

1 **Aggregates reduce transport distance of soil organic carbon: are** 2 **our balances correct?**

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8 **Abstract**

9 The effect of soil erosion on global carbon cycling, especially as a source or sink for
10 greenhouse gases, has been the subject of intense debate. The controversy arises mostly
11 from the lack of information on the fate of eroded soil organic carbon (SOC) whilst in-transit
12 from the site of erosion to the site of longer-term deposition. Solving this controversy requires
13 an improved understanding of the transport distance of eroded SOC, which is principally
14 related to the settling velocity of sediment fractions that carry the eroded SOC. Although
15 settling velocity has already been included in some erosion models, it is often based on
16 mineral particle size distribution. For aggregated soils, settling velocities are affected by their
17 actual aggregate size rather than by mineral particle size distribution. Aggregate stability is,
18 in turn, strongly influenced by SOC. In order to identify the effect of aggregation of source
19 soil on the transport distance of eroded SOC, and its susceptibility to mineralization after
20 transport and temporary deposition, a rainfall simulation was carried out on a silty loam. Both
21 the eroded sediments and undisturbed soils were fractionated into six different size classes
22 using a settling tube apparatus according to their settling velocities: > 250, 125 to 250, 63 to
23 125, 32 to 63, 20 to 32 and < 20 μm . Weight, SOC content and instantaneous respiration
24 rates were measured for each of the six class fractions. Our results indicate that: 1) 41% of
25 the eroded SOC was transported with coarse aggregates that would be likely re-deposited
26 down eroding hillslopes, rather than with fine particles likely transferred to water courses; 2)
27 erosion was prone to accelerate the mineralization of eroded SOC, and thus might contribute
28 more CO₂ to the atmosphere than current estimates which often ignore potential effects of

29 aggregation; 3) preferential deposition of SOC-rich coarse aggregates potentially causes an
30 increase of SOC remaining in the colluvial system and a reduction of SOC flux to the alluvial
31 or aquatic system. These findings identify a potential error of overestimating net erosion-
32 induced carbon sink effects, and thus add an additional factor to consider when improving
33 our current understanding of SOC erosion and deposition on hillslopes.

34

35 *Key words: eroded organic carbon, instantaneous respiration rate, transport distance, carbon*
36 *source*

37

38 **1. Introduction**

39 The net effect of soil erosion as a source or sink of CO₂ in the global carbon cycle
40 has been the subject of intense debate (Lal, 2003; van Oost et al., 2007; Quinton et
41 al., 2010; Dlugoß et al., 2012; Doetterl et al., 2012). On one hand, erosion exposes
42 the previously incorporated soil organic carbon (SOC), which may accelerate the
43 mineralization of eroded SOC (Jacinthe et al., 2002, 2004; Kuhn, 2007; Mora et al.,
44 2007; Lal and Pimentel, 2008). On the other hand, deposition limits the
45 decomposition of SOC upon burial, while inputs of decomposing plant material on the
46 surface of eroding sites partially replaces the lost SOC (Harden et al., 1999; van Oost
47 et al., 2007; Wang et al., 2010). So far, effects of erosion on CO₂ emissions have
48 mostly been assessed by comparing SOC stocks at the assumed site of erosion and
49 the site of colluvial deposition (Stallard, 1998; Berhe, 2012; van Hemelryck et al.,
50 2011; Nadeu et al., 2012; van Oost et al., 2012). One underlying assumption
51 associated with this approach is that the redistribution of eroded SOC across
52 landscapes is non-selective. However, several recent publications showed (at least)
53 temporary enrichment of SOC in sediment, as well as preferential deposition of

54 aggregates with size distribution and SOC content that differ from original soils
55 (Schiettecatte et al., 2008; Kuhn et al., 2009; Hu et al., 2013a; Kuhn, 2013). As a
56 consequence, carbon balances drawn only from the SOC stocks on sites of erosion
57 and colluvial deposition may not adequately consider the potential SOC re-deposition
58 into the terrestrial system.

59 Regardless of selective or non-selective erosion, sediment fractions are all likely to
60 experience preferential deposition. Therefore, SOC redistribution, either after
61 selective or non-selective erosion, is strongly depending on the transport distance of
62 eroded aggregates. This is thus related to the respective settling velocities of
63 sediment fractions carrying the eroded SOC (Dietrich, 1982; Kinnell, 2001, 2005).
64 Although settling velocity has already been included in some erosion models, it is
65 often based on mineral particle size distribution (Morgan et al., 1998; Beuselinck et
66 al., 1999; Flanagan and Nearing, 2000; van Oost et al., 2004; Aksoy and Kavvas,
67 2005). But for aggregated soils, settling velocities are affected by the actual size,
68 irregular shape, porosity of soil fractions and their incorporation with light-dense SOC
69 (Kinnell and McLachlan, 1988; Loch, 2001; Hu et al., 2013b). In addition, the upper
70 limits of the mineral particle size classes used in current erosion models are often
71 smaller than the actual aggregate sizes. For instance, the largest class in van Oost et
72 al. (2004) is only 90 μm , whereas up to 250 μm for rill erosion in Morgan et al. (1998).
73 Such limits may also skew the estimation on settling velocity of eroded sediment.
74 Hence, the mineral particle size classes, no matter how efficiently applicable in
75 erosion models, are not the decisive factor that determines the actual settling
76 behaviour or movement of aggregates. Aggregation of original soil potentially
77 increases the settling velocities of soil particles, and thereby likely reduces their

