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Soil aggregates as biogeochemical reactors: Not a way forward in the research on soil-atmosphere exchange of greenhouse gases

Article type : Letters to Editor

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Over the last two decades, the fact that soils are significant sources of greenhouse gases (GHG), e.g., carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and water vapor, has received considerable attention from the scientific community. Many laboratory and field experiments have been carried out to investigate the release of GHG by soils, and a wide range of computer modeling approaches have been explored to encapsulate what is known about the process, as well as to improve its prediction at various spatial and temporal scales. In this context, in an article published recently in Global Change Biology, Wang et al. (2018) note that knowledge about GHG dynamics at the relatively small scale of soil "aggregates" is still scarce, and advocate that significant new insight about GHG release could be obtained from

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1111/gcb.14640

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considering "aggregate reactors" in a systematic and dynamic way. Wang et al. (2018) contend that by "proposing this aggregate reactor concept in a dynamic framework, ecological theory can be applied to studies of GHG exchange by examining both the reactivity of an aggregate reactor (physiology) and the compositional dynamics of differing aggregate reactors (community ecology)." These authors "recommend integration of soil science, ecology, and climate science communities to advance the aggregate reactor concept and to develop a predictive framework based on aggregate reactors in the context of global change."

Superficially, this recommendation, and the concept of "aggregate reactor" may sound very appealing, all the more so that a huge literature exists on the topic of soil aggregates, providing a wealth of information from which one could draw to develop the predictive framework to which Wang et al. (2019) refer. Unfortunately, due to the opacity of soils and the virtual absence of direct observation methods, soil research relied heavily in the past on destructive experimental tools, among which are dry and wet soil sieving techniques used to procure aggregates. While importance and usefulness of these experimental tools can not be overstated, their limitations must not be forgotten either. Just like any other ecosystem, soil is not a simple sum of its individual components. That is, the outcomes of the processes taking place within individual aggregates obtained from sieving intact soil will not add to the outcome from the intact soil itself. However, while most ecologists would not see a value in building an ecosystem model from a disjoint collection of individual sites without considering links and connectivity among them, such links and connections are easily forgotten when we get to soils.

It should be noted that the general idea proposed by Wang et al. (2019) is not necessarily new. The ease with which intact soil can be transformed into a pile of aggregates misled many into thinking that the pile can just as easily be assembled back into the intact whole. Soil physicists, who for a brief period of time in the 1980s toyed with the same general idea of starting from aggregates to upscale soil processes (e.g., van Genuchten, 1985), dismissed it rapidly and have not been pursuing it since, realizing that in more ways than one, it is a dead end road. Instrumental to this realization was the landmark article of Nkedi-Kizza et al. (1984), which demonstrated that macroscopic data could not help discriminate models assuming

non-equilibrium processes associated with the presence of aggregates from other models involving chemical non-equilibrium instead. The general sentiment among soil physicists about starting from aggregates to upscale the description of soil processes is stated clearly in the recent thorough review of the literature by Rabot et al. (2018), who conclude that "although appealing, the aggregate perspective does not seem to be the most appropriate to link soil structure with soil functions and processes."

Without going in great detail through all the relevant evidence, we would like to briefly discuss here the most salient objections to an aggregate-based framework such as that outlined by Wang et al. (2019), and thereby prevent researchers from venturing on a path that leads nowhere. The key objections have to do with size- and boundary conditions indeterminacies, as illustrated in Figure 1.

The first issue, of size indeterminacy, has been discussed extensively in the literature on soil aggregates (e.g., Dexter, 1988; Letey, 1991; Baveye, 2006). In a soil like that depicted in Figure 1, this issue does not matter much in the surface (Ah1) horizon, where the soil easily breaks down into aggregates, largely of biological origin, that have relatively well-defined sizes and round shapes (Figure 1b). But the situation can be very different for different soil types and for lower horizons in the soil profile. In the natural state, there is no clear evidence of the presence of aggregates. With the use of a knife, one can free from the soil mass some chunks of soil of relatively large size, which with further effort can be broken down in "aggregates" of progressively smaller sizes (Figure 1c). This feature is consistent with the hypothesis of a hierarchical architecture of aggregates, identified and described in detail by Tisdall and Oades (1982), and shows that the distribution of sizes of aggregates one obtains when dismantling a soil sample depends on the amount of energy that is applied to take soils apart. This operational issue, discussed by Amezketa (1999), is particularly well illustrated by the experimental results of Diaz-Zorita et al. (2002), who show that the size of fragments obtained by sieving soils is inversely related to the mechanical stress applied. Hallett et al. (2013) also point out that breakdown of soils by dynamic or static mechanical loading yields different fragmentations of soil aggregates. This dependence of the aggregate size distribution on the operational conditions under which it is obtained raises the question of

whether aggregates exist in soils in their natural state and are not merely artefacts (Young et al., 2001). This may perhaps explain, e.g., why some authors have failed to observe anticipated correlations between organic matter content and aggregation (Razafimbelo et al., 2013).

