

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.

******We profoundly thank the reviewer for providing a list of relevant and excellent reviews on N bioavailability. These reviews have been read and incorporated into the introduction of this manuscript to support the need to study Fe minerals impact on N bioavailability.

1. L33-51. This section might read better as prose rather than a list that doesn't seem to be ordered in a particular way.

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:

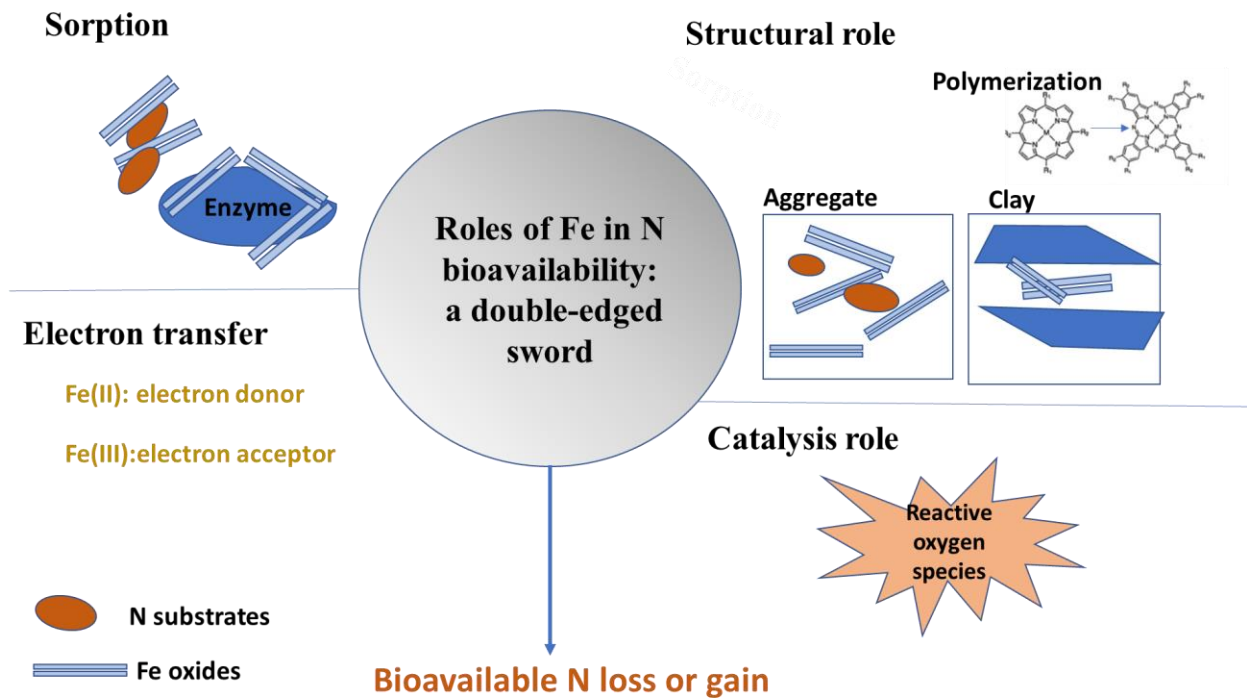
Line 30: "Since C and N cycles are interconnected in soils (Feng et al., 2019; Gärdenäs et al., 2011), they should be regulated by the same factors, including mineralogy type (Wade et al., 2018). Increasing evidence shows that Fe specifically represents a major control over N biological transformations, 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 (N₂O). 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. L53-70. This section provides a nice structure for the paper.

Response: Thank you for this helpful comment. We like this structure too !

3. Fig. 1. Is this an original figure or taken from another source? If the latter, the source needs to be provided. If the former, I would suggest using color to make it more interesting/readable and also link it to the overarching structure of the paper. That is, if you're going to follow the structure outlined in L53-70, then indicate how these four "roles" tie into the figure. Or develop a different overarching figure that gives a conceptual overview of the paper.

Response: Thank you very much for bringing this to our attention. This figure was adapted from (Zhu-Barker et al. 2016). The original figure from Zhu-Barker et al. did not have all the processes that I added to it. We felt adding the 4 roles of Fe to this figure would make it cluttered and the purpose was just to show that Fe takes part in almost every N process. So, we decided to develop another figure to give a conceptual overview of the paper (as per the recommendation). See attached.



