The fluvial flux of particulate organic matter from the UK: the emission factor of soil erosion.
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# 8 Abstract

Soil erosion has been identified as a potential global carbon sink since eroded organic matter 9 10 is replaced at source and eroded material is readily buried. However, this argument has relied on poor estimates of the total fate of in-transit particulates and could erroneously imply soil 11 erosion could be encouraged to generate carbon stores. These previous estimates have not 12 considered that organic matter can also be released to the atmosphere as a range of 13 greenhouse gases, not only  $CO_2$ , but also the more powerful greenhouse gases  $CH_4$  and  $N_2O$ . 14 As soil carbon lost by erosion is only replaced by uptake of CO<sub>2</sub>, this could represent a 15 considerable imbalance in greenhouse gas warming potential, even if it is not significant in 16 terms of overall carbon flux. This work therefore considers the flux of particulate organic 17 matter through UK rivers with respect to both carbon fluxes and greenhouse gas emissions. 18 The results show that, although emissions to the atmosphere are dominated by  $CO_2$ , there are 19 also considerable fluxes of CH<sub>4</sub> and N<sub>2</sub>O. The results suggest that soil erosion is a net source 20 of greenhouse gases with median emission factors of 5.5, 4.4 and 0.3 tonnes  $CO_{2eq}/yr$  for 1 21 tonne of fluvial carbon, gross carbon erosion and gross soil erosion, respectively. This study 22 concludes that gross soil erosion would therefore only be a net sink of both carbon and 23

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greenhouse gases if all the following criteria are met: the gross soil erosion rate were very low (< 91 tonnes/km<sup>2</sup>/yr); the eroded carbon were completely replaced by new soil organic matter; and if less than half of the gross erosion made it into the stream network. By establishing the emission factor for soil erosion, it becomes possible to properly account for the benefits of good soil management in minimising losses of greenhouse gases to the atmosphere as a by-product of soil erosion.

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31 Keywords: particulate organic carbon, particulate organic nitrogen, soil erosion, greenhouse
32 gas emissions.

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## 34 1. Introduction

35 It has been argued that the erosion of particulate organic carbon (POC) from soils constitutes a global carbon sink because the eroded soil organic carbon lost to POC is replaced whilst the 36 eroded POC is stored by downstream burial (Stallard, 1998; Harden et al., 1999; Smith et al., 37 38 2005). On this premise, Van Oost et al. (2007) suggested global agricultural land was a net carbon sink of 120 Mtonnes C/yr, based on a soil erosion rate of between 470 and 610 39 Mtonnes C/yr. However, Van Oost et al. (2007) explicitly stated that their method made no 40 allowance for in-stream loss of the POC to atmosphere once out of the immediate catchment 41 area, or for the burial efficiency in marine waters; in effect, they assumed that, once outside 42 43 of the immediate source area, the POC would be buried into a long-term store (e.g. alluvium). Van Oost et al. (2007) reported between 470 and 610 Mtonnes C/yr were lost globally due to 44 soil erosion of which between 240 and 570 Mtonnes C/yr was retained in the immediate 45 catchment, which meant between 30 and 220 Mtonnes C/yr were exported to streams and 46 assumed to be buried. For the UK, Quinton et al. (2006) suggested that the amount of carbon 47 stored due to soil erosion from agricultural land was up to 0.75 Mtonnes C/yr (3.1 tonnes 48

C/km<sup>2</sup>/yr) based on a POC flux to the fluvial network equivalent to between 0.8 and 2.9 tonnes C/km<sup>2</sup>/yr. Whether just soil erosion from agricultural land is considered or erosion of organic particles as a whole, the basic argument remains the same: while eroded carbon particles are transported to permanent burial, the lost carbon is replaced at source from the atmosphere.

The potential for soil erosion to be a net carbon sink arises from the balance between 54 replacement of the eroded soil and the burial of the eroded material. Soil erosion can expose 55 fresh mineral surfaces that have a greater capacity to stabilise accumulated organic carbon 56 57 than the original uneroded soil surface (Quinton et al., 2010). Deposition of the eroded particles can rapidly bury organic matter and so protect it from decomposition (Berhe et al., 58 2007). Eroded mineral particles can adsorb and protect dissolved organic matter upon 59 60 entering rivers (Aufdenkampe et al., 2011). However, these processes are countered by enhanced decomposition of organic matter as aggregates are broken up in the erosion process 61 (Alewell et al., 2009) and by reduced fertility and primary production at the erosion site (Lal 62 63 et al., 2003). Discussion has tended to focus on in-field processes (e.g. Ni et al., 2012) and not the fate of organic matter once it has entered the fluvial system. It must always be 64 remembered that not all particular organic matter entering the fluvial network will come from 65 soil erosion from agricultural land and may come, for example, from bank erosion or 66 67 landslides. Hilton et al. (2015) estimated 15% weathering of particulate fossil carbon, not 68 derived from soil particles, across catchments in Taiwan. Hoffmann et al. (2013a) cited the loss rates reported by Tranvik et al. (2009) in their discussion of whether soil erosion is a 69 carbon sink but did not then use those values to conclude whether soil erosion was a C sink or 70 71 not. Equally, Aufdenkampe et al. (2011) discussed the outgassing of  $CO_2$  and the potential for soil erosion to sequester carbon but did not explicitly consider the turnover of POM to 72  $CO_2$ . If there is loss of the carbon to the atmosphere while the particles, whatever their 73

