



## **Evolution of soil organic matter energetic and chemical composition during long-term bare fallow: implications for soil carbon vulnerability to global change**

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Determining the relative stability of soil organic carbon (SOC) is a critical step to better understanding its vulnerability to global change. The absence of convincing physical or chemical procedures to define, characterize or isolate relatively labile versus stable pools of SOC makes it difficult to study. Long-term bare fallow (LTBF) experiments, in which C inputs have been stopped for several decades, provide a unique opportunity to study stable SOC without the inherent artefacts induced by extraction procedures, the hypothesis being that SOC is gradually enriched in stable C with time as labile components decompose. We determined the evolution of energetic and chemical characteristics of bulk SOC in five LTBF experiments across Europe: Askov (DK), Grignon (FR), Rothamsted (UK), Ultuna (SW) and Versailles (FR), using simultaneous thermal analysis (i.e. thermogravimetry (TG), differential scanning calorimetry (DSC) and evolved CO<sub>2</sub> gas analysis (CO<sub>2</sub>-EGA)) and diffuse reflectance Fourier transformed mid-infrared spectroscopy (DRIFT-MIRS).

Results of TG analyses showed that the temperature needed to combust the first half of the SOC (i.e. TG-T50) increased with bare fallow duration at all sites. Conversely, the energy density (in mJ mg<sup>-1</sup> C) decreased with bare fallow duration. Combined together, these results provide a means to contrast the stable, mineral-associated SOC pool from any potential pyrogenic C, which would have much greater energy density. DRIFT-MIRS results showed that the “carboxylation index” (the ratio of C=O bonds peak area over (C=C + C=O) bonds peak areas) decreased with bare fallow duration, that aromaticity (C=C bond peak area over C content) increased with bare-fallow duration, and that the “reticulation index” (CH<sub>3</sub> peak area over CH<sub>2</sub> peak area) decreased with increasing bare fallow duration at Rothamsted, Versailles and Ultuna. These trends were less clear or not observed at Grignon due to the presence of carbonates or at Askov due to greater variability, thus precluding unequivocal conclusions.

Our results showed that in spite of the heterogeneity of the soils at the LTBF sites, generalized energetic and chemical pathways exist for SOC stabilization. The DRIFT-MIRS indices demonstrate that SOC stabilization is accompanied by a consistent evolution of its bulk chemical composition across most sites, over decades (ca. 50 years). The general decarboxylation of SOC observed in bare fallow challenges the current view of SOM decomposition, raising questions about the oxidative state of the stable C pool. The increased burning temperature and lower energy density of stable SOC suggest that decomposition of the stable C pool may be more temperature sensitive and thus vulnerable to increased temperature. Conversely, decreasing energy density suggests that priming might be the only means available for the microbial community to decompose this pool of SOC, and that SOC stability may be a function of low potential energy gain from decomposition of this material. Finally, this study illustrates the enormous value of such long-term field experiments, and a potential multi-faceted approach to quantify SOC stability as an ecosystem property.