78 transport distances after erosion (Hu et al., 2013b). This, in theory, would also reduce
79 the transport distance of eroded SOC incorporated into soil aggregates.

80 The effect of aggregation of source soil may also affect the movement of SOC down
81 eroding hillslopes. Aggregation is related to SOC content, and SOC is often
82 increased in both macro- and micro-aggregates (Tisdall and Oades, 1982;
83 Cambardella and Elliott, 1994). The quality and stabilizing mechanisms of SOC in the
84 soil matrix also vary with different aggregate conditions. For instance, physically-
85 stabilized SOC within macro- and micro-aggregates is protected by forming physical
86 barriers between microbes and enzymes and their substrates, and thus very
87 susceptible to mineralization after aggregates break-up (Six et al., 2002). Chemically-
88 stabilized SOC results from the chemical or physicochemical binding between SOC
89 and soil minerals (i.e., clay and silt particles). Such stabilization is also likely to be
90 disturbed by aggregates break-up, as often occurs during erosion and transport
91 (Starr et al., 2000; Lal and Pimentel, 2008; van Hemelryck et al., 2010).
92 Biochemically-stabilized SOC is resulted from the inherent or acquired biochemical
93 resistance to decomposition. Aggregates break-up might also affect the
94 biodegradability of the SOC or the exposure to hydrolyzation. Therefore, erosion,
95 either detaching aggregates from the soil matrix or disintegrating larger aggregates,
96 may have diverse impacts on mineralization of eroded SOC (van Hemelryck et al.,
97 2010; Fiener et al., 2012).

98 In this study, we aim to conduct an initial test of the theoretical deductions made
99 above by fractionating eroded loess sediments, generated during a laboratory rainfall
100 simulation, according to their settling velocities, and then measure their SOC content
101 and instantaneous respiration rates.

102

103 **2. Materials and Methods**

104 **2.1. Soil sampling and preparation**

105 A silty loam from the conventionally managed Bäumlhof Farm in Möhlin (47°33' N,
106 7°50' E), near Basel in northwest Switzerland, was used in this study. The soil
107 supports a maize-wheat-grass rotation. A-horizon material (top 20 cm) was sampled
108 from a gentle shoulder slope (< 5%) in March 2012. Previous research on the same
109 silty loam showed that aggregation increased the settling velocity of original soil
110 fractions, particularly the medium sized fractions, in comparison with that expected
111 based on the texture of the original soil (Hu et al., 2013b). The mineral particle
112 specific SOC distribution, average SOC content (LECO RC 612 at 550°C), and
113 aggregate stability of original soil (method adapted from Nimmo and Perkins, 2002),
114 are shown in Table 1. The mineral particle size distribution was fractionated by wet-
115 sieving, after dispersion by ultrasound using a Sonifier Model 250 from Branson, USA.
116 The energy dissipated in the water/soil suspension was $60 \text{ J}\cdot\text{ml}^{-1}$ (i.e. Energy =
117 output power $70 \text{ W} \times$ time 85 s / suspension volume 100 ml). The SOC mass
118 proportions across mineral particle size classes were calculated only from average
119 values of individual weight and SOC content. Although the ultrasound energy used in
120 Hu et al. (2013b) was not enough to thoroughly disperse the original soil into real
121 mineral particles (Kaiser et al., 2012), such extent of dispersion was notable enough
122 to demonstrate the potential under-estimation of applying mineral particle size
123 distribution to predict the settling velocity of eroded SOC. Hence, it is speculated that
124 similar increasing effects would also occur to sediment fractions, and thus make the
125 silty loam suitable to investigate the potential effects of aggregation of original soil on
126 the transport distances of differently sized sediment fractions. While this study
127 investigated only one soil, similar loess soils cover about 10% (ca., 14.9 million km^2)

128 of the global land area (Sartori, 2000). This study was thus considered relevant as it
129 generally reflects the erodible nature of similar loess soils under similar management
130 regimes. In the future, more experiments with soils of different aggregation and
131 various SOC contents have to be carried out to expand our knowledge on the effects
132 of aggregation to a wider range of soils.

