UC Irvine
UC Irvine Previously Published Works

Title

Soil aggregates as biogeochemical reactors and implications for soil–atmosphere exchange of greenhouse gases—A concept

Permalink

Journal

ISSN

Authors

Wang, Bin
Brewer, Paul E
Shugart, Herman H
et al.

Publication Date

DOI

Peer reviewed

1   **Title:** Soil aggregates as biogeochemical reactors and implications for soil-atmosphere exchange

2   of greenhouse gases—a concept

3   **Running head**: Soil aggregates as GHG reactors

4

5   **Author list:** Bin Wang [1,2], Paul E. Brewer [3], Herman H. Shugart [2], Manuel T. Lerdau [2], Steven

6   D. Allison [1,4]

7

8   [1] Department of Ecology and Evolutionary Biology, University of California, Irvine, CA, USA

9   [2] Department of Environmental Sciences, University of Virginia, Charlottesville, VA, USA

10   [3] Smithsonian Environmental Research Center, Edgewater, MD, USA

11   [4] Department of Earth System Science, University of California, Irvine, CA, USA

12

13   **Correspondence author:**

14   Bin Wang

15   Department of Ecology and Evolutionary Biology

16   University of California, Irvine

17   321 Steinhaus Hall, Irvine, CA 92697-2525 USA

18   wbwenwu@gmail.com or bw8my@virginia.edu

19

20   **Key words**: greenhouse gas, soil heterogeneity, aggregate reactor, soil organic matter,

21   microorganism, individual-based model, aggregate-based model

22

23   **Type of paper**: Opinion

24

Abstract

Soil-atmosphere exchange significantly influences the global atmospheric abundances of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). These greenhouse gases (GHGs) have been extensively studied at the soil profile level and extrapolated to coarser scales (regional and global). However, finer scale studies of soil aggregation have not received much attention, even though elucidating the GHG activities at the full spectrum of scales rather than just coarse levels is essential for reducing the large uncertainties in the current atmospheric budgets of these gases. Through synthesizing relevant studies, we propose that aggregates, as relatively separate micro-environments embedded in a complex soil matrix, can be viewed as biogeochemical reactors of GHGs. Aggregate reactivity is determined by both aggregate size (which determines the reactor size) and the bulk soil environment including both biotic and abiotic factors (which further influence the reaction conditions). With a systematic, dynamic view of the soil system, implications of aggregate reactors for soil-atmosphere GHG exchange are determined by both an individual reactor's reactivity and dynamics in aggregate size distributions. Emerging evidence supports the contention that aggregate reactors significantly influence soil-atmosphere GHG exchange and may have global implications for carbon and nitrogen cycling. In the context of increasingly frequent and severe disturbances, we advocate more analyses of GHG activities at the aggregate scale. To complement data on aggregate reactors, we suggest developing bottom-up aggregate-based models (ABMs) that apply a trait-based approach and incorporate soil system heterogeneity.

1 Introduction

Biogeochemical reactions are manifested at spatial scales ranging from molecule to globe (McClain et al., 2003). In the case of the biogeochemical processes specifically responsible for GHG production/consumption in soils, our understanding, however, is mostly derived from the soil profile level because of methodological constraints (Mosier et al., 1991; Smith et al., 2003). Larger scale understanding (from landscape through regional to global) is extrapolated from knowledge gained at this scale through scale-up exercises via a combination of land surface modeling and remote sensing of land cover (e.g., McClain et al., 2003). From these scaling efforts, it is clear that soil-atmosphere exchange significantly affects the atmospheric abundances of CO_2, CH_4, and N_2O regionally and globally (Ciais et al., 2013). This conclusion is confirmed by top-down constraints derived from satellite measurements (Ciais et al., 2013). Nevertheless, we still have very poor constraints on the global and regional balances of GHGs (e.g., Ciais et al., 2013; Saunois et al., 2016), though the underlying biogeochemical reactions responsible for the production and/or consumption of these gases in soils are relatively well understood [but see Wang et al. (2017a) for an emerging paradigm of GHG production via non-microbial pathways].

These uncertainties necessitate a more complete understanding of GHG activities at the full spectrum of spatial scales, especially the soil aggregate scale which is relatively less understood. Such information is essential for further elucidating complex processes resulting from soil heterogeneity and for guiding bottom-up modeling of soil-atmosphere exchange of GHGs (e.g., Hinckley et al., 2014; Ebrahimi & Or, 2016). This endeavor will eventually contribute to a better understanding of the mechanisms regulating atmospheric concentrations of GHGs and to improved strategies for mitigating soil GHG emissions in the context of global environmental changes, i.e., achieving 'climate-smart' soils (Paustian et al., 2016).

The soil system shows extremely high heterogeneity, and microbial activities are not spatially homogeneous in the soil matrix (Young & Crawford, 2004). From the perspective of structure, soil aggregates and pore spaces create fine-scale spatial heterogeneity (e.g., Elliott & Coleman, 1988; Horn et al.,1994; Rillig et al., 2017). Primary soil mineral particles (clay, silt, and sand) chemically interact with organic matter (historically classified as 'primary organo-mineral complexes'), forming the basic units of soil aggregates. These basic units can accrete into larger aggregates, depending on the availability of a diverse suite of binding agents (e.g., polyvalent cations: Ca^{2+} or Al^{3+}) and various forms of organic matter (e.g., polysaccharides, organic acids, plant debris, roots, and hyphae). From these aggregation processes a hierarchical system of soil aggregates emerges (Tisdall & Oades, 1982; Oades, 1991; Lehmann *et al*., 2007). Such aggregates generate additional soil heterogeneity, along with other 'hotspots' associated with the rhizosphere, detritusphere, and biopores that affect the distribution of substrates and microbial communities (Kuzyakov & Blagodatskaya, 2015).

Since the soil aggregate concept was first proposed about a century ago, extensive studies have been conducted on physiochemical and biological properties at this scale, as well as their responses to disturbances including soil management, land use change, and global change (*e.g*., Elliott, 1986; Oades, 1991; Jastrow, 1996; Six *et al*., 2000a, 2000b, 2004; Blanco-Canqui & Lal, 2004; Lehmann *et al.* 2017). However, knowledge about aggregate-scale GHG dynamics is still fairly scarce. To address GHG activities at this fine scale and hence their implications for coarse scale GHG exchange, here we propose that aggregates—as relatively independent micro-habitats in the soil matrix—can be viewed as biogeochemical reactors (hereafter referred to as aggregate reactors) that produce GHGs including CO_2, CH_4, and N_2O. Correspondingly, aggregate reactivity is defined as the potential for GHG production and the rate and duration of these reactions. Because

94   of varying sizes and turnover rates, we further argue that aggregate reactors in a soil system should

95   be viewed in a systematic and dynamic way. By proposing this aggregate reactor concept in a

96   dynamic framework, ecological theory can be applied to studies of GHG exchange by examining

97   both the reactivity of an aggregate reactor (physiology) and the compositional dynamics of

98   differing aggregate reactors (community ecology).

99   The overarching purpose of this paper is to demonstrate the rationale behind this concept,

100   discuss its implications for soil-atmosphere GHG exchange, and propose ways forward to improve

101   our understanding of aggregate reactors. By synthesizing previous studies, we first examine basic

102   characteristics of aggregate reactors. We then discuss factors influencing aggregate reactivity, such

103   as aggregate size, soil bulk properties, and fractionation methodology. Implications of aggregate

104   reactors for soil-atmosphere GHG exchange are also addressed. In light of increasingly frequent

105   and severe perturbations to soil systems, we emphasize impacts of soil management and global

106   change on aggregate reactors. We conclude by identifying current knowledge gaps and research

107   opportunities, including the potential to develop aggregate-based models (ABMs) that can

108   explicitly incorporate the structural heterogeneity of soil systems. We recommend integration of

109   soil science, ecology, and climate science communities to advance the aggregate reactor concept

110   and to develop a predictive framework based on aggregate reactors in the context of global change.

111   These efforts should eventually help reduce the large uncertainties in GHG exchange associated

112   with soil heterogeneity.