4. Sec. 2. This section interrupts the flow from the overarching structure of the paper laid out in L53-70 and Sec. 3 (i.e., "sorbent role"). Perhaps this material could be shortened and included in the intro section before laying out the paper objectives/structure

We agree with this helpful comment. Material has been shortened and added to the introduction of this manuscript. This section reads now as:

Starting at the 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 (N₂O). 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).

The characteristic properties of individual Fe minerals and N compounds and how these properties are influenced by the soil environment likely drive the aforementioned reactions as well. First, Fe exists in a variety of polymorphs (Navrotsky et al., 2008) and is a redox-sensitive element that cycles between Fe(II) and Fe(III) states as controlled by soil Eh and pH. While Fe(III) promotes N stabilization within mineral associations, Fe(III) mobilization when it is reduced to Fe(II) can release N into solution. Fe reactivity is also driven by the amount and sign of surface charge, surface topography, particle size, crystallinity (Li et al., 2015a) (Petridis et al., 2014) and the presence and the type of organic matter (OM) coverage (Gao et al., 2018; Kleber et al., 2007; Poggenburg et al., 2018) (Boland et al., 2014; Henneberry et al., 2016) (Kaiser and Zech, 2000a) (Daugherty et al., 2017). Second, soil N exists predominantly in organic forms (ON); mostly as protein and peptides, and to a lesser extent as amino-sugars and nucleic acids (Kögel-Knabner, 2006) (Knicker, 2011; Schulten and Schnitzer, 1997). Proteins are intrinsically reactive towards soil minerals, due to a number of properties, including hydrophobicity, surface charge distribution, surface area, number and type of functional groups, conformation, and size (Lützow et al., 2006). N from these compounds is generally not directly bioavailable due to molecular size constraints on microbial cell uptake (Schimel and Bennett, 2004). Depolymerization reactions, carried out by the activity of extracellular enzymes, such as peptidases, transform these polymers into soluble, low molecular weight (MW) organic monomers (e.g., short oligopeptides, amino acids (AAs)). Recent research shows that the size of AAs available for mineralization is controlled by peptidase activity, but more so by protein availability, both of which are affected by the interactions with Fe minerals. Therefore, Fe may drive gross AA production in soils (Noll et al., 2019).

5. L127. This question doesn't lead well into the overarching theme of this section and detracts from the main thread. Start with an introductory paragraph that

introduces the topic and provides a roadmap for the rest of the section. I assume that the “sorberent role” isn’t just about enzymes, right?

Response: We agree with this helpful comment. An introductory paragraph has been added to all the sections.

For section 1 for example, this paragraph reads as: “This section explores the role of Fe in regulating N bioavailability through sorption processes, which alter both the availability of N substrates and enzyme activity. This section also highlights the multiple mechanisms of destabilization of sorbed N; such as Fe reduction, desorption by local disequilibrium in soil chemistry and the displacement of N by competitive sorption”.

6. Sec. 4. As noted above, starting the section with a question is not the best way to introduce the topic and provide an overview for the section. In the first paragraph, lay out what the questions or topics are and then follow up with a succinct discussion of each. Ditto this comment for all sections.

Response: We agree with this helpful comment. An introductory paragraph has been added to all the sections. For section 4, this paragraph reads as:

“The impact of structural Fe on N bioavailability in soils is a complex phenomenon that can be influenced by various factors. This section provides a detailed examination of this subject, focusing on how the structural Fe in clay and aggregates influence N turnover, as well as Fe- induced organic nitrogen (ON) polymerization”

7. Fig. 2. I would encourage you to think about how this figure could be a bit more nuanced rather than just having “clouds” for enzymes, N substrates, etc.

Response: This figure has been eliminated from the manuscript to address the first reviewer comment. An updated figure has been incorporated into the manuscript (see attached).