133 TABLE 1

134 **2.2. Experimental set-up**

135 *2.2.1. Rainfall simulation*

136 The experiments consisted of three separate components: 1) rainfall simulation to
137 sufficiently destroy aggregates, so as to ensure that the eroded sediments were less
138 likely to experience further breakdown during the subsequent settling velocity
139 measurements; 2) fractionation of the eroded sediments by a settling tube apparatus
140 into six settling velocity classes; and 3) measurements of the instantaneous
141 respiration rates of each settling class. The experiments were repeated three times in
142 order to generate reliable erosion and respiration data.

143 Soils (0 - 5 cm depth) were placed in a 150 × 80 cm flume, which was pitched at a 15%
144 gradient (Figure 1a). Preliminary tests revealed that a flume of this size could
145 generate sufficient runoff to initiate non-selective erosion on this particular silt loam.
146 The soil was sieved into aggregates of a diameter less than 10 mm and over-sized
147 clods were excluded in order to reduce variations in surface roughness, both within
148 the flume and between replicates. Levelling the surface also ensured that large
149 roughness elements, in particular depressions, did not inhibit movement of
150 aggregates across the flume and thereby prevent selective deposition on the soil
151 surface. To assist drainage, the base of the flume was perforated and covered with a

152 fine cloth and a layer of sand (c.a., 5 cm). A FullJet nozzle of ¼ HH14WSQ, installed
153 1.8 m above the soil surface, was used to generate rainfall. Soil of each replicate was
154 then subjected to simulated rainfall at an intensity of 55 mm·h⁻¹ for 3 h. The kinetic
155 energy of the raindrops, detected by a Joss-Waldvogel-Disdrometer, was on average
156 200 J·m⁻²·h⁻¹.

157 Natural precipitation events of 55 mm·h⁻¹ are unlikely in the Möhlin region, where
158 precipitation intensity is mostly less than 35 mm·h⁻¹ (return period of 0.33 years,
159 MeteoSwiss, 2013). Increased intensity is often considered necessary to compensate
160 for the deficiency of kinetic energy associated with simulated rainfalls in order to
161 recreate conditions that were as comparable as possible with natural rainfalls
162 (Dunkerley, 2008; Iserloh et al., 2012, 2013). A previous study had shown that full
163 crust formation on the Möhlin silty loam (aggregates < 8 mm) requires a cumulative
164 kinetic energy of about 340 J·m⁻² (Hu et al., 2013a). This corresponds to natural
165 precipitation of 35 mm·h⁻¹ for 30 min (Iserloh et al., 2012). Therefore, a simulated
166 rainfall of 55 mm·h⁻¹ lasting for 3 h with a cumulative kinetic energy about 600 J·m⁻²
167 was chosen in this study to make sure that the aggregates (coarser than those in Hu
168 et al., 2013a) would experience full crust formation to equilibrium conditions, i.e.
169 resistance of crust against erosion equals the erosive force of raindrops and runoff.

170

FIGURE 1

171 Tap water, with an electric conductivity of 2220 µs·cm⁻¹, which is five times higher
172 than natural rainwater in Basel, was used during each rainfall simulation. In general,
173 the increased electric conductivity associated with tap water increases soil dispersion
174 during tests using simulated rainfall (Borselli et al., 2001). Despite this, however,
175 comparative aggregate stability tests showed only a 7% difference between rainwater
176 sampled in Basel and ordinary tap water; thus making it acceptable.

177 2.2.2. *Sediment collection and fractionation by a settling tube apparatus*

178 During each rainfall event, runoff and sediment were continuously collected over 30
179 min intervals. A 1.8m settling tube (Figure 1b) was used to fractionate the eroded
180 sediment fractions according to their respective settling velocities. The settling tube
181 apparatus consists of four components (Figure 1a): the settling tube, through which
182 the soil sample settles; the injection device, by which the soil sample is introduced
183 into the tube; the turntable, within which the fractionated subsamples are collected;
184 and the control panel, which allows an operator to control the rotational speed and
185 resting/moving intervals of the turntable. Details about the settling tube apparatus
186 were described in Hu et al. (2013b). The injection device within this particular settling
187 tube has a volume of 80 cm³. As this limits the amount of sediment used during each
188 test, only sediment collected during the first 10 min of each 30 min interval was used
189 to determine settling velocities. In total, there were six sediment collection intervals
190 over the 3 h rainfall events, and a settling fractionation test was carried for each of
191 the six sediment collection intervals. Prior to being subjected to settling fractionation,
192 the eroded sediment was allowed to settle for 1 h in collection beakers (height of 20
193 cm). Measurements confirmed that > 95% of the total mass settled after this pre-
194 treatment. The supernatant and remaining suspended sediment (corresponding to
195 EQS < 8 μm) was then decanted off and added to the < 20 μm fraction remaining in
196 suspension in the settling tube (described as following).