113

114   **2 The aggregate as a 'biogeochemical reactor' of GHG**

115   Embedded in the soil matrix, soil aggregates exhibit physical, chemical, and biological

116   properties that differ from the bulk soil (**Fig. 1**). Soil aggregates contain a three-dimensional

117 structure with pores of varying sizes (e.g., Ebrahimi & Or, 2016). Organic matter becomes

118 occluded during the aggregation process. Identifiable components of the occluded fraction include

119 small particles of incompletely decomposed organic residues, pollen grains, and particles of plant

120 tissue such as lignin coils and phytoliths. This physically bound organic matter, compared with

121 free organic matter, often has relatively higher carbon and nitrogen concentrations and contains

122 more alkyl carbon that is recalcitrant (Golchin *et al*. 1994a, b; Six et al., 2001b). Further, oxygen

123 (O_2), water, nutrients, and dissolved organic carbon diffuse into the aggregates from the inter-

124 aggregate voids or macropores of bulk soil (Keiluweit et al., 2016). For example, O_2 diffusion

125 from the soil matrix (where the O_2 concentration is further controlled by bulk soil moisture, plant

126 roots, and other factors) is limited by the pore networks of aggregates, whereas O_2 consumption in

127 aggregates is controlled by microbial activities, organic carbon, and nutrient availability. The net

128 result is that aggregates can experience O_2 limitations even within aerobic, well-drained soils,

129 making anoxia the most notable environmental characteristics of aggregates (e.g., Tiedje et al.

130 1984; Sexstone *et al*., 1985; Elliott & Coleman, 1988; Sexstone et al. 1988; Højberg *et al*., 1994;

131 Diba *et al*., 2011; Keiluweit et al., 2016).

132   A variety of anaerobic metabolic pathways can occur in aggregates (Ebrahimi & Or, 2015;

133 Keiluweit et al. 2016), including processes responsible for CH_4 and N_2O production such as

134 denitrification and methanogenesis (e.g., Sexstone et al. 1988; von Fischer & Hedin 2002;

135 Keiluweit et al. 2016; summarized in **Tables 1-3**). Therefore, aggregates can be viewed as

136 segregated biogeochemical reactors of GHG embedded in a complex soil matrix (**Fig. 1**). The

137 connectivity and tortuosity of pores and other bulk soil properties (e.g., soil texture, moisture, and

138 biological activities) determine the micro-environment in aggregates by regulating O_2 diffusion,

139 distribution of water films, and substrate and nutrient accessibility for microbes, as well as the

140 composition and structure of soil microbial community.

141   Microbial communities inhabit soil aggregates and exhibit dynamics in composition and

142 activity (e.g., Ebrahimi & Or, 2016). Recently, based on aggregates' isolation feature Rillig et al.

143 (2017) proposed that aggregates are 'incubators' of microbial evolution that allow processes

144 including genetic drift, natural selection, and mutation to occur and that likely produce an overall

145 effect of an increase of microbial diversity. This microbial evolution dimension further adds

146 complexity to understanding aggregate reactors.

147

148 **3 Factors regulating aggregate reactivity**

149 **3.1 Aggregate reactor size**

150   Aggregate reactor size is typically measured in terms of diameter. Aggregate reactor size

151 is an important factor for GHG reactivity because of size impacts on other physical, chemical, and

152 biological activities. Just as GHG exchange exhibits substantial variation at coarse spatial scales,

153 aggregate reactivity is also expected to vary across aggregate size classes. Here, we offer a non-

154 exhaustive review of the major differences between macro- (>0.25 mm) and micro-aggregates

155 (<0.25 mm), focusing on the aspects that could potentially result in reactivity differences.

156   First, aggregates of different sizes have differing geometry. One important aspect of the

157 geometry is mean pore size which is smaller for micro-aggregates than for macro-aggregates

158 (Dexter, 1988). This difference affects diffusion of O_2, nutrients, and dissolved organic carbon

159 (DOC). Compared with macro-aggregates, O_2 diffusion is slower into micro-aggregates (Sexstone

160 *et al*., 1985; Elliott & Coleman, 1988; Højberg *et al*., 1994; Denef *et al*., 2001; Diba *et al*., 2011).

161   Second, the chemical composition of substrates for carbon and nitrogen mineralization is

162 different. Macro-aggregates often have higher carbon and nitrogen concentrations (*e.g*., Elliott,

163 1986; Gupta & Germida, 1988; Cambardella & Elliott, 1993). Younger and more labile organic

164 matter (with a higher C/N ratio) constitutes more of the organic matter pool in macro-aggregates

165 than in micro-aggregates (Elliott, 1986; Elliott and Coleman 1988; Six et al., 2004).

166   Third, microbial community composition and structure are influenced by aggregate size

167 (e.g., Van Gestel et al., 1996; Mummey *et al*., 2006; Kravchenko et al., 2014; Rabbi et al., 2016;

168 Ebrahimi and Or, 2016). For instance, Mummey *et al*., 2006 found that microaggregates select for

169 specific microbial lineages across disparate soils. Bach et al. (2018) reported that microaggregates

170 hold more diverse microbial communities than macroaggregates.

171   Many studies have found differences in GHG process rates among aggregates of varying

172 sizes, though some studies have found no differences (**Table 1-3**). Sexstone *et al*. (1985) and later

173 studies found that the composition of microbial communities responsible for N_2O production

174 varies with aggregate size. Sey et al. (2008) suggested that ammonium oxidizers are most abundant

175 in macro-aggregates, while denitrifiers, which preferentially colonize anaerobic environments, are

176 more abundant in micro-aggregates. This study also found that denitrification pathways of N_2O

177 production dominate in smaller aggregates, whereas nitrification dominates N_2O production in

178 larger aggregates. In addition, greater denitrification rates occurred in the smallest aggregate size

179 fractions when acetylene was applied to prevent the complete reduction of N_2O to N_2 during

180 denitrification. Higher denitrification in smaller aggregates was also reported by Seech and

181 Beauchamp (1988) and Uchida *et al*., (2008). Sey et al. (2008) attributed this pattern to a higher

182 proportion of complete denitrification ($NO_3^- \rightarrow N_2O \rightarrow N_2$) because of very low or effectively

183 absent O_2 in smaller aggregates. An alternative explanation, proposed by Ebrahimi and Or (2016),

184   is that N_2O takes longer to diffuse out of larger aggregates and thus has more time to be completely

185   reduced from N_2O to N_2.

186   Overall, consistent relationships between aggregate reactor size and reactivity are difficult

187   to establish, even qualitatively (**Table 1-3**). Although the majority of studies with different types

188   of soils (>60%) support an overall positive relationship between aggregate size and CO_2

189   production, some studies observed a negative relationship. For N_2O production, the majority of

190   studies (almost 70%) support an overall positive relationship with aggregate size, of which more

191   than half found more N_2O production from macro-aggregates than micro-aggregates (**Table S3**).

192   Regarding CH_4, more studies observe that smaller aggregates act as consumers and larger

193   aggregates as producers, but this is rarely true for the specific comparison of macro- and micro-

194   aggregates (**Table S4**).

195

196   **3.2 Bulk soil properties**

197   Bulk soil properties determine the environment surrounding the aggregate reactors, which

198   largely shapes conditions in the reactors. The observed inconsistent relationships between

199   aggregate reactor size and reactivity across different studies, as discussed above (**Table 1-3**),

200   support this postulation: aggregate reactivity is not just determined by size but also by the bulk

201   soil properties. These bulk properties can be abiotic or biotic factors that regulate the soil physic-

202   chemical and biological environment.

203   Although it remains challenging to establish direct causal connections between aggregate

204   reactivity and bulk soil properties, a few studies have addressed these relationships. For instance,

205   soil water content can significantly affect aggregate-level CH_4 activities (Sey *et al.* 2008).

206   Aggregates with sizes < 0.25 mm and 0.25-2 mm consumed CH_4 at low water content but began

207   to produce CH_4 at higher water content (Sey et al. 2008). This same study also observed a

208   maximum CH_4 production rate at 40% water-filled pore space (WFPS) for 2-6 mm aggregates.

209   This pattern can be explained by greater methanogenesis when high water content prevents O_2

210   diffusion (Yavitt *et al*., 1990). Regarding CO_2, studies by Drury *et al*. (2004) and Mangalassery *et*

211   *al*. (2013) both observed the largest CO_2 effluxes from small sized aggregates in clay loam soils,

212   in contrast to other studies using different soils (**Table 1**). This observed difference in CO_2

213   production may have resulted from texture differences that influence soil porosity and water film

214   distribution.

215   Many other factors, especially biotic ones, that can also affect aggregate reactor conditions

216   have not been studied at all. For instance, a soil O_2 concentration decline can be induced directly

217   by root respiration and/or by root exudation that stimulates heterotrophic respiraton in the

218   rhizosphere (Keiluweit *et al*., 2016). Other soil organisms, such as soil fauna and fungal hyphae,

219   can affect soil porosity and change the diffusivity of O_2 into aggregates or significantly affect the

220   formation of soil aggregates and their associated C pools (Lehmann *et al.,* 2017). Based on this

221   reasoning, biological activities should exert a variety of effects on aggregate-level GHG dynamics.

