

We greatly appreciate the amount of time and effort put into this review as evidenced by the extremely constructive comments provided! We will address the reviewer's concerns by reorganizing, structuring, re-writing, and summarizing the text in the manuscript as described below.

1. Lines 33-52: This list of evidence reflects the structure of the paper overall – many sections in the paper are stand alone “chunks” of ideas that do not cohesively tie together. As written, the sections appear as a list of ideas instead of a defined structure with a beginning, middle, and end.

**Response:** We agree with this helpful comment. This list is transformed into a paragraph with a beginning, middle and an end. It reads now as follows:

**Line 30:** “Since C and N cycles are interconnected in soils (Feng et al., 2019; Gärdenäs et al., 2011), they should be regulated by the same factors, including mineralogy type (Wade et al., 2018). Increasing evidence shows that Fe specifically represents a major control over N biological transformations, including mineralization (Wade et al., 2018), nitrification (Huang et al., 2016a) (Han et al., 2018) denitrification (Zhu et al., 2013) (Wang et al., 2016), as well as their abiotic analogous reactions, such as chemo-denitrification (Burger and Venterea, 2011) and Fe-mediated hydroxylamine oxidation to nitrous oxide (N<sub>2</sub>O). These reactions and others (Fig.1) are likely to operate ubiquitously in soils, due to the close proximity between Fe minerals and SOM since most of the latter is contained in association with the former (Lalonde et al., 2012; Wagai and Mayer, 2007)”.

2. Section 4: It is not clear how the structural role is distinct from the sorbent role. The mechanisms presented in Figure 2 and many described in this section are referring to adsorption/desorption processes.

**Response:** The purpose of this section is to highlight the role of Fe in the formation and stability of micro-aggregates and the impact this has on N bioavailability. This section refers to the fact that Fe-mediated aggregate stability increases N stability inside microaggregates by limiting its physical accessibility to microbes. The intent was to highlight a physical rather than a chemical phenomenon, therefore, we eliminated all the text referring to sorption-adsorption mechanisms. Thank you so much for bringing this to our attention, that was a great review comment!

\*\*This section now reads as:

Fe oxides are one of the most important constituents of soil microaggregates (Peng et al., 2015), serving as nuclei for their formation and mediating their stability (Barral et al.,

1998; Pronk et al., 2012; Wei et al., 2016), acting as a cementing agent (Colombo and Torrent, 1991; Krause et al., 2020) and binding OM (Giovannini and Sequi, 1976; Totsche et al., 2017). Recent studies demonstrated that colloidal-sized Fe promotes the formation of smaller-sized microaggregates (<20  $\mu\text{m}$ ) and Fe-rich biosolids induce rapid formation of microaggregates and significantly increase soil organic carbon (SOC). (Krause et al., 2020) (Silva et al., 2015). N is also an important component of microaggregate-SOM (Golchin et al., 1994) (Aufdenkampe et al., 2001) (Sollins et al., 2006). Using density fractionation, Wagai et al., (2020) observed joint accumulation of OM with low C:N ratio and pedogenic Fe and Al oxides in the meso-density fractions (1.8–2.4  $\text{g cm}^{-3}$ ) of five soil orders collected from different climate zones. Moreover, Rodionov et al., (2001) observed high concentrations of amino sugars in microaggregates (Rodionov et al., 2001). These observations have implications for N bioavailability, given the fact that Fe mediated micro-aggregation may slow down or suppress N mineralization (Mendes et al., 1999). Indeed, N compounds located inside microaggregates have lower availability to microbes than those located on more accessible surfaces. Microaggregate-N is also relatively more persistent than macroaggregate-N because microaggregates' turnover is relatively slow, which provides longer-term stabilization of OM (Cambardella and Elliott, 1993; Six et al., 2002). Similarly, Bugeja and Castellano (2018) observed positive correlation between ammonium oxalate-extractable Fe (AmOx-F), C and N in microaggregate, indicating that Fe and microaggregate stabilization are interconnected (Bugeja and Castellano, 2018). Partial or complete removal of mineral-forming components, for example due to Fe reduction, can initiate aggregate turnover and destabilization (Michalet, 1993; Cornell and Schwertmann, 2003) which eventually exposes OM to microbial degradation (Lützw et al., 2006) and organic carbon (OC) and ON loss from SOM (Cambardella and Elliott, 1993). We also hypothesize that there is another pathway by which Fe-promoted aggregation may decrease N mineralization. Aggregates of different sizes influence microbial community composition differently and therefore the activities of N mineralization enzymes (Muruganandam et al., 2009). Therefore, it will be useful to examine the distribution and the activities of these enzymes among soil aggregate size classes along a gradient of increased Fe mineral content in soils.

