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## Redox-driven changes in the distribution of Fe minerals between aggregate-size classes in illuvial and eluvial horizons of a hydromorphic soil

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Paddy soils experience long-term redox alternations affecting the interactions between the biogeochemical cycling of iron (Fe) and carbon (C). Although the higher soil organic matter (SOM) accumulation rates in paddy topsoils with respect to non-paddy soils is generally assumed to be due to limited mineralization under anoxic soil conditions resulting from frequent field flooding, there is growing evidence questioning this assumption. Moreover, differences in particle aggregation and SOM turnover are likely to both affect and be affected by the trajectory of Fe mineral evolution/crystallinity with redox fluctuations. We hypothesized that redox cycling in paddy soils will affect the particle aggregation, the distribution and mineralogy Fe (hydr)oxides between aggregate size fractions, and consequently the mechanisms of SOM stabilization. In particular, we expect finer aggregate and particle size classes to have a higher proportion of short-range ordered (SRO) Fe oxides with respect to larger aggregates under paddy management, compared to non-paddy management, and that paddy management can result in lower amounts of Fe(hydr)oxides in the topsoil with respect to non-paddy soils.

We tested these hypotheses by evaluating mineralogical changes, and the distribution of Fe species and organic C between different aggregate and particle-size fractions in topsoil (eluvial) and subsoil (illuvial) horizons of soils under long-term paddy (P) horizons (Arp1, Arp2, Arpd, Brd1, Brd2) and non-paddy (NP) horizons (Ap1, Ap2, Bgw) in NW Italy. Soil aggregates (microaggregates: <200  $\mu\text{m}$ , free silt: (53-2  $\mu\text{m}$ ), free clay: <2  $\mu\text{m}$ , and, after sonication, fine sand, silt and clay within microaggregates) have been obtained from bulk soils using an aggregate and particle size physical fractionation method. After fractionation, Fe phases were evaluated by selective extraction procedures, X-ray diffraction (XRD) and Fe K-edge extended X-ray fine structure (Fe EXAFS) spectroscopy (Elettra XAFS beamline).

Our results indicate: (1) a depletion in the contents of ferrihydrite in the P topsoil horizons with respect to NP, though redox cycling favoured an increase in ferrihydrite in the P subsoil, possibly due to Fe(II) translocation from topsoil to subsoil, with consequent ferrihydrite precipitation and

aggregates formation; (2) more crystalline Fe mineral phases were associated with intra-aggregate clay fraction in the P topsoil. In the clay fraction in the Brd2 subsoil horizon magnetite was observed. In the NP soil the illuvial horizons were not characterized by a significant increase in ferrihydrite. Our hypothesis that finer aggregate and particle size classes have a higher proportion of SRO Fe oxides with respect to larger aggregates under P management, with respect to NP management, was confirmed; (3) more organic C was associated with the fine fraction in P with respect to NP suggesting that redox cycling enhances the chemical stabilization of mineral-associated SOM.

These findings focused on localized Fe dynamics and biogeochemical coupling with SOM, suggesting that redox-driven changes in aggregate-size classes distribution were also linked to the differences in organic C and Fe stocks in these two agro-ecosystems.