222

223   **3.3 Fractionation method**

224   Theoretically, aggregate reactivity should be determined by both soil properties and

225   aggregate reactor size. In practice, however, the observed variability of aggregate reactivity (**Table**

226   **1-3**) may also reflect differences in fractionation techniques. Separation methods are not uniform

227   across studies; specifically, dry-sieving is used more often than wet-sieving (**Table 1-3**).

228   Separation exerts significant influences on physical, chemical, and biological properties of

229   different sized aggregates (Ashman *et al*., 2003; Bach & Hofmockel, 2014; Kaiser *et al*., 2015).

230 For instance, air-drying can increase the mechanical strength of aggregates, and wet-sieving can

231 increase potential enzyme activity (Bach & Holmockel, 2014). A recent review by Kaiser *et al*.

232 (2015) provides further details on air-drying and rewetting effects on soil aggregate stability.

233 Currently, we know of only one study by Beauchamp & Seech (1990) that evaluated impacts of

234 dry- and wet-sieving methods on aggregate reactivity. They observed decreased denitrification

235 rates as the dry-sieved aggregate size increased, but the opposite relationship for wet-sieved

236 aggregates. More experiments testing the effects of separation techniques on GHG activities across

237 aggregates of different sizes are needed to inform future studies of environmental effects on

238 aggregate reactivity.

239

240 **4 Aggregate reactors and soil-atmosphere GHG exchange**

241 To link the fine-scale aggregate reactors and soil profile GHG exchange, here we argue

242 that a systematic, dynamic view of the soil system is required. Specifically, we need to focus on

243 both the reactivity of an individual aggregate and also the composition of aggregate reactors of

244 different sizes in a soil system. This dual focus is necessary because aggregate reactors of different

245 sizes collectively make different contributions to soil profile GHG exchange. For example,

246 Bandyopadhyay & Lal (2014) report a much higher contribution to bulk soil CO_2 emissions from

247 macro-aggregates than micro-aggregates. The relative proportions of different size aggregate

248 reactors in a soil system are dynamically changing, and these changes are determined by the

249 turnover of aggregates (i.e. aggregate stability). Aggregate turnover is strongly influenced by

250 external disturbances to soil systems (Six et al. 2004). This by analogy is similar to the space-

251 lifetime hypothesis for organisms proposed by Ginzburg & Damuth (2008). We should view an

252 aggregate reactor in four dimensions—in addition to its three dimensional spatial structure, one

253   more temporal dimension, aggregate lifetime, should be included. Therefore, as with ecological

254   systems (e.g., Levin 1998; Grimm et al. 2005; Wang et al. 2017b), we need a dynamic view of

255   aggregate reactors to account for their compositional dynamics (**Fig. 1**).

256   Current evidence has already unequivocally suggested that aggregate reactors have

257   significant implications for soil-atmosphere exchange of GHG. Formation of these reactors

258   suppresses carbon oxidation and CO_2 release because of oxygen limitation, promoting carbon

259   sequestration (Six et al. 2002; Keiluweit et al. 2016; Keiluweit et al. 2017). While revisiting two

260   prior studies (Greenwood 1961; Sexstone et al. 1985), Keiluweit et al. (2016) offered an initial

261   estimate of aggregation effects on bulk soil carbon mineralization, showing a striking suppression

262   in the range of 23-97.5% relative to fully aerobic soils. By contrast, when these reactors are

263   'destroyed' (e.g., by disturbance from tillage), carbon mineralization increases rapidly because of

264   increased O_2 availability (e.g. Elliott 1986, Beare et al. 1994, and Drury et al. 2004). In particular,

265   Keiluweit et al. (2017) recently reported that shifting from anaerobic to aerobic conditions leads

266   to a 10-fold increase in volume-specific mineralization rate, illustrating the sensitivity of

267   anaerobically protected carbon to disturbance. These results, meanwhile, indirectly substantiate

268   the strong physical protection of organic matter against decomposition offered by the aggregates

269   (Six et al., 2000a, 2002).

270   In addition, the aggregate reactor concept illustrates that most field measurements of net

271   soil–atmosphere exchange mask significant gross production and consumption of CH_4 and N_2O.

272   Fine-scale activities are disguised in the traditional metrics of soil-atmosphere exchange of CH_4 at

273   larger scales (von Fischer & Hedin, 2002), where a soil is considered either a sink or a source.

274   Multiple studies have shown that higher *in situ* gross CH_4 production could stimulate higher gross

275   consumption, resulting in little difference in surface fluxes (Kammann *et al*., 2009; Mangalassery

12

276 *et al.*, 2013; Yang & Silver, 2016). Similar to CH$_4$, N$_2$O is could also be both produced and

277 consumed within a soil (Chapuis-Lardy *et al.*, 2007), so aggregate scale production of N$_2$O may

278 not always increase surface fluxes (Yang & Silver, 2016). In summary, aggregate reactors have

279 significant implications for soil-atmosphere exchange of GHG.

280

281 **5 Aggregate reactors in the context of global change**

282 The soils beneath our feet are strongly affected by coarse-scale disturbances including soil

283 management practices (mostly agronomic practices), land use change, and global changes resulting

284 from growing human activities in the Anthropocene Epoch (Bronick & Lal 2005; Hinckley, 2014;

285 Paustian et al. 2016). With the aggregate reactor concept and a systematic view of soil systems as

286 discussed above, coarse scale perturbations of soil systems are postulated to affect soil-atmosphere

287 GHG exchange both directly by influencing conditions for aggregate reactivity and indirectly by

288 altering the distribution of aggregate reactor sizes (**Fig. 1**). Soil management (e.g., soil cultivation,

289 fertilization, crop rotation, irrigation, biochar addition, and compaction) and land use change can

290 significantly affect the size distribution of aggregates (Six et al., 1999, 2000b; Young et al., 2000;

291 Wang et al., 2013b). Climate warming increases the soil temperature, while precipitation shifts

292 alter soil moisture content, shaping the environment of aggregate reactors and thus aggregate-level

293 microbial activities (e.g., Fang et al. 2015). Moreover, climate change can also indirectly affect

294 soil aggregate properties by influencing vegetation activity (Torn *et al.*, 2015). Additionally,

295 atmospheric changes can indirectly affect soil structure by influencing above-ground vegetation

296 activities and carbon and nitrogen allocation. Rising CO$_2$ levels can alter soil structure and increase

297 soil aggregation and carbon sequestration (Cotrufo *et al.*, 2013; Rillig *et al.*, 1999; Six *et al.*, 2001a;

298 Jastrow *et al.*, 2005; Dorodnikov *et al.*, 2009). Nitrogen deposition can also shape soil aggregate

299　properties by influencing rhizodeposition, microbial biomass and microbial activity (Janssens *et*

300　*al.,* 2010). Increasing abundance of tropospheric ozone (O_3), the most important secondary air

301　pollutant, can modify the soil structure in terms of aggregate properties and distribution and soil-

302　atmosphere GHG exchange (Kou *et al*., 2014; Wang et al. 2017c).

303　　　　Still, relatively few studies have addressed direct connections between these disturbances

304　and reactions responsible for GHG production from aggregate reactors. The available studies

305　mainly focus on aggregate responses to tillage and fertilization, while the indirect effects mediated

306　by aggregate turnover and size distribution changes are still unknown. In general, macro-

307　aggregates from no-till soils have higher CO_2 production than those from soils under conventional

308　tillage (Franzluebbers & Arshad, 1997; Fernández *et al*., 2010). Moreover, the tillage impact

309　depends upon soil depth. Fernández *et al*. (2010) demonstrated that differences in CO_2 production

310　between tillage practices disappear for deeper soils. One possible explanation is that no-till soils

311　show a pattern of decreasing of SOC with depth whereas conventional soils have uniformly

312　distributed SOC (Fernández *et al*., 2010; Plaza-Bonilla *et al*., 2014). This pattern might also

313　explain why Plaza-Bonilla *et al*. (2014) did not observe differences between no-till and

314　conventional tillage.

315　　　　Similar to CO_2, CH_4 production and consumption is affected by tillage. A study by Plaza-

316　Bonilla *et al*. (2014) reported that macro-aggregates act as CH_4 sources under conventional tillage

317　and sinks under no tillage. This change could be attributed to inhibited methanotrophic activity

318　induced by aggregate destruction under tillage, or, alternatively, to a smaller quantity of anoxic

319　microsites within the no-tillage macro-aggregates maintained by intra-aggregate pore architecture

320　and connectivity (*e.g*., Brewer *et al.,*2018). In contrast, both Jiang *et al*. (2011) and Plaza-Bonilla

321　*et al*. (2014) reported that soil tillage did not affect aggregate N_2O production.