197 Six particle size classes, based on the concept of Equivalent Quartz Size (EQS)
198 described in Hu et al. (2013b), were selected according to their likely transport
199 distances after erosion (Table 2). EQS represents the diameter of a nominal
200 spherical quartz particle that would fall with the same velocity as the aggregated
201 particle for which fall velocity is measured (Loch, 2001). The six EQS classes were

202 converted to six settling velocities and corresponding settling times using Stokes'
203 Law (Hu et al., 2013b). The use of Stokes' Law to convert EQS into settling velocity is,
204 in the strictest sense, limited to particles < 70 μm (Rubey, 1933). From the
205 perspective of terrestrial and aquatic systems, however, sediment fractions coarser
206 than 63 μm are considered as one group that is likely to be re-deposited along
207 hillslopes. Therefore, the potential error when using Stokes' Law to calculate the
208 settling velocities of fractions of all sizes is considered acceptable. For soils
209 dominated by larger mineral grains, different relationships should be used (Ferguson
210 and Church, 2004; Wu and Wang, 2006). In addition, fine suspended fractions are
211 considered as one group exported out of the terrestrial system. Hence, with the
212 current settling tube (length of 1.8 m), any fractions finer than 20 μm (settling time
213 longer than 1.5 h) were not further fractionated to save time. After fractionation, the
214 six EQS classes were air-dried for 72 h in a dark environment at ambient temperature
215 (20°C). Despite the possibility of biasing the mineralization SOC potential through the
216 process of air-drying, as the first step to unwrap the complex effects of aggregation
217 onto SOC erosion, transport, and deposition, our aim was to produce quasi-natural
218 sediments, i.e., subjecting to a single rainfall event, successively re-deposited after
219 increasing transport distances and immediately dried afterwards. Further effects of
220 multiple rainfall events, other soil moisture conditions (e.g., wet sediments) and long-
221 term incubation will be investigated in future research, once the role of aggregation
222 on eroded SOC has been studied.

223 TABLE 2

224 *2.2.3. Instantaneous respiration rate measurement*

225 Instantaneous respiration rates were measured, based on the method described in
226 Robertson et al. (1999) and Zibilske, (1994). In brief, two grams (dry weight) of each

227 EQS size fraction were placed into a 30 ml vial and re-wetted using distilled water in
228 order to obtain a gravimetric moisture content equivalent to ca. 60%. Preliminary
229 tests revealed that the gravimetric moisture of 60% represented a proper
230 intermediate moisture level for sediment fractions of various surface areas, and thus
231 exerted comparable effects on soil respiration rates (Xu et al., 2004; Bremenfeld et
232 al., 2013). The re-wetting was done on the previous day before the respiration
233 measurements. This way, the initial CO₂ pulses of rewetted soils should be largely
234 excluded (Orchard and Cook, 1983). Even if there were any CO₂ pulses induced by
235 rewetting, this exactly mimics the natural processes, where dry sediments deposited
236 from previous rainfall events, experience a second time of erosion and transport
237 processes. All the re-wetted fractions were then incubated over night at 25°C (vials
238 open). Two grams of original undisturbed soil were also prepared in the same way
239 and used to generate reference measurements. Prior to soil respiration
240 measurements, all vials were sealed using rubber stoppers. Gas from the headspace
241 of each sealed vial was extracted by a 1 cc syringe at the beginning and end of the 1
242 h sampling period. Differences in CO₂ concentrations between these two
243 measurements, as measured on a SRI8610C Gas Chromatograph, were used to
244 calculate the instantaneous respiration rate.

245 *2.2.4. Laboratory measurements and data analyses*

246 Soil erosion rates for each 30 min interval were estimated by the mass of sediment
247 samples both from the beginning 10 min (sum of the six EQS classes) and the late 20
248 min (not fractionated by the settling tube). Runoff samples collected during the late
249 20 min of each 30 min interval were allowed to settle for more than 48 h. The
250 supernatant was then decanted off and the sediment was dried at 40°C and weighed.
251 The SOC content of all the samples was measured by a LECO RC 612 at 550°C.

252 Data analysis was carried out using Microsoft Excel 2010 and R studio software
253 packages (R version 2.15.1).

254

255 **3. Results**

256 Runoff began after 20 min. of rainfall and attained steady state conditions equivalent
257 to $18 \text{ mm}\cdot\text{h}^{-1}$ after 120 min. Sediment discharge rates followed a similar pattern and
258 reached steady state of $168.7 \text{ g}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$. Detailed erosional responses are listed in
259 Table 3. During the simulated rainfall events, the sediments were seen to move
260 continuously with runoff, and no evident selective deposition was observed on the
261 soil surface.

