DOI: 10.1111/gcb.14403

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RESPONSE TO THE EDITOR

Contribution of soil inorganic carbon to atmospheric CO₂: More important than previously thought

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Funding Information

Deutsche Forschungsgemeinschaft, Grant/Award Number: KU 1184/34-1

Keywords: acidification, CO2 efflux, global warming, liming, nitrogen fertilization, soil inorganic carbon

 $\rm CO_2$ production from soil inorganic carbon (SIC) by neutralization of nitrogen (N) fertilization-induced acidity is globally relevant. Here we analyzed factors that may affect $\rm CO_2$ production from SIC after N fertilization: (1) buffering capacity of soil organic matter (SOM) and of clays, (2) increasing crop growth and C input belowground by N fertilization, (3) acidity localization at the fertilization point, (4) SIC localization in the sub-soil, (5) application of CaO and basic slag instead of lime, (6) inability of farmers in low income countries to apply lime. We conclude that our previous estimation of $\rm CO_2$ fluxes from carbonates by N fertilization (7.5 \times 10^{12} g C year $^{-1}$) and from liming of acidic soils (273 \times 10^{12} g C year $^{-1}$) is possibly an underestimation and consequently, the contribution of SIC to atmospheric $\rm CO_2$ is more important than previously thought.

 CO_2 production from soil inorganic carbon (SIC) by neutralization of nitrogen (N) fertilization-induced acidity is globally relevant, but up to now was not considered as a CO_2 source. We analyzed in detail the seven factors mentioned by Datta and Mandal (2018) to hone our estimation of CO_2 efflux from SIC (Zamanian, Zarebanadkouki, & Kuzyakov, 2018).

SOM and clays buffer the acidity only in soils with pH below 6.5—the soils without carbonates. The increased belowground C input by N fertilization can only be of very minor importance because the portion of belowground allocated C decreases with N availability (Figure 1) and the soil organic carbon (SOC) in most soils is under steady state. Consequently, the increase of root biomass by N fertilization is less than that of the shoots, because of decreasing root/shoot ratio. So, despite root C contributes 2.5–3.0 times more to the SOM formation (Rasse, Rumpel, & Dignac, 2005), the relative increase of root biomass is of minor importance. Further, the root/shoot ratio will decrease in future because the breeding is mainly aimed on the grain yield.

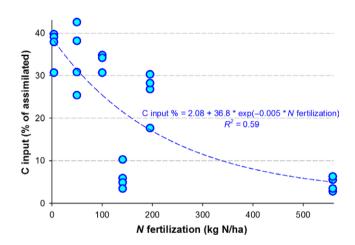


FIGURE 1 Belowground C allocation by Lolium perenne depending on N fertilization. The contribution of CO₂ assimilated by plants to SOM formation mentioned by Datta and Mandal (2018)about 1/3 of plant-assimilated C—is strongly overestimated. We agree that about 20%-33% of assimilated C will be allocated belowground by grasses or agricultural cereals (Pausch & Kuzyakov, 2018), but: (1) the C input into the soil is much larger than its contribution to SOM formation, because most of the root-derived C will be decomposed to CO₂, and (2) the portion of assimilated C strongly decreases with N fertilization. Therefore, only a small part of the belowground allocated C contributes to SOM formation: According to the review of (Kuzyakov & Domanski, 2000), only 3.5% (wheat), 2.3% (barley), and 3.7%-4.9% (various grasses) of the plantassimilated C is incorporated into SOM. From this C incorporated into SOM, only a delta—the increase (Δ) of total C input by plants into the soil by N fertilization—should be considered. This Δ is so marginal that its potential importance disappears against the backdrop of other factors. Modified from figure 8 of Pausch and Kuzyakov (2018); See there for the original references of the studies [Colour figure can be viewed at wileyonlinelibrary.com]

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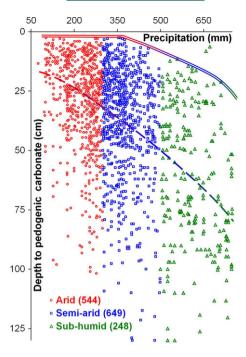


FIGURE 2 Depth to the upper level of the calcic horizon (horizon containing pedogenic carbonates) in 1,436 analyzed soil profiles under three climatic conditions (arid, semi-arid, sub-humid) depending on the mean annual precipitation (modified after Zamanian, Pustovoytov, & Kuzyakov, 2016). Note that pedogenic carbonates are formed via dissolution of geogenic carbonates, that is carbonates in parent material, and further reprecipitation of dissolved ions (i.e., Ca²⁺ and HCO₃⁻). The dissolved ions might also be translocated down the soil profile, whereby reprecipitation would take place at horizons deeper than the original calcareous parent material. About 39% and 46% of the analyzed profiles contain pedogenic carbonates starting at depths of ca. 25 and 30 cm, respectively. The double line at the top shows the minimal depth and the dashed line in the middle show the average depth of the upper level of calcic horizon; both decrease with precipitation. Note that only the upper depth of the calcic horizon is presented; all these soils contain CaCO₃ at least to the parent material. The numbers in the brackets close to the legend show the number of points for each climate zone [Colour figure can be viewed at wile yonlinelibrary.com]

The initial acidity induced by fertilization is limited to the point at which fertilizers are applied, but the produced H^+ ions migrate easily to the $CaCO_3$ depth and are neutralized with CO_2 production. Furthermore, erosion (common in croplands even in semi-arid climates) places the sub-soil in contact to the atmosphere and thus to the applied fertilizers. When SIC is present, its amount is endlessly large compared to the applied N fertilizers (Figure 2).

CaO and Ca $^{2+}$ -containing slag are produced by heating CaCO $_3$ and metal ores, respectively, at temperatures exceeding 800°C. The production of CaO and basic slag leads to CO $_2$ emission from CaCO $_3$, but in a sector other than agriculture. The area amended with CaO and basic slag is minimal compared to the liming area. N fertilization in low-income countries is very low (see the N

fertilization map: Figure 1 in Zamanian et al., 2018) and, accordingly, less acidification is expected. The annual release of 3.2-7.9 10¹² g C from SIC as estimated by Perrin, Probst and Probst (2008) is based on a field trial and direct measurement of NO₃and HCO₃⁻ concentrations in surface water. They clearly showed the correspondence of alkalinity neutralization in surface water (not soil) with increasing nitrate concentrations. So far, our estimation based on the CaCO₃ distribution in soils and N fertilization is the first approach for a global estimation of CO₂ from SIC. Using more accurate and detailed maps accompanying local measurements will no doubt improve the estimation. Nonetheless, higher CO₂ fluxes are expected because over-fertilization always takes place and N use efficiencies at local scales are much lower than the assumed averages. The wetting/drying of paddy soils, fertilizers other than urea (e.g., (NH₄)₂SO₄, Ca(H₂PO₄)₂), and mineralization of animal dung in pastures should also be included. Furthermore, not only N fertilization, but also global N pollution leads to acidification (Averill, Dietze, & Bhatnagar, 2018) and unaccounted CO₂ efflux from CaCO₃. We conclude that, considering the factors mentioned by Datta and Mandal (2018), our estimation (Zamanian et al., 2018) of CO₂ fluxes from carbonates by N fertilization $(7.5 \times 10^{12} \text{ g C year}^{-1})$ and from liming of acidic soils $(273 \times 10^{12} \text{ g C year}^{-1})$ as accurate as it can be done to date is possibly an underestimation.

ACKNOWLEDGEMENTS

We acknowledge German Research Foundation (DFG) for their support (KU 1184/34-1).

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