322   Fertilization has been reported to affect aggregate-level N_2O production with the effects

323   dependent on fertilizer type. For example, Plaza-Bonilla *et al.* (2014) concluded that mineral and

324   organic nitrogen fertilizers can lead to differences in the relative importance of nitrification versus

325   denitrification in macro-aggregates: nitrification dominates with mineral fertilizer whereas

326   denitrification dominates with organic fertilizer (pig slurry). This difference was attributed to

327   changes in the proportion of C and N substrates and in microbial activities (Plaza-Bonilla *et al.*

328   2014).

329

330   **6 Prospects and aggregate-based modeling (ABM)**

331   Generations of research have built a relatively solid knowledge of reactions broadly

332   responsible for GHG production and of aggregates' inherent biophysical and chemical properties.

333   Yet an understanding of the direct relationships between these properties and reactions specifically

334   at the aggregate level remains elusive. Emerging new mechanisms, such as anaerobic oxidation of

335   CH_4, require additional investigation. Likewise, bulk soil conditions are still not yet fully linked

336   to aggregate reactivity. Aside from the soil hydration-O_2 diffusion relationship (e.g., Ebrahimi &

337   Or, 2017), many abiotic and biotic factors that could regulate aggregate reactivity have not been

338   studied. As pointed out by Torn *et al.* (2015), two of the most widespread impacts of anthropogenic

339   activities on soils in this century will be warmer temperatures and altered plant allocation

340   belowground because of rising CO_2 and nitrogen deposition. Therefore, more studies are needed

341   to understand how soil temperature and biotic factors (*e.g.*, root activity, plant species, and soil

342   macro-fauna), as well as soil management practices, affect aggregate GHG fluxes. Additionally,

343   how microbial community composition and dynamics control these activities in aggregates is

344   almost unknown (Allison et al. 2013; Ebrahimi & Or, 2016; Buchkowski et al. 2017). Finally,

345 future studies should move beyond the reactivity of an individual aggregate. A systematic,

346 dynamic view of these reactors in soil systems is equally important for fully quantifying the

347 implications of aggregate reactors for soil-atmosphere GHG exchange.

348   Driven by these grand questions, we should simultaneously refine and develop more

349 standardized methodologies for the study of aggregate reactors. Undoubtedly, the ideal route is to

350 conduct *in situ* measurements, which is a great challenge at the micron-scale with current

351 techniques. Therefore, we recommend a combination of the following techniques. First, it is

352 essential to establish a uniform experimental protocol for aggregate fractionation. The approach

353 of optimal moisture fractionation by Bach and Hofmockel (2014) has a high potential to separate

354 aggregates with minimal disturbance to chemical and biological properties and might be ideal for

355 studies of aggregre reactors. As a complement to aggregate fractionation, artificial aggregates are

356 a good means for exploring the relationships between aggregate reactivity and physical structure

357 (e.g., Ebrahimi and Or 2017; Schlüter et al. 2018). Another area of technique development could

358 exploit isotope pool dilution to measure gross fluxes of GHGs, followed by separation of soil

359 aggregates to determine correlations between aggregate size distributions, physio-chemical

360 properties, and the gross gas fluxes [*e.g*., CH_4 by von Fischer & Hedin (2002) and N_2O by Yang

361 *et al*., (2011)]. These methods could be combined with technique advancements in computer-aided

362 tomography (CT) and electron microscopy (e.g., SEM and TEM) (Williams & Carter 1996) that

363 provide soil structural information in terms of aggregate reactor size and distribution (e.g., Young

364 & Crawford 2004; Rabbi et al. 2016).

365   Finally, a major research need is to develop computational models that can quantify and

366 predict aggregate reactivity. These models could untangle the non-linearities between aggregate

367 reactors and soil profile GHG exchange. Such research would address the grand challenge of

368 modelling soil biogeochemical processes at larger spatial and temporal scales in the context of

369 global environmental changes. With a systematic, dynamic view of aggregate reactors in soil

370 systems as discussed above, we propose a bottom-up strategy to develop aggregate-based models

371 (ABM) that explicitly represent the 'behavior' of aggregate reactors of different sizes (**Fig. 1**). This

372 approach is inspired by the agent-based or individual-based modeling (IBM) strategy that largely

373 originated in ecological systems in the 1960s (Grimm et al. 2005; Shugart et al. 2018). For a soil

374 system composed of aggregate reactors of different sizes, an ABM framework could be developed

375 to represent these different aggregates. For each aggregate reactor, a single IBM would be

376 developed to explicitly simulate microbial communities and their functions. Therefore, an ABM

377 is expected to be a hierarchy of individual-based models simulating each of an aggregate element

378 and its dynamic properties. Such a hierarchically constructed ABM contrasts with traditional

379 models that represent soils as a set of discrete carbon fractions with an implicit treatment of

380 microbial diversity (e.g., reviewed in Bradford *et al.,* 2016). We also advocate the incorporation

381 of a trait-based approach in the ABM based on trait data from aggregate reactors. These data could

382 include the physical (e.g., aggregate size, pore size, gas diffusion coefficient), chemical (e.g.,

383 substrates, enzyme, O_2, and moisture), and biological properties (i.e., turnover rate and microbial

384 diversity) of soil aggregates and could be obtained by the techniques described above (i.e. 'top-

385 down experiment'; **Fig.1**). We anticipate that a global soil aggregate trait database can be

386 established and that tradeoffs among aggregate traits (*e.g.*, reactor size and O_2 abundance) could

387 be uncovered to facilitate the model construction, similar to previous successful applications with

388 plant traits (*e.g.,* Kattge et al. 2011; Wright et al. 2014) and even litter decomposition (Allison

389 2012). Biophysical equations that may be helpful in building such an ABM are listed in the

390 Supporting Information.

391   Previous workers have built a solid foundation for developing ABMs based on mechanistic

392   modelling of soil processes that occur in aggregate reactors. Smith (1980) developed a model of

393   the variation in the extent of anaerobiosis in aggregated soils by extending previously published

394   models of radial diffusion into individual aggregates (e.g., Currie 1962). This work assumed a log-

395   normally distributed population of aggregate sizes to calculate denitrification rates. Recently,

396   Ebrahimi & Or (2015) embedded an individual-based microbial model [inherited from Kreft et al.

397   (1998)] into an idealized artificial aggregate and developed an analytical model for

398   biogeochemical processes in aggregates. The model was later expanded to include aggregates of

399   different sizes to simulate CO_2 and N_2O fluxes (Ebrahimi & Or 2016). Moreover, Ebrahimi & Or

400   (2018) applied their aggregate-based model to scale up microbial processes in aggregates of

401   different sizes. They used spatial data on soil type and land cover to simulate GHG exchange at

402   the landscape scale. Future modeling efforts should dynamically represent aggregate reactivity and

403   distribution and microbial community composition over space. The resulting models should be

404   validated under different soil conditions and management practices across spatial and temporal

405   scales. These efforts will likely require cooperation among modelers, ecologists, microbiologists,

406   and climate scientists to advance a predictive science of land-atmosphere exchange of GHG in the

407   context of global environmental change (BERAC 2017).

408

7 Conclusions

410   Soil GHG exchanges are manifested at a wide spectrum of scales from the aggregate

411   through the soil profile to the landscape, region, and globe. Understanding GHG exchange at these

412   different scales is essential for a more accurate quantification of trace gas fluxes and better

413   evaluation of land-atmosphere interactions in the context of accelerating global change. Based on

414 a synthesis of prior studies, we argue that aggregates can be viewed as biogeochemical reactors of

415 GHGs, with reactivities dependent on aggregate size and bulk soil abiotic and biotic factors that

416 shape the reaction environment. We also suggest a systematic, dynamic approach to link the

417 individual aggregate reactor with GHG exchange from the soil profile. Prior work has already

418 revealed implications of aggregate reactors for the soil-atmosphere exchange of GHG—for

419 example, soil carbon oxidation can be suppressed in aggregate reactors to facilitate carbon

420 sequestration. However, our understanding of aggregate reactors is far from complete. We

421 advocate for more research on techniques, environmental drivers, and cross-scale linkages related

422 to the aggregate reactor concept. There is also great potential for developing mechanistic,

423 aggregate-based models that use a trait-based approach to represent soil systems and reduce

424 uncertainties about soil-atmosphere GHG exchange in the face of human impacts.

425

426

435

436

437   **References**

438   Allison SD (2012) A trait-based approach for modelling microbial litter decomposition.

439   Ecology Letters, **15**, 1058-1070.

440   Allison SD, Lu Y, Weihe C, Goulden ML, Martiny AC, Treseder KK, Martiny JBH (2013)

441   Microbial abundance and composition influence litter decomposition response to

442   environmental change. Ecology, **94**, 714–725.