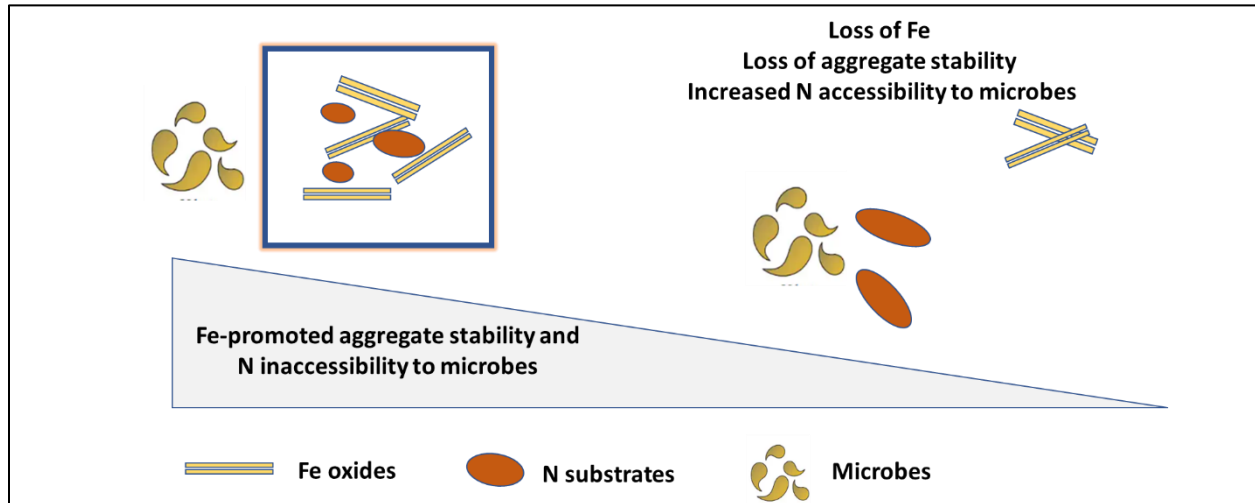


Figure: Schematic representation of the effects of Fe-promoted aggregate formation and stability on N accessibility to microbial degradation.

8. Sec. 5. You skip the “electron transfer role” and go straight to “catalytic role”. If there isn’t a separate section for “electron transfer role” then remove from overview paragraph as a separate, defined “role”.

Response: The electron Transfer role of Fe is presented in lines 325-350

9. Sec. 7 and 8. As noted by the other reviewer, these sections lack structure and were not adequately introduced earlier in the manuscript.