262

TABLE 3

263 The fraction mass and SOC in the six EQS classes of sediment are presented in
264 Figures 2 and 3. Preliminary data analysis had shown that while the absolute
265 sediment mass increased during the simulated rainfall events, the proportional
266 composition of six EQS classes in each sediment collection interval did not differ
267 significantly between samples (ANOVA, single factor, $P > 0.05$, $n = 18$). Hence, only
268 proportional values are presented, and each EQS class could be considered to have
269 18 replicates (6 sediment collection intervals during each of the 3 rainfall events).
270 Nevertheless, the distribution of fraction mass and SOC content considerably differed
271 across six EQS classes: about 61% of the sediment fractions were in EQS of 32 to
272 $63 \mu\text{m}$ and 63 to $125 \mu\text{m}$, containing about 65% of the SOC. This SOC distribution in
273 six EQS classes of sediment also contrasts against the association of SOC with
274 mineral particles in the original soil (Table 1).

275

FIGURE 2

276

FIGURE 3

277 The instantaneous respiration rates from EQS classes of 32 to 63 μm and 63 to 125
278 μm were on average lower than that from other fractions (Figure 4a). However, after
279 multiplying the respiration rate from each class with its fraction mass (Figure 2), EQS
280 classes of 32 to 63 μm and 63 to 125 μm on average released even more amount of
281 CO_2 than all other finer or coarser classes did (Figure 4b). The instantaneous
282 respiration rates per gram of SOC also differ among different EQS classes. In EQS
283 classes $< 20 \mu\text{m}$ and 20 to 32 μm , the instantaneous respiration rates per gram of
284 SOC were lower than that in the original soil (Figure 5). In contrast, all the other four
285 EQS classes ($> 32 \mu\text{m}$) had higher instantaneous respiration rates per gram of SOC
286 than the original soil (Figure 5). We attribute the increased respiration rates per gram
287 of SOC in EQS classes $> 32 \mu\text{m}$ to the detachment and transport of eroded soils,
288 during which time the structural aggregates were broken down, thereby exposing the
289 previously protected SOC to microbial processes (Six et al., 2002; Lal and Pimentel,
290 2008; van Hemelryck et al., 2010).

291

FIGURE 4

292

FIGURE 5

293

294 **4. Discussion**

295 **4.1. Likely fate of eroded SOC in the terrestrial and aquatic system**

296 Fractionation of eroded sediment by settling velocity shows that aggregation of
297 source soil has a clear potential to affect the movement of sediment fractions and
298 thus the fate of the associated SOC after erosion. According to the conceptual model

299 developed by Starr et al. (2000) (Figure 6), the six EQS classes can be further
300 grouped into three separate groups, each with a different likely fate: EQS < 20 μm
301 would be likely to remain suspended in runoff and hence, transferred to rivers, and all
302 EQS > 63 μm would be re-deposited along eroding hillslopes (Table 2). The
303 intermediate EQS of 20 to 32 μm and 32 to 63 μm can have either fate, depending on
304 localised flow hydraulics. In accordance with this model, approximately 41% of the
305 eroded SOC from the silt loam used in this study would be re-deposited along
306 eroding hillslopes (Figure 7b). This proportion strongly contrasts against the
307 approximately 11% SOC mass associated with coarse mineral particles > 63 μm in
308 the original soil (Table 1), and is also contrary to the high SOC content (24.3 $\text{mg}\cdot\text{g}^{-1}$)
309 in sediment fraction of EQS < 20 μm (Figure 3a). These results support our
310 theoretical deduction that aggregation of source soil reduces the likely transport
311 distance of eroded SOC. This would then decrease the likelihood of eroded SOC
312 being transferred from eroding hillslopes to aquatic systems, but increase the amount
313 of eroded SOC being re-deposited into terrestrial systems. These findings are
314 consistent with those reported by Hu et al. (2013b), in which 79% of the eroded SOC
315 mass in a silty loam was associated with mineral particles of size < 32 μm , whereas
316 73% of the SOC mass was actually contained in aggregates of EQS > 63 μm . The
317 distinct SOC distribution across aggregate size classes also agrees with the field
318 investigation by Polyakov and Lal (2008), where the coarse aggregates (1 to 0.5 mm)
319 fractionated by wet-sieving, contained up to 4.5 times more SOC than the finest
320 fraction (< 0.05 mm). More experiments are required to describe the effects of
321 different aggregation degrees and SOC contents.