This size indeterminacy issue may not be too serious a problem as long as one is interested strictly in what happens entirely within an aggregate or, in columns that are repacked with aggregates, in the pore space between aggregates. In the first case, aggregates have been used extensively to understand at a very local scale in soils the interactions between pore geometry, chemical composition, and microbial activity. As long as aggregates are viewed strictly as chunks of 98 soil that are convenient to manipulate because they do not fall apart too easily, e.g., when they are rotated on the stage of a CT scanner, no harm is done in using aggregates to gain insight into microscale processes, as various authors have done successfully (Remusat et al., 2012; Ananyeva et al., 2013; Kravchenko et al., 2015; Voltolini et al., 2017; Yu et al., 2017). Similarly, for various reasons, in particular the need to use replicate soil columns, some authors have found it useful to isolate uniformly-sized aggregates from soils, and to repack them in a reproducible manner, prior to experiments to study a wide range of processes occurring within inter-aggregate pores (e.g., Pot et al., 2015; Juyal et al., 2018, 2019). As long as the repacked soil materials are not confused with the original soils, one could again consider that no real harm is done. Given the opportunity, it would always make more sense to work directly with undisturbed soil samples, in spite of their inherent heterogeneity, but as more reproducible alternatives, one has to acknowledge that repacked columns may occasionally be useful.

The situation is entirely different in the case of processes where energy transfer or material movement takes place across the external surfaces of aggregates, a situation that definitely occurs in many dynamical processes, and in particular during the release of GHG. In such situations, for the concept of soil aggregates as biogeochemical reactors to have operational meaning, i.e., in order for researchers to be able to perform representative experiments on aggregates, one would have to be able to replicate the exact same boundary conditions (of temperature, external moisture content and chemical composition, acidity, water potential, etc) that an

aggregate would have experienced had it remained in its original state in the soil. At the moment, this is completely beyond our capabilities. Even with the most advanced X-ray computed tomography (CT) scanners at our disposal, we would not be able to determine the size and shape of peripheral pores, and therefore their likely moisture content under field conditions. This in turn makes it virtually impossible to estimate the rate of release of GHG, which we know is strongly linked to moisture content (Rabot et al., 2018). Equally so these aggregates may have bordered large pores or cracks. As the nature of the neighboring space cannot be predicted from the aggregate itself, it is not possible now, and it does not seem feasible either in the foreseeable future, in spite of the major technological advances that are forthcoming (Baveye et al., 2018), to impose on artificially isolated soil aggregates the same type of boundary conditions that these aggregates would have experienced 130 in their undisturbed state.

Thus, aggregates are not a viable option in practice to measure the dynamic aspects of the release of GHG by soils, and therefore should not be regarded as "biogeochemical reactors" for upscaling purposes. Much more fruitful will be to focus on undisturbed soil samples of various sizes, taking advantage of technological developments that enable quantification of soil biochemistry in connection to soil physical processes, i.e., fluxes and transports, in the soil as a whole ecosystem (Baveye et al., 2018). There are still boundary conditions to be handled in undisturbed samples as well, but for the most part, especially when dealing with GHG release, one only has to ensure that the bottom boundary condition does not lead to artefacts, like water logging. This is far simpler to achieve with undisturbed, e.g., cylindrical, samples than in the case of oddly-shaped aggregates. If intact samples are taken carefully from soil, as soil scientists have done for many years, the boundary conditions are less likely to display the same discontinuity and uncertainty that occur with aggregates.

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Figure captions

Figure 1: (a) Profile of a soil located in Grignon (France) and classified alternatively as an Orthic luvisol (FAO classification) or hapludalf (U.S. Soil Taxonomy), (b) Round-shaped aggregates, of biological origin, found in the surface (Ah1) horizon, (c) Aggregates of progressively smaller sizes obtained by breaking downm by hand, large chunks of soil initially dislodged from the profile with a knife. The existence of a sequence of arbitrary aggregate sizes gives rise to a "size indeterminacy" problem, (d) Schematic illustration of the indeterminacy associated with the boundary conditions to be imposed on an aggregate, were one to try to characterize its dynamics in laboratory experiments.