443   Aoyama M, Angers D, N'dayegamiye A, Bissonnette N (1999) Protected organic matter in

444   waterstable aggregates as affected by mineral fertilizer and manure applications. Canadian

445   Journal of Soil Science, **79**, 419-425.

446   Ashman M, Hallett P, Brookes P (2003) Are the links between soil aggregate size class, soil

447   organic matter and respiration rate artefacts of the fractionation procedure? Soil Biology

448   Biochemistry, **35**, 435-444.

449   Bach EM, Hofmockel KS (2014) Soil aggregate isolation method affects measures of

450   intra-aggregate extracellular enzyme activity. Soil Biology and Biochemistry, **69**, 54-62.

451   Bach EM, Williams RJ, Hargreaves SK, Yang F, & Hofmockel KS (2018) Greatest

452   soil microbial diversity found in micro-habitats. Soil Biology and Biochemistry, **118**, 217-226.

453   Bais HP, Weir TL, Perry LG, Gilroy S, Vivanco JM (2006) The role of root exudates in

454   rhizosphere interactions with plants and other organisms. Annual Review of Plant Biology, **57**,

455   233-266.

456   Bandyopadhyay KK, Lal R (2014) Effect of land use management on greenhouse gas emissions

457   from water stable aggregates. Geoderma, **232–234**, 363-372.

458   Bardgett RD, Mommer L, De Vries FT (2014) Going underground: root traits as drivers of

459   ecosystem processes. Trends in Ecology and Evolution, **29**, 692-699.

460 Bayer C, Gomes J, Zanatta JA, Vieira FCB, de Cássia Piccolo M, Dieckow J, Six J (2015) Soil

461   nitrous oxide emissions as affected by long-term tillage, cropping systems and nitrogen

462   fertilization in Southern Brazil. Soil and Tillage Research, **146**, 213-222.

463 Beare M, Hendrix P, Cabrera M, Coleman D (1994) Aggregate-protected and unprotected organic

464   matter pools in conventional-and no-tillage soils. Soil Science Society of America

465   Journal, **58**, 787-795.

466 Beauchamp EG, Seech AG (1990) Denitrification with different sizes of soil aggregates obtained

467   from dry-sieving and from sieving with water, Biology and Fertility of Soils, **10**,

468   188-193.

469 Bimüller C, Kreyling O, Kölbl A, von Lützow M, Kögel-Knabner I (2016) Carbon and nitrogen

470   mineralization in hierarchically structured aggregates of different size. Soil and Tillage

471   Research, **160**, 23-33.

472 Blanco-Canqui H, Lal R (2004) Mechanisms of Carbon Sequestration in Soil Aggregates.

473   Critical Review in Plant Sciences, **23**, 481-504.

474 Bradford, M. A., Wieder, W. R., Bonan, G. B., Fierer, N., Raymond, P. A., & Crowther, T. W.

475   (2016). Managing uncertainty in soil carbon feedbacks to climate change. Nature Climate

476   Change, 6, 751-758.

477 BERAC. 2017. Grand Challenges for Biological and Environmental Research: Progress and Future

478   Vision; A Report from the Biological and Environmental Research Advisory Committee,

479   DOE/SC–0190, BERAC Subcommittee on Grand Research Challenges for Biological and

480   Environmental Research (science.energy.gov/~/media/ber/berac/pdf/Reports/BERAC-2017-

481   Grand-Challenges-Report.pdf).

482 Brewer PE, Calderón F, Vigil M, von Fischer JC (2018) Impacts of moisture, soil respiration, and

483   agricultural practices on methanogenesis in upland soils as measured with stable isotope pool

484   dilution. Soil Biology and Biochemistry, **127**, 239-251.

485 Bronick C, Lal R (2005) Soil structure and management: a review. Geoderma, **124**, 3-22.

486 Buchkowski RW, Bradford MA, Grandy AS, Schmitz OJ, Wieder WR (2017) Applying

487   population and community ecology theory to advance understanding of belowground

488   biogeochemistry. Ecology Letters, **20**, 231-245.

489 Cambardella C, Elliott E (1993) Carbon and nitrogen distribution in aggregates from cultivated

490   and native grassland soils. Soil Science Society of America Journal, **57**, 1071-1071.

491 Chapuis-Lardy L, Wrage N, Metay A, Chotte JL, Bernoux M (2007) Soils, a sink for N_2O? a

492   review. Global Change Biology, **13**, 1-17.

493 Ciais P, Sabine C, Bala G *et al.* (2013) Carbon and other biogeochemical cycles. In: *Climate*

494   *Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth*

495   *Assessment Report of the Intergovernmental Panel on Climate Change.* (eds Stocker TF, Qin

496   D, Plattner GK, Tignor M, Allen SK, Boschung J, Nauels A, Xia Y, Bex V, Midgley PM) pp

497   465-570. Cambridge University Press, Cambridge.

498 Cotrufo MF, Gorissen A (1997) Elevated CO_2 enhances below-ground C allocation in three

499   perennial grass species at different levels of N availability. New Phytologist, **137**, 421-431.

500 Currie JA (1962) The importance of aeration in providing the right conditions for plant growth.

501   Journal of the Science of Food and Agriculture, 13, 380–385.

502 Del Galdo I, Six J, Peressotti A, Francesca Cotrufo M (2003) Assessing the impact of land-use

503   change on soil C sequestration in agricultural soils by means of organic matter fractionation

504   and stable C isotopes. Global Change Biology, **9**, 1204-1213.

505   Denef K, Six J, Bossuyt H, Frey SD, Elliott ET, Merckx R, Paustian K (2001) Influence of dry–

506   wet cycles on the interrelationship between aggregate, particulate organic matter, and

507   microbial community dynamics. Soil Biology and Biochemistry, **33**, 1599-1611.

508   Dexter, AR (1988) Advances in characterization of soil structure. Soil and tillage Research, **11**,

509   199-238.

510   Diba F, Shimizu M, Hatano R (2011) Effects of soil aggregate size, moisture content and fertilizer

511   management on nitrous oxide production in a volcanic ash soil. Soil Science and Plant

512   Nutrition, **57**, 733-747.

513   Dorodnikov M, Blagodatskaya E, Blagodatsky S, Marhan S, Fangmeier A, Kuzyakov Y (2009)

514   Stimulation of microbial extracellular enzyme activities by elevated CO_2 depends on soil

515   aggregate size. Global Change Biology, **15**, 1603-1614.

516   Drury CF, Yang XM, Reynolds WD, Tan CS (2004) Influence of crop rotation and aggregate

517   size on carbon dioxide production and denitrification. Soil & Tillage Research, **79**, 87-100.

518   Ebrahimi A & Or D (2015) Hydration and diffusion processes shape microbial community

519   organization and function in model soil aggregates. Water Resources Research, **51**, 9804-9827.

520   Ebrahimi A, Or D (2016) Microbial community dynamics in soil aggregates shape

521   biogeochemical gas fluxes from soil profiles–upscaling an aggregate biophysical model.

522   Global Change Biology, **22**, 3141-3156.

523   Ebrahimi A, Or D (2018) Dynamics of soil biogeochemical gas emissions shaped by remolded

524   aggregate sizes and carbon configurations under hydration cycles. Global Change Biology, **24**,

525   378-392.

526   Elliott E (1986) Aggregate structure and carbon, nitrogen, and phosphorus in native and

527   cultivated soils. Science Society of America Journal, **50**, 627-633.

528 Elliott ET, Coleman DC (1988) Let the soil work for us. Ecological Bulletins, 23-32.

529 Fang X, Zhou G, Li Y, Liu S, Chu G, Xu Z, Liu J (2015) Warming effects on biomass and

530 composition of microbial communities and enzyme activities within soil aggregates in

531 subtropical forest. Biology and Fertility of Soils, **52**, 1-13.

532 Fernández R, Quiroga A, Zorati C, Noellemeyer E (2010) Carbon contents and respiration rates

533 of aggregate size fractions under no-till and conventional tillage. Soil & Tillage Research, **109**,

534 103-109.

535 Franzluebbers A, Arshad M (1997) Soil microbial biomass and mineralizable carbon of water-

536 stable aggregates. Soil Science Society of America Journal, **61**, 1090-1097.

537 Lehmann A, Zheng W, Rillig MC (2017) Soil biota contributions to soil aggregation.

538 Nature Ecology and Evolution, **1**, 1828.

539 Ginzburg L, Damuth J (2008) The space-lifetime hypothesis: viewing organisms in

540 four dimensions, literally. The American Naturalist, **171**, 125-131.

541 Golchin A, Oades JM, Skjemstad JO, Clarke P (1994a) Study of free and occluded particulate

542 organic matter in soils by solid state ^{13}C Cp/MAS NMR spectroscopy and scanning electron

543 microscopy. Soil Research **32**, 285-309.