322 FIGURE 6

323 FIGURE 7

324 4.2. Erosion as a source of CO₂ flux

325 The effect of aggregation on the likely fate of SOC may also cast new light on
326 understanding the effect of soil erosion on global carbon cycling. Based on the EQS
327 specific SOC content (Figure 3), the potential SOC stock of a nominal 25 cm layer of
328 topsoil on the foot-slope of a colluvial depositional site, assumingly composed of
329 aggregates EQS > 63 μm, would be 5.1 kg·m⁻² on average (Table 4). The potential
330 SOC stock from the same 25 cm layer of original soil would be only 4.5 kg·m⁻², or
331 15.5% lower than that on the foot-slope of a colluvial depositional site. Such a large
332 difference implies that a combined model approach (Equation 1), integrating the
333 effects of aggregation on the likely fate of SOC, is demanded to adequately
334 distinguish the proportion of SOC likely re-deposited along hillslopes from the portion
335 potentially transferred to aquatic systems.

$$336 S_d \times C_d = S_e \times C_e - S_a \times C_a - C_{min} \quad (\text{Equation 1})$$

337 Where, S_d : mass of sediment likely to be re-deposited along hillslopes ; C_d : carbon
338 content of sediment likely to be re-deposited along hillslopes ; S_e : mass of eroded soil;
339 C_e : carbon content of eroded soil; S_a : mass of sediment potentially to be transferred
340 to aquatic systems; C_a : carbon content of sediment potentially to be transferred to
341 aquatic systems; C_{min} : carbon mineralized during transport.

342 TABLE 4

343 In previous reports, C_{min} was considered to be a minor constituent in the overall
344 carbon budget, and C_d was often assumed equivalent to the average of C_e (Stallard,
345 1998; Harden et al., 1999; Berhe et al., 2007; van Oost et al., 2007; Quinton et al.,
346 2010). However, if presuming that C_e equals C_d observed in topsoil of colluvial
347 depositional sites (e.g., in van Oost et al., 2012), then the C_d would lead to an

348 overestimation of total SOC loss from eroding sites, because C_d is likely enriched by
349 SOC-rich aggregates compared to C_e due to preferential deposition. Conversely,
350 assuming C_d corresponds to C_e observed in topsoil at eroding sites (e.g., in Dlugoß et
351 al., 2012), would neglect the potential enrichment of SOC in sediment fractions
352 preferentially deposited on hillslopes. This would thus lead to an underestimation of
353 C_{min} during transport. In both cases, SOC transferred to aquatic systems would be
354 overestimated. The observed enrichment of SOC by 15.5% in sediment fractions
355 composed only of EQS > 63 μm , indicates that the potential error of above-described
356 estimates could be considerable. A 15.5% SOC enrichment of sediment re-deposited
357 in the terrestrial system corresponds to the proportion of eroded SOC estimated to be
358 deposited in permanent sinks (e.g., 0.12 Pg of SOC eroded per year by van Oost et
359 al. 2007). While the effects of aggregation on SOC redistribution and subsequent fate
360 cannot be assessed based on one experiment, most sediment is transported in form
361 of aggregates (Walling, 1988; Walling and Webb, 1990). Ignoring the effect of
362 aggregation on erosion and redistribution of SOC, therefore, bears the risk of
363 overestimating the erosion-induced carbon sink effect. As a consequence, the
364 behavior of aggregated sediment requires a reconsideration of existing approaches
365 of sediment behavior in erosion models. Further study of different soil types, their
366 aggregation and aggregate breakdown while moving through landscapes of varying
367 topography during rainfall events of different intensity, frequency and duration, is
368 required to assess the relevance of aggregation for SOC movement and fate
369 identified in this study.

370 The risk of falsely estimating SOC losses during transport is further exacerbated by
371 the observed instantaneous respiration rates. The instantaneous respiration rates
372 probably merely represent a spike of SOC mineralization after erosion, and therefore,

373 should not be extrapolated over longer periods of time. However, the 41% proportion
374 of eroded SOC, which would likely be re-deposited along hillslopes, generated 53%
375 of the entire instantaneous respiration (Figure 7c). This implies that the immediately
376 deposited SOC is more susceptible to mineralization than both the mass of coarse
377 sediment fractions and their SOC content would suggest. These findings are
378 consistent with those observed by van Hemelryck et al. (2010), who reported that a
379 significant fraction of SOC eroded from initially dry soil aggregates is mineralized
380 after deposition. As a consequence, the preferentially deposited SOC could
381 potentially generate a further error in the carbon source-sink balance. Such error
382 would be particularly significant, when repeated erosion and deposition processes
383 along hillslopes cause further disintegration of large aggregates (Kuhn et al., 2003;
384 van Hemelryck et al., 2010). This would thereby result in additional SOC exposure
385 and mineralization (Jacinthe et al., 2002; Six et al., 2002). Overall, as a result of
386 preferential deposition of SOC-enriched sediment fractions and enhanced
387 mineralization during transport, the carbon losses during transport, so far assumed to
388 be small (van Oost et al., 2007; Quinton et al., 2010), would actually be
389 underestimated.