source, are in transit from source to burial, then this limits the carbon store that soil erosion
could represent. A knowledge of in-stream particulate organic matter (POM) turnover is thus
vital to determine whether soil erosion is a net source or a net sink of carbon.

77 The existing approach used to consider atmospheric impacts of soil erosion is potentially misleading: it is common to consider the POC and the carbon component of 78 79 eroded particles as they are transported through the catchment to shelf seas, but the 80 transported organic carbon is only one component of the total organic matter. Naturally-81 occurring organic matter is generally only 50% carbon, so an assessment of only POC can 82 underestimate the importance of organic matter because it ignores organic oxygen and nitrogen. The oxygen content of the organic matter controls its oxidation state which, in turn, 83 84 controls its impact on atmospheric CO<sub>2</sub> (Worrall et al., 2013). The N content represents an 85 important nutrient source to river biota and so can drive organic matter turnover and, perhaps 86 of more consequence, the N could be released as the powerful greenhouse gas (GHG) N<sub>2</sub>O. Not only does the organic matter consist of elements other than C that play a role in its 87 88 turnover, but also the form of release, or species, can vary: nitrogen could be released as  $N_2$ or N<sub>2</sub>O, and the carbon can be released as either CO<sub>2</sub> or CH<sub>4</sub>, the latter, like N<sub>2</sub>O, being the 89 90 more powerful greenhouse gas than  $CO_2$  (Houghton et al. 1995).

In many studies there is a tacit assumption that a discussion of carbon sinks and 91 92 sources is equivalent to discussion of the overall impact upon atmosphere (e.g. Hoffmann et 93 al., 2013a), but the real impact on the atmosphere comes only from understanding the greenhouse gas fluxes; the carbon budget and the greenhouse gas budget are not the same 94 thing. To understand the greenhouse gas budget, the form of release is important, not just a 95 96 consideration of carbon but to understand the greenhouse gas warming potential from eroded organic matter; we must therefore distinguish between CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O. Unless the whole 97 98 greenhouse gas budget is considered, it is possible to reach the potentially misleading 99 conclusion that, if erosion of organic matter can represent a carbon sink, then soil erosion100 should be encouraged.

An alternative approach would be to consider the flux of carbon from rivers to 101 102 estuaries and shelf seas rather than starting from fluxes of suspended sediment. Meybeck (1993) estimated that the carbon flux (dissolved organic carbon, particulate organic carbon 103 104 and dissolved inorganic carbon) from the world's rivers to the oceans was around 542 Mtonnes C/yr in proportions DOC:POC:DIC as 37:18:45, respectively, i.e. global river flux 105 of POC is about 98 Mtonnes C/yr. Ludwig et al. (1996) used a spatially-explicit model of 106 global fluvial carbon fluxes to suggest fluxes of 800 Mtonnes C/yr with a split of 107 approximately 50:25:25 for DOC:POC:DIC, i.e. a global POC flux of 200 Mtonnes C/yr. 108 109 These figures provided useful estimates of fluvial POC losses from the land to the oceans at 110 the tidal limit, but they did not account for in-stream losses along the length of the river, between the carbon sources (e.g. soils) and the ocean. Battin et al. (2008) used a value of 180 111 Mtonnes C/yr for the global flux of POC from rivers to oceans based on values from Cauwet 112 (2002). Bauer et al. (2013) suggested that the flux of DOC+POC from rivers to shelf seas 113 (including estuaries) was 450 Mtonnes C/yr, based on values from Syvitski et al. (2005). 114 Syvitski et al. (2005) suggested that pre-human POC fluxes were between 140 and 470 115 Mtonnes C/yr decreasing to between 126 and 380 Mtonnes C/yr in modern times with the 116 difference being the role of reservoir storage outstripping the influence of increased soil 117 118 erosion.

For the UK, a more detailed analysis of carbon fluxes through and from rivers has been possible. Worrall et al. (2007) used nationally-collected monitoring data to assess national-scale loss at the tidal limit, the loss from the terrestrial biosphere and the loss from rivers to the atmosphere. They found that in total 10.1 tonnes C/km<sup>2</sup>/yr were lost from the terrestrial biosphere to rivers of which 2.5 tonnes C/km