544 Golchin A, Oades JM, Skjemstad JO, Clarke P (1994b) Soil structure and carbon cycling. Soil

545 Research **32**, 1043-1068.

546 Grimm V, Revilla E, Berger U, Jeltsch F, Mooij WM, Railsback SF, ... and DeAngelis,

547 DL (2005) Pattern-oriented modeling of agent-based complex systems: lessons from

548 ecology SCIENCE **310**, 987-991.

549 Gupta V, Germida J (1988) Distribution of microbial biomass and its activity in different soil

550 aggregate size classes as affected by cultivation. Soil Biology and Biochemistry, **20**, 777-786.

551   Hallett P, Feeney D, Bengough AG, Rillig M, Scrimgeour C, Young I (2009) Disentangling the

552   impact of AM fungi versus roots on soil structure and water transport. Plant and Soil, **314**,

553   183-196.

554   Hinckley, ELS, W Wieder, N Fierer, E Paul (2014) Digging into the world beneath our feet:

555   bridging across scales in the age of global change. Eos Transaction AGU, **95**, 96-97.

556   Højberg O, Revsbech NP, Tiedje JM (1994) Denitrification in soil aggregates analyzed with

557   microsensors for nitrous oxide and oxygen. Soil Science Society of America Journal, **58**,

558   1691-1698.

559   Horn R, Taubner H, Wuttke M, Baumgartl, T (1994) Soil physical properties related to soil

560   structure. Soil & Tillage Research, **30**, 187-216.

561   Jäckel U, Schnell S, Conrad R (2001) Effect of moisture, texture and aggregate size of paddy

562   soil on production and consumption of CH_4. Soil Biology and Biochemistry, **33**, 965-971.

563   Janssens IA, Dieleman W, Luyssaert S, Subke JA, Reichstein M, Ceulemans R, ... &

564   Papale D (2010) Reduction of forest soil respiration in response to nitrogen deposition. Nature

565   Geoscience, **3**, 315-322.

566   Jastrow J (1996) Soil aggregate formation and the accrual of particulate and mineral-associated

567   organic matter. Soil Biology Biochemistry, **28**, 665-676.

568   Jastrow JD, Michael Miller R, Matamala R, Norby RJ, Boutton TW, Rice CW, Owensby, CE

569   (2005) Elevated atmospheric carbon dioxide increases soil carbon. Global Change Biology,

570   **11**, 2057-2064.

571   Jastrow JD, Amonette JE, Bailey VL (2007) Mechanisms controlling soil carbon turnover and

572   their potential application for enhancing carbon sequestration. Climate Change, **80**, 5-23.

573 Jiang X, Shi X, Liu W, Wright A (2011) Kinetics of net nitrification associated with soil

574 aggregates under conventional and no-tillage in a subtropical rice soil. Plant and Soil, **347**,

575 305-312.

576 Kaiser M, Kleber M, Berhe AA (2015) How air-drying and rewetting modify soil organic matter

577 characteristics: An assessment to improve data interpretation and inference. Soil Biology and

578 Biochemistry, **80**, 324-340.

579 Kammann C, Hepp S, Lenhart K, and Müller C (2009) Stimulation of methane consumption

580 by endogenous CH_4 production in aerobic grassland soil. Soil Biology and Biochemistry, **41**,

581 622-629.

582 Keiluweit M, Nico PS, Kleber M, Fendorf S (2016) Are oxygen limitations under recognized

583 regulators of organic carbon turnover in upland soils? Biogeochemistry, **127**, 157-171.

584 Keiluweit M, Wanzek T, Kleber M, Nico P, Fendorf S (2017) Anaerobic microsites have

585 an unaccounted role in soil carbon stabilization. Nature communications, **8**, 1771.

586 Kimura SD, Melling L, Goh KJ (2012) Influence of soil aggregate size on greenhouse gas emission

587 and uptake rate from tropical peat soil in forest and different oil palm development years.

588 Geoderma, **185–186**, 1-5.

589 Kou T, Wang L, Zhu J, Xie Z, Wang Y (2014) Ozone pollution influences soil carbon and nitrogen

590 sequestration and aggregate composition in paddy soils. Plant and Soil, **380**, 305-313.

591 Kravchenko AN, Negassa WC, Guber AK, Hildebrandt B, Marsh TL, & Rivers ML

592 (2014) Intra-aggregate pore structure influences phylogenetic composition of bacterial

593 community in macroaggregates. Soil Science Society of America Journal, **78**, 1924-1939.

594 Kreft JU, Booth G, Wimpenny JW (1998) BacSim, a simulator for individual-based

595 modelling of bacterial colony growth. Microbiology, **144**, 3275-3287.

596   Kuzyakov Y, Blagodatskaya E (2015) Microbial hotspots and hot moments in soil: Concept and

597   review. Soil Biology Biochemistry, **83**, 184-199.

598   Lehmann J, Kinyangi J, Solomon D (2007) Organic matter stabilization in soil microaggregates:

599   implications from spatial heterogeneity of organic carbon contents and carbon forms.

600   Biogeochemistry, **85**, 45-57.

601   Levin SA (1998) Ecosystems and the biosphere as complex adaptive systems. Ecosystems,

602   **1**, 431-436.

603   Mangalassery S, Sjögersten S, Sparkes D, Sturrock C, Mooney S (2013) The effect of soil

604   aggregate size on pore structure and its consequence on emission of greenhouse gases. Soil

605   Tillage Research, **132**, 39-46.

606   Manucharova N, Stepanov A, Umarov M (2001) Microbial transformation of nitrogen in water-

607   stable aggregates of various soil types. Eurasian Soil Science, **34**, 1125-113.

608   McClain ME, et al. (2003) Biogeochemical hot spots and hot moments at the interface of terrestrial

609   and aquatic ecosystems. Ecosystems, **6**, 301-312.

610   Mosier A, Schimel D, Valentine D, Bronson K, Parton W (1991) Methane and nitrous

611   oxide fluxes in native, fertilized and cultivated grasslands. Nature, **350**, 330-332.

612   Mummey D, Holben W, Six J, Stahl P (2006) Spatial stratification of soil bacterial populations in

613   aggregates of diverse soils. Microbial Ecology, **51**, 404-411.

614   Muñoz C, Torres P, Alvear M, Zagal E (2012) Physical protection of C and greenhouse gas

615   emissions provided by soil macroaggregates from a Chilean cultivated volcanic soil. Acta

616   Agriculturae Scandinavica, Section B–Soil & Plant Science, **62**, 739-748.

617   Oades J (1991) Aggregate hierarchy in soils. Soil Research, **29**, 815-828.

618 Papadopoulos A, Bird NRA, Whitmore AP, Mooney SJ (2009) Investigating the effects of
619   organic and conventional management on soil aggregate stability using X-ray computed
620   tomography. European Journal of Soil Science, **60**, 360-368.

621 Paustian K, Lehmann J, Ogle S, Reay D, Robertson GP, and Smith P (2016) Climate-smart
622   soils. Nature, **532**, 49-57.

623 Plaza-Bonilla D, Cantero-Martínez C, Álvaro-Fuentes J (2014) Soil management effects on
624   greenhouse gases production at the macroaggregate scale. Soil Biology and Biochemistry, **68**,
625   471-481.

626 Rabbi, SMF et al. (2016) Physical soil architectural traits are functionally linked to carbon
627   decomposition and bacterial diversity. Scientific Reports 6, 33012.

628 Rabbi SMF, Hua Q, Daniel H, Lockwood PV, Wilson BR, Young IM (2013) Mean residence
629   time of soil organic carbon in aggregates under contrasting land uses based on radiocarbon
630   measurements. Radiocarbon, **55**, 127-139.

631 Rabbi SMF, Wilson BR, Lockwood PV, Daniel H, Young IM (2015) Aggregate hierarchy and
632   carbon mineralization in two Oxisols of New South Wales, Australia. Soil & Tillage Research,
633   **146**, 193-203.

634 Rillig MC, Muller LAH, Lehmann A (2017) Soil aggregates as massively concurrent evolutionary
635   incubators. The ISME Journal, **11**, 1943–1948

636 Rillig MC, Wright SF, Allen MF, Field CB (1999) Rise in carbon dioxide changes soil structure.
637   Nature, **400**, 628-628.

638 Robinson A, Di HJ, Cameron KC, Podolyan A (2014) Effect of soil aggregate size and
639   dicyandiamide on N_2O emissions and ammonia oxidizer abundance in a grazed pasture soil.
640   Soil Use and Management, **30**, 231-240.

641   Saunois M, Bousquet P, Poulter B *et al*. (2016) The global methane budget 2000–2012. Earth
642   System Science Data, **8**, 697-751.

643   Schutter ME, Dick RP (2002) Microbial community profiles and activities among aggregates of
644   winter fallow and cover-cropped soil. Soil Science Society of America Journal, **66**, 142-153.