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

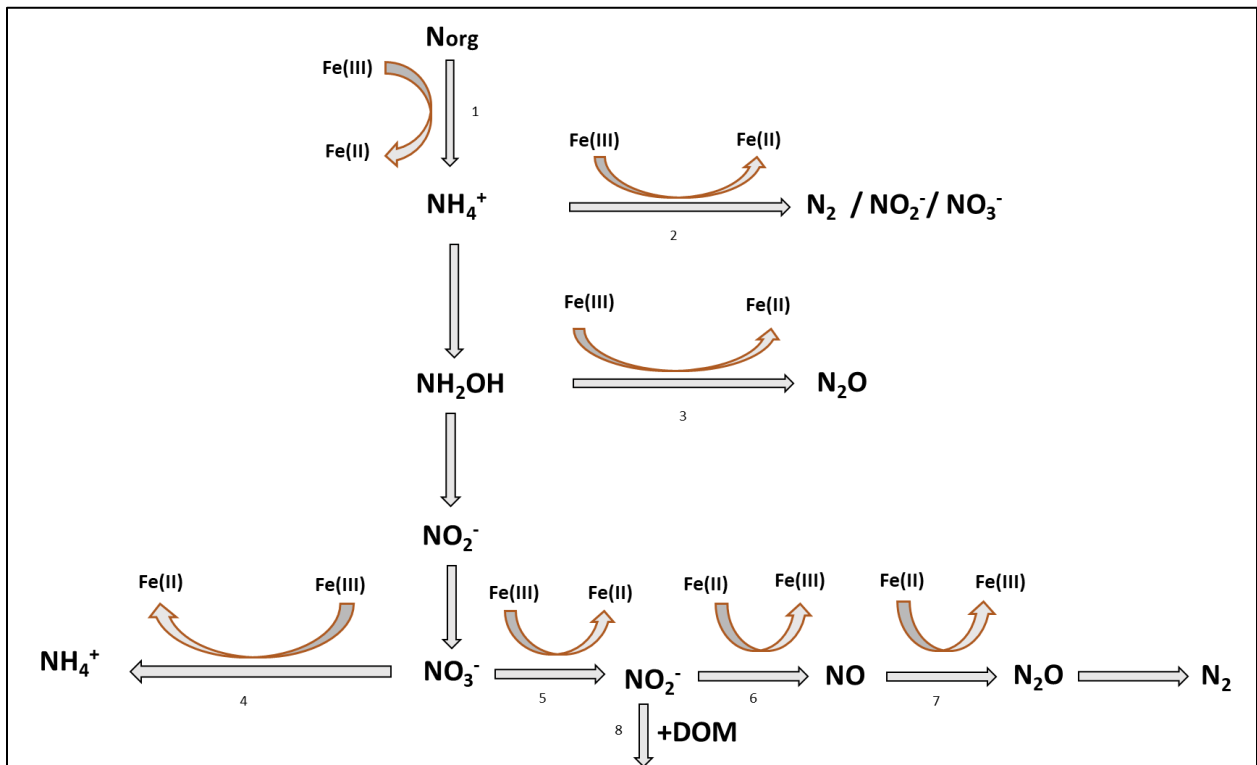
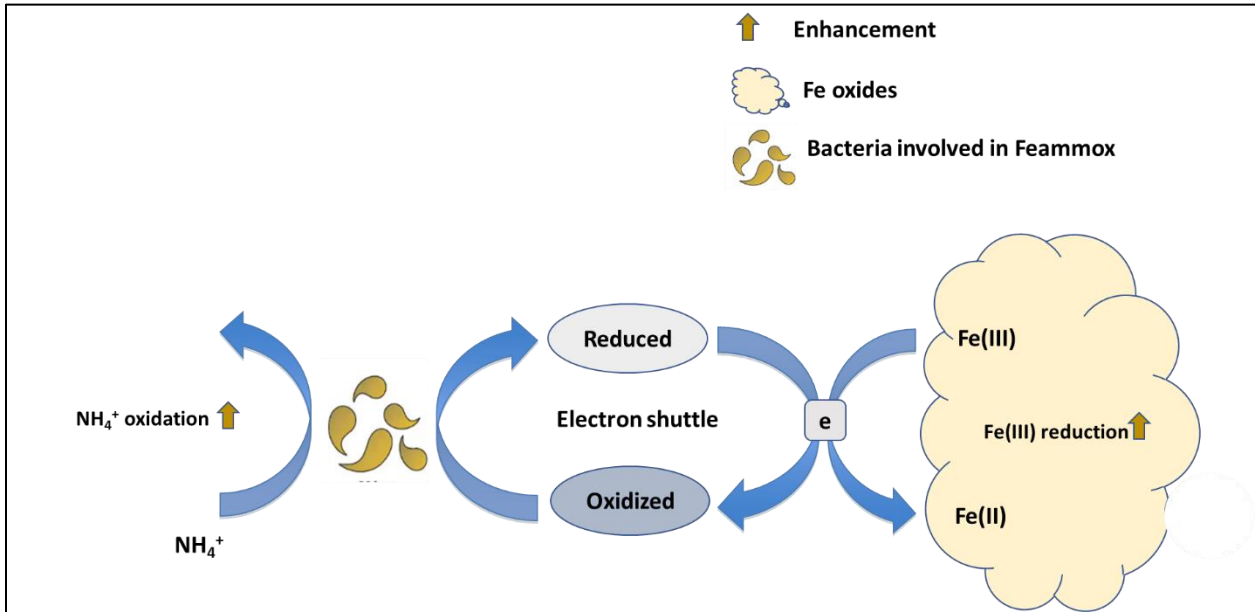
8.1 Impact of variability in precipitation

8.2 Impact of variability in temperature

8.2 Impact of elevated CO₂

8.3 Impact of land use change

All the figures of the manuscript have been updated with colors and revised. Here are examples:



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