390

391 **5. Conclusion**

392 This study aimed to identify the effects of aggregation of source soil on the likely
393 transport distance of eroded SOC and its susceptibility to mineralization after single-
394 event transport and deposition. Our data show that 41% of the eroded SOC from a
395 silty loam was incorporated into aggregates of EQS > 63 μm , and hence would likely
396 be re-deposited into the terrestrial system rather than being transferred to the aquatic
397 system. This proportion is much greater than the approximately 11% SOC mass

398 associated with coarse mineral particles > 63 μm in the original soil (Table 1), and the
399 high SOC content (24.3 $\text{mg}\cdot\text{g}^{-1}$) in sediment fraction of EQS < 20 μm would suggest.
400 Respiration rates from sediment fractions of EQS > 63 μm also increased
401 immediately after erosion and deposition. Both results indicate that aggregation of
402 source soil and preferential deposition of SOC-rich coarse sediment fractions may
403 skew the re-deposition of eroded SOC towards the terrestrial system, rather than
404 further transfer to the alluvial or aquatic system. Consequently, a risk of
405 overestimating lateral SOC transfer exists when mineral grain size rather than actual
406 size of aggregated sediment is applied in erosion models. Our very limited data
407 indicates that this error could be potentially within the same range as the current
408 estimate of annual net erosion-induced carbon sink rate.

409 While based on a laboratory experiment and thus with very limited applicability to real
410 landscapes, the potential effects of aggregation of source soil on reducing the
411 transport distance of eroded SOC appear to be considerable. This illustrates the
412 need to integrate the effect of aggregation of source soil on SOC transport distance
413 into soil erosion models (e.g., as a soil erodibility parameter), in order to adequately
414 distinguish SOC likely re-deposited in the terrestrial system from the portion
415 potentially transferred to aquatic systems, and further assess the implications to the
416 global carbon cycle. Further research should, therefore, focus on the effects of
417 preferential deposition of eroded aggregates and the fate of SOC in these
418 aggregates whilst in-transit and during multiple rainfall events. More simulations as
419 well as field experiments are also needed to examine the effects of various transport
420 processes (such as slope length, slope gradients, field barriers) onto the mechanism
421 of aggregate breakdown and aggregate specific SOC distribution. The effects of
422 varying rainfall characteristics, crust formation, soil management and topography

423 (e.g., Wang et al., 2008; Hu et al., 2013a) onto SOC transport should also be
424 investigated.

425

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435

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- 587

588 **Table 1** Mineral particle size distribution, soil organic carbon (SOC) distribution
 589 across mineral particle size, average SOC of bulk soil, and the percentage of stable
 590 aggregates greater than 250 μm in the silty loam used in this study.

	Mineral particle size (μm)					SOC of original soil (g kg^{-1})	Aggregates greater than 250 μm (%)
	< 32	32-63	63-125	125-250	> 250		
Weight (%)	62.0 ± 0.3	29.1 ± 0.4	6.6 ± 0.3	1.2 ± 0.1	1.1 ± 0.1		
SOC (g kg^{-1})	13.7 ± 0.7	3.0 ± 0.3	8.9 ± 2.6	21.9 $\pm 0.8^a$	26.4 $\pm 1.3^a$	10.8 ± 0.4	67.2 ± 6.9
SOC mass proportion (%)	80.8	8.3	5.6	2.5	2.8		

591 NOTE: a. It might be over-estimated due to the mixture of minute amount of residue or straw, which was previously incorporated
 592 into the aggregates but then released by dispersion and blended with coarse particles.
 593 Lower case numbers indicate the range of minimum and maximum values ($n = 3$).
 594
 595

596 **Table 2** Six settling velocities based on the Equivalent Quartz Size (EQS) classes,
 597 and the likely fate of eroded fractions based on the conceptual model developed by
 598 Starr et al. (2000).

EQS (μm)	Settling velocity ($\text{m}\cdot\text{s}^{-1}$)	Likely fate
< 20	Suspension	Likely transferred to rivers
20 - 32	$3.3 \times 10^{-4} - 1.0 \times 10^{-3}$	Possibly transferred to rivers
32 - 63	$1.0 \times 10^{-3} - 3.0 \times 10^{-3}$	
63 - 125	$3.0 \times 10^{-3} - 1.5 \times 10^{-2}$	Deposited along eroding hillslopes
125 - 250	$1.5 \times 10^{-2} - 4.5 \times 10^{-2}$	
> 250	$> 4.5 \times 10^{-2}$	

599

600 **Table 3** Summary of the erosional responses of Möhlin soil over 180 min of rainfall
 601 time. Subscripted numbers indicate the minimum and maximum range of the
 602 parameters ($n = 3$).

Steady state (after 120 min)			Total runoff (kg)	Runoff coefficient (%)	Total sediment yield (g)
Runoff rate (mm·h ⁻¹)	Sediment discharge rate (g·m ⁻² ·h ⁻¹)	Sediment concentration (g·L ⁻¹)			
18.0 ±0.9	168.7 ±14.4	9.4 ±0.1	40.7 ±3.1	20.6 ±1.6	475.8 ±74.6

603

604 **Table 4** Comparison between soil organic carbon (SOC) stock in top layer of 25 cm
 605 from a temporary depositional site which is theoretically composed of only three
 606 Equivalent Quartz Size (EQS) classes, and SOC stock of average original soil in the
 607 same top layer of 25 cm, as often applied in previous literature.