645   Schlüter S, Henjes S, Zawallich J, Bergaust L, Horn M, Ippisch O, ... Dörsch P (2018)
646   Denitrification in soil aggregate analogues-effect of aggregate size and oxygen diffusion.
647   Frontiers in Environmental Science, **6**, 17.

648   Seech AG, Beauchamp EG (1988) Denitrification in soil aggregates of different sizes. Soil
649   Science Society of America Journal, **52**, 1616-1621.

650   Sexstone AJ, Parkin TB, and Tiedje JM (1988) Denitrification response to soil wetting in
651   aggregated and unaggregated soil. Soil Biology and Biochemistry, **20**, 767-769.

652   Sexstone AJ, Revsbech NP, Parkin TB, Tiedje JM (1985) Direct measurement of oxygen profiles
653   and denitrification rates in soil aggregates. Soil Science Society of America Journal, **49**, 645-
654   651.

655   Sey BK, Manceur AM, Whalen JK, Gregorich EG, Rochette P (2008) Small-scale heterogeneity
656   in carbon dioxide, nitrous oxide and methane production from aggregates of a cultivated
657   sandy-loam soil. Soil Biology and Biochemistry, **40**, 2468-2473.

658   Shugart HH, Wang B, Fischer R, Ma J, Fang J, Yan X, ... and Armstrong AH (2018)
659   Gap models and their individual-based relatives in the assessment of the consequences of global
660   change. Environmental Research Letters, **13**, 033001.

661   Six J, Bossuyt H, Degryze S, Denef K (2004) A history of research on the link between
662   microaggregates, soil biota, and soil organic matter dynamics. Soil & Tillage Research, **79**, 7-
663   31.

Six J, Carpentier A, van Kessel C, Merckx R, Harris D, Horwath WR, Lüscher A (2001a) Impact of elevated CO_2 on soil organic matter dynamics as related to changes in aggregate turnover and residue quality. Plant and Soil, **234**, 27-36.

Six J, Conant RT, Pau EA, Paustian K (2002) Stabilization mechanisms of soil organic matter: Implications for C-saturation of soils. Plant and Soil, **241**, 155-176.

Six J, Elliott E, Paustian K (2000a) Soil macroaggregate turnover and microaggregate formation: a mechanism for C sequestration under no-tillage agriculture. Soil Biology and Biochemistry, **32**, 2099-2103.

Six J, Elliott E, Paustian K (1999) Aggregate and soil organic matter dynamics under conventional and no-tillage systems. Soil Science Society of America Journal, **63**, 1350-1358.

Six J, Guggenberger G, Paustian K, Haumaier L, Elliott E, Zech W (2001b) Sources and composition of soil organic matter fractions between and within soil aggregates, European Journal of Soil Science, **52**, 607-618.

Six J, Paustian K, Elliott ET, Combrink C (2000b) Soil structure and organic matter I. Distribution of aggregate-size classes and aggregate-associated carbon. Soil Science Society of America Journal, **64**, 681-689.

Smith KA (1980) A model of the extent of anaerobic zones in aggregated soils, and its potential application to estimates of denitrification. Journal of Soil Science, **31**, 263–277.

Smith KA, Ball T, Conen F, Dobbie KE, Massheder J, Rey A (2003) Exchange of greenhouse gases between soil and atmosphere: interactions of soil physical factors and biological processes. European Jouranl of Soil Science, **54**, 779-791.

Stepanov A, Manucharova N, Polyanskaya L (1997) Bacteria producing nitrous oxide in soil aggregates. Eurasian Soil Science, **30**, 863-866.

687 Tiedje, J. M., Sexstone, A. J., Parkin, T. B., Revsbech, N. P. and Shelton, D. R. 1984. Anaerobic

688   processes in soil. - In: Tinsley, J. and Darbyshire, J. F. (eds). Biological processes and soil

689   fertility. Vol II, Martinus Nijhoff/Dr. W. Junk Publishers, The Hague, Netherlands, pp. 197-

690   212.

691 Tisdall JM, Oades JM (1982) Organic matter and water-stable aggregates in soils. Journal of Soil

692   Science, **33**, 141-163.

693 Torn MS, Chabbi A, Crill P (2015) A call for international soil experiment networks for

694   studying, predicting, and managing global change impacts. SOIL, **1**, 575-582.

695 Uchida Y, Clough TJ, Kelliher FM, Sherlock RR (2008) Effects of aggregate size, soil

696   compaction, and bovine urine on N_2O emissions from a pasture soil. Soil Biology

697   Biochemistry, **40**, 924-931.

698 Van Gestel M, Merckx R, Vlassak K (1996) Spatial distribution of microbial biomass in

699   microaggregates of a silty-loam soil and the relation with the resistance of microorganisms to

700   soil drying. Soil Biology Biochemistry, **28**, 503-510.

701 von Fischer JC, Hedin LO (2007) Controls on soil methane fluxes: tests of biophysical

702   mechanisms using stable isotope tracers. Global Biogeochemical Cycle,

703   **21**, GB2007.

704 Wang B, Hou LY, Liu W, Wang ZP (2013a) Non-microbial methane emissions from soils.

705   Atmospheric Environment, **80**, 290-298.

706 Wang B, Lerdau MT, He YL (2017a) Widespread production of non-microbial greenhouse gas

707   production in soils. Global Change Biology, **23**, 4472-4482.

708 Wang B, Shugart HH, Lerdau MT (2017b) An individual-based model of forest volatile

709   organic compound emissions—UVAFME-VOC v1. 0. Ecological Modelling, **350**, 69-78.

710   Wang B, Shugart HH, Lerdau MT (2017c) Senstitivity of global budgets of greenhouse gases to

711   tropospheric ozone pollution mediated by the biosphere. Environmental Research Letters, **12,**

712   084001.

713   Wang Y, Zhang JH, Zhang ZH (2015) Influences of intensive tillage on water-stable

714   aggregate distribution on a steep hillslope. Soil & Tillage Research, **151**, 82-92,.

715   Wang ZP, Han XG, Chang SX, Wang B, Yu Q, Hou LY, Li LH (2013b) Soil organic and

716   inorganic carbon contents under various land uses across a transect of continental steppes in

717   Inner Mongolia. Catena, **109**, 110-117.

718   Williams DB, Carter CB (1996) The transmission electron microscope. In Transmission

719   electron microscopy (pp. 3-17). Springer, Boston, MA.

720   Wright IJ, Reich PB, Westoby, M, Ackerly, DD, Baruch Z, Bongers F, ... Flexas, J

721   (2004). The worldwide leaf economics spectrum. Nature, **428**, 821.

722   Yang, WH, Silver WL (2016) Net soil–atmosphere fluxes mask patterns in gross production and

723   consumption of nitrous oxide and methane in a managed ecosystem. Biogeosciences, **13**,

724   1705-1715.

725   Yang WH, Teh YA, Silver WL (2011) Nitrous oxide production and consumption in a peatland

726   pasture: a test of a field-based ^{15}N-nitrous oxide pool dilution technique. Global Change

727   Biology **17**, 3577-3588.

728   Yavitt JB, Downey DM, Lang GE, Sexstone AJ (1990) Methane consumption in two temperate

729   forest soils. Biogeochemistry, **9**, 39-52.

730   Young IM, Crawford JW (2004) Interactions and self-organization in the soil-microbe

731   complex. Science, **304**, 1634-1637.

732   Young IM, Ritz K (2000) Tillage, habitat space and function of soil microbes. Soil & Tillage

733   Research, **53**, 201-213
734

Table 1 A compilation of research on aggregate reactors in terms of CO_2.