We also updated Figure 2:

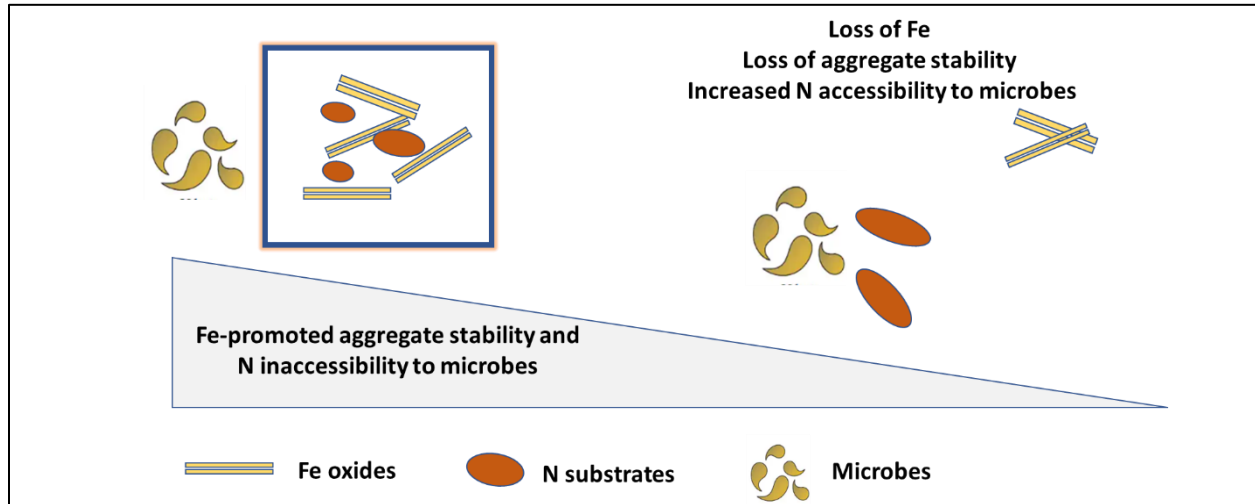


Figure: Schematic representation of the effects of Fe-promoted aggregate formation and stability on N accessibility to microbial degradation.

3. Sections 7 and 8, in particular, lack an overall structure. A potential solution to the organizational issues with the writing would be to separate the properties and processes into distinct spatial scales: 1) The molecular scale at which sorption/desorption, catalysis, electron transfer occurs, 2) the micro-scale, at which iron mediates soil aggregation, and 3) the meso/ecosystem-scale, at which iron may influence the priming of soil nitrogen in the rhizosphere or the response of SON cycling to global change.

**Response:** We thank the reviewer for this helpful and well thought of comment. We love the idea of separating processes/mechanisms into scales, but we didn't feel that it would follow the flow of the narrative and may make the review too long. To make the structure clearer to the reader, we decided to introduce section 7 and 8; section 7 which details the role of Fe in the three complex phenomena that affects N bioavailability in soils; priming, birch effect and freeze-thaw cycle and section 8 with the focus on how is Fe-N bioavailability influenced by global change.

\*\*The paragraph in the introduction now reads as:

**line 70:** 'While these roles of Fe in controlling C cycling have been studied extensively, their effects on N bioavailability are not well explored. This review seeks to underpin these suggested relationships and provide mechanistic descriptions of how Fe controls N bioavailability in soils. Moreover, we detail how Fe participates in three complex

phenomena that influence N bioavailability; priming, birch effect, and freeze-thaw cycle. We also highlight how Fe-N interactions are affected by global change. This information are needed to construct reliable models with improved predictive power of N cycling in terrestrial ecosystems (Wade et al., 2018), and will offer new possibilities for land management”.

\*\*We also included an introductory paragraph at the beginning of section 7 where we talk about how the three phenomena influence N bioavailability and why the role of Fe should be examined.

\*\*To better structure section 8, we synthesized its information and introduced subheads to highlight the driver of change and potential impact on Fe-N bioavailability. We also included an introductory paragraph to this section.

This section subheads are:

## **8 Impact of global change on Fe-N bioavailability interactions**

8.1 Impact of variability in precipitation

8.2 Impact of variability in temperature

8.2 Impact of elevated CO<sub>2</sub>

8.3 Impact of land use change

We hope these changes are satisfactory and address the reviewer concerns.