	EQS	SOC content (mg g ⁻¹)	SOC stock (kg m ⁻²) ^a	Differences (%) ^b	Average SOC stock (kg m ⁻²)	Average differences (%) ^b
Re-distributed fractions	> 250	12.2	5.0	-11.1	5.1	-15.5
	125 - 250	15.7	6.5	-44.4		
	63 - 125	9.6	4.0	+11.1		
Original soil	NA	10.8	4.5	NA	4.5	NA

608 NOTE: a. Accurate bulk densities for sediment fractions of different aggregate sizes are not available, so only particle density
 609 1.65 g cm⁻³ is applied here to form a preliminary comparison.

610 b. Minus (-) means underestimation compared to the original soil; Plus (+) means overestimation compared to the
 611 original soil.
 612

613 **Figure 1** The rainfall simulation flume (a), and the settling tube apparatus (b). The
614 settling tube apparatus consists of four components: the settling tube, through which
615 the soil sample settles; the injection device, by which the soil sample is introduced
616 into the tube; the turntable, within which the fractionated subsamples are collected;
617 and the control panel, which allows an operator to control the rotational speed and
618 rest intervals of the turntable (operations see Hu et al., 2013b)).

619 **Figure 2** The weight distribution of different Equivalent Quartz Size (EQS) classes of
620 the sediment. Colors of the boxes correspond to the likely fate of each fraction after
621 erosion. See section 4.1 and Figure 6 for definitions and explanation of the three
622 manners of likely fate. Bars in the boxes represent median values. Whiskers indicate
623 the lowest datum within 1.5 interquartile range of the lower quartile, and the highest
624 datum within 1.5 interquartile range of the upper quartile ($n = 18$).

625 **Figure 3** The distribution of soil organic carbon content (SOC) (a), and soil organic
626 carbon (SOC) mass (b) in different Equivalent Quartz Size (EQS) classes of the
627 sediment. The bold and dashed lines in (a) denote the average and standard
628 deviation of soil organic carbon (SOC) of the original soil. Colors of the boxes
629 correspond to the likely fate of each fraction after erosion. See section 4.1 and Figure
630 6 for definitions and explanation of the three manners of likely fate. Bars in the boxes
631 represent median values. Whiskers indicate the lowest datum within 1.5 interquartile
632 range of the lower quartile, and the highest datum within 1.5 interquartile range of the
633 upper quartile ($n = 18$).

634 **Figure 4** The distribution of instantaneous respiration rate (a); and potential CO₂
635 emission (b) in different Equivalent Quartz Size (EQS) classes of the sediment. The
636 bold and dashed lines in (a) denote the average and standard deviation of
637 instantaneous respiration rate of the original soil. Colors of the boxes correspond to
638 the likely fate of each fraction after erosion. See section 4.1 and Figure 6 for
639 definitions and explanation of the three manners of likely fate. Bars in boxes
640 represent median values. Whiskers indicate the lowest datum within 1.5 interquartile
641 range of the lower quartile, and the highest datum within 1.5 interquartile range of the
642 upper quartile ($n = 18$).

643 **Figure 5** The distribution of instantaneous respiration rate per gram of soil organic
644 carbon (SOC) in different Equivalent Quartz Size (EQS) classes of the sediment. The
645 bold and dashed lines denote the average and standard deviation of instantaneous
646 respiration rate per gram SOC of the original soil. Colors of the boxes correspond to
647 the likely fate of each fraction after erosion. See section 4.1 and Figure 6 for
648 definitions and explanation of the three manners of likely fate. Bars in boxes
649 represent median values. Whiskers indicate the lowest datum within 1.5 interquartile
650 range of the lower quartile, and the highest datum within 1.5 interquartile range of the
651 upper quartile ($n = 18$).

652 **Figure 6** Likely fate of eroded soil organic carbon (SOC) as a function of aggregate
653 size, re-drawn from the conceptual model developed by Starr et al. (2000). Blocks of
654 different colors represent three manners of likely fate of eroded SOC, divided by the
655 two convenient cut-off points: aggregate size of 6.3 μm and 63 μm . See section 4.1
656 for definitions and explanation of the three manners of likely fate.

657 **Figure 7** The likely fate of sediment fractions (a), eroded SOC (b), and potential
658 share of CO₂ emission (c) by fractions that would have been likely transferred to

659 rivers, possibly transferred to rivers, and deposited along eroding hillslopes. The bar
660 in box represents the median value, while numbers written in each box denote the
661 average value. Whiskers indicate the lowest datum within 1.5 interquartile range of
662 the lower quartile, and the highest datum within 1.5 interquartile range of the upper
663 quartile ($n = 18$).