Land use*	Soil texture	Method	Size class (mm)	Moisture‡	Rate vs size‖	Reference
Grassland	Silty loam	Dry†	6.3-2, <2	FC	+	Bimüller et al. 2016
Pasture	Clay	Wet	2-0.25, 0.25-0.053, <0.053	70% FC	=	Rabbi et al. 2015
Crop	Clay	Wet	2-0.25, 0.25-0.053, <0.053	70% FC	=	Rabbi et al. 2015
Forest	Clay	Wet	2-0.25, 0.25-0.053, <0.053	70% FC	=	Rabbi et al. 2015
Crop	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	+	Bandyopadhyay & Lal 2014
Crop (NT)	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	+	Bandyopadhyay & Lal 2014
Forest	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	+	Bandyopadhyay & Lal 2014
NA	Clay loam	Dry	4-2, 2-1, 1-0.5, <0.5	FC	-	Mangalassery et al. 2013
NA	Sandy loam	Dry	4-2, 2-1, 1-0.5, <0.5	FC	+	Mangalassery et al. 2013
Crop	Silt loam	Dry	>6, 6-4, 4-2, 2-1, 1-0.5, 0.5-0.25	60% FC	-	Muñoz et al. 2012
Tropical forest	Peat	Dry	20-8, 8-2, <2	30%&70% FC	+	Kimura et al. 2012
Oil palm	Peat	Dry	20-8, 8-2, <2	30%&70% FC	=	Kimura et al. 2012
Grassland	NA	Dry	4.5-2, <2	60% & 80% FC	+	Diba et al. 2011
Crop (NT/CT)	Sandy loam	Dry	>4, 4-1, <1	80% FC	+	Fernández et al. 2010
Crop	Sandy loam	Dry	6-2, 2-0.25, <0.25	20% - 80% WFPS	-	Sey et al. 2008
Crop	Clay loam	Dry	8-4, 4-2, 2-1, 1-0.5, 0.5-0.25, <0.25	30% GWC	-	Drury et al. 2004
Crop	Silt loam	Dry	5-2, 2-0.25,<0.25	67% FC	+	Schutter & Dick 2002
Crop	Loam	Wet	>1, 1-0.5, 0.5-0.25, <0.25	NA	+	Aoyama et al. 1999
Crop	Loam	Wet	5.6-1.0, 1.0-0.25, 0.25-0.05, <0.05	FC	+	Franzluebbers & Arshad 1997
Crop	Silt loam	Wet	5.6-1.0, 1.0-0.25, 0.25-0.05, <0.05	FC	+	Franzluebbers & Arshad 1997
Crop	Clay loam	Wet	5.6-1.0, 1.0-0.25, 0.25-0.05, <0.05	FC	+	Franzluebbers & Arshad 1997
Crop	Clay	Wet	5.6-1.0, 1.0-0.25, 0.25-0.05, <0.05	FC	+	Franzluebbers & Arshad 1997
Crop	Clay loam	Dry	20-10, 10-5, 5-2, 2-1, 1-0.5, 0.5-0.25, <0.25	FC	-	Seech & Beauchamp 1988
Prairie	Sandy loam	Dry†	8-1, 1 - 0.5, 0.5 - 0.25, 0.25 - 0.1, < 0.1	Field-moist	+	Gupta & Germida 1988
Prairie (CT)	Sandy loam	Dry†	8-1, 1 - 0.5, 0.5 - 0.25, 0.25 - 0.1, < 0.1	Field-moist	+	Gupta & Germida 1988

* The blank denotes information not available. NT and CT denote no tillage and conventional tillage, respectively.

† These studies directly used field-moist soils for dry sieving, which remain being labeled as dry sieving.

‡ Incubation moisture expressed in % field capacity (FC), in water-filled pore space (WFPS), or in gravimetric water content (GWC) based on literature.

‖ Sign '+' generally denotes a positive relationship of CO_2 production rate with aggregate size, '-' negative, while '=' no significant relationship.

For more detailed information of each study, see Table S1,S2 in supplementary information.

735

736

737

Table 2. A compilation of research on aggregate reactors in terms of N_2O.

Land use	Soil texture	Method	Size class (mm)	Moisture	Rate vs size	Reference
Crop	Clay loam	Dry	5.6-4, 4-2, 2-1	Aeration	+	Robinson et al. 2014
Crop	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	+	Bandyopadhyay & Lal 2014
Crop(NT)	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	+	Bandyopadhyay & Lal 2014
Forest	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	+	Bandyopadhyay & Lal 2014
NA	Sandy loam	Dry	4-2, 2-1, 1-0.5, <0.5	FC	=	Mangalassery et al. 2013
NA	Clay loam	Dry	4-2, 2-1, 1-0.5, <0.5	FC	=	Mangalassery et al. 2013
Crop	Silt loam	Dry	>6, 6-4, 4-2, 2-1, 1-0.5, 0.5-0.25	60% FC	-	Muñoz et al. 2012
Oil palm	Peat	Dry	20-8, 8-2, <2	30%&70% FC	+	Kimura et al. 2012
Tropical forest	Peat	Dry	20-8, 8-2, <2	30%&70% FC	+	Kimura et al. 2012
Crop	*NA*	Wet	>2, 2-0.25, 0.25-0.053, <0.053	60% FC	+	Jiang et al. 2011
Grassland	*NA*	Dry	4.5-2, <2	60% & 80% FC	+	Diba et al. 2011
Crop	Sandy loam	Dry	6-2, 2-0.25, <0.25	20% - 80% WFPS	+	Sey et al. 2008
Pasture	Silt loam	Dry	5.6-4, 4-2, 2-1, <1	FC	-	Uchida et al. 2008
Crop	Clay loam	Dry	8-4, 4-2, 2-1, 1-0.5, 0.5-0.25, <0.25	30% GWC	+	Drury et al. 2004
NA	Loamy	Wet	5.0-3.0, 3.0-2.0, 2.0-1.0, 1.0-0.5, 0.5-0.25,<0.25	Dry	+	Manucharova et al. 2001
NA	*NA*	*NA*	10-4.0	*NA*	+	Stepanov et al. 1997
Crop	Silt loam	Wet	>4.7, 4.7-2, 2-1, 1-0.5, 0.5-0.25, 0.25-0.1, <0.1	FC	+	Beauchamp et al. 1990
Crop	Silt loam	Dry	>20, 20-10, 10-5, 5-2, 2-1, 1-0.5, 0.5-0.25, 0.25-0.15,0.15-0.05,<0.05	FC	-	Beauchamp et al. 1990
Crop	Silt loam	Dry	20-10, 10-5, 5-2, 2-1, 1-0.5, 0.5-0.25, <0.25	Saturation	-	Seech & Beauchamp 1988

See Table 1 notes on information listed.

For more detailed information of each study, see Table S1,S3 in supplementary information.

738

739

740

741

742

743

744

Table 3. A compilation of research on aggregate reactors in terms of CH_4.

Land use	Soil texture	Method	Size class (mm)	Moisture	Rate vs size†	Reference
Crop	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	=	Bandyopadhyay & Lal 2014
Crop (NT)	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	=	Bandyopadhyay & Lal 2014
Forest	Silt loam	Wet	>2, 2-0.25, 0.25-0.053, <0.053	FC	=	Bandyopadhyay & Lal 2014
NA	Clay loam	Dry	4-2, 2-1, 1-0.5, <0.5	FC	-	Mangalassery et al. 2013
NA	Sandy loam	Dry	4-2, 2-1, 1-0.5, <0.5	FC	-	Mangalassery et al. 2013
Tropical forest	Peat	Dry	20-8, 8-2, <2	30% & 70% FC	+	Kimura et al. 2012
Oil palm	Peat	Dry	20-8, 8-2, <2	30% & 70% FC	+	Kimura et al. 2012
Crop	Sandy loam	Dry	6-2, 2-0.25, <0.25	20% - 80% WFPS	+/=/-	Sey et al. 2008
Crop	Loam	Dry	<2, 2-4, 4-6, 6-8, 8-10, >10	7-97% FC	+	Jäckel et al. 2001
Forest	Loam	Wet	>2, 2-0.25, 0.25-0.053, < 0.053	Dry	+	Wang et al. 2013a‡

* See Table 1 notes on information listed;

† Pattern '+' denotes larger aggregates are CH_4 producers and smaller aggregates are consumers (or less production than larger aggregates); '-' denotes larger aggregates have less production than smaller aggregates; and '=' denotes no significant pattern is observed;

‡ The only study on aggregate-scale non-microbial CH_4.
For more detailed information of each study, see Table S1,S4 in supplementary information.

745

746

747

748

749

750

751

752

753

Fig. 1 Schematic of soils as a system of aggregate reactors of different sizes. At the profile level, soils act as a source of CO_2, either a source or a sink of CH_4 (denoted by the upward and downward arrow, respectively), and a source of N_2O. At fine scales, soil consists of aggregate reactors of differing sizes. Each individual aggregate reactor can be described by physical (e.g., pore size, diffusion coefficient, and aggregate size), chemical (e.g., concentration of O_2, H_2O, dissolved organic carbon—DOC, substrates), and biological traits (turnover rate, microbial community composition and dynamics). Top-down experiment refers to studying these properties by 'digging' into soils. Aggregate reactivity depends on aggregate size (denoted by the irregular circles with differing colors) and bulk soil properties including both abiotic and biotic factors, as well as coarser scale anthropogenic disturbances. Different widths of the red arrows denote the reactor size-induced variations in GHGs. Soil systems composed of aggregates of different sizes are dynamically changing because of aggregate turnover (or aggregate stability), which is not illustrated here. Bottom-up modelling refers to building models based on aggregate reactor that can represent soil system composition and dynamics and simulate soil profile GHG exchange as an emergent process.

769

770

771

772

773

774

775