et al., 2008) if calcium (Ca^{2+}) ions have been released to the soil via sources

other than carbonate-containing minerals, for instance from weathering of igneous rocks, decomposition of organic matter or dissolved Ca^{2+} in rainwater (Boettinger and Southard, 1991; Emmerich, 2003; Monger et al., 2015). This calls for investigating SIC stocks, forms and their dynamics to understand the role of SIC in the C cycle at regional and global scales, the fast and slow processes of C cycling, as well as the link between biotic and abiotic parts of the C cycle.

1.2. Soil inorganic carbon: worldwide distribution

Large SIC stocks are mostly found in regions with low water availability (i.e. arid, semi-arid and sub-humid regions) (Fig. 1) (Eswaran et al., 2000). Low precipitation and high potential evapotranspiration strongly limit the dissolution and leaching of carbonates from soil (Eswaran et al., 2000; Royer, 1999). Accordingly, the highest SIC content – around 320 to 1280 Mg C ha^{-1} – is accumulated in soils of arid regions with mean annual precipitation (MAP) below 250 mm such as middle east, African Sahara and west USA (Fig. 1). As MAP increases, the SIC content decreases and <40 Mg C ha^{-1} may accumulate at MAP exceeding 1000 mm for example in Amazonian forests and monsoonal forests in south-east Asia. However, partial eluviation and redistribution of carbonates may concentrate SIC deeper in the soil profile (Díaz-Hernández et al., 2003; Wang et al., 2010).

1.3. Soil inorganic carbon pools, classification and definitions

Based on origin, formation processes and morphology, the SIC can be subdivided into three large groups:

1. Geogenic carbonates (GC)¹: carbonates which have remained or are inherited from soil parent materials such as limestone particles or allocated onto the soil from other locations by calcareous dust or landslides etc.
2. Biogenic carbonates (BC)¹: carbonates formed within terrestrial or aquatic animals and plants as part of their skeleton, for example shells, bones and calcified seeds, or released from or within certain organs such as the esophageal glands of earthworms.
3. Pedogenic carbonates (PC): carbonates formed and redistributed in soils via dissolution of the SIC pool (i.e. geogenic, biogenic or previously formed pedogenic carbonates) and re-precipitation of dissolved ions in various morphologies such as carbonate nodules.

This review focuses solely on the origin, morphology and processes of PC formation. The GC and BC are mentioned only if relevant for PC formation.

¹ Here we do not review the forms and formation of geogenic and biogenic carbonates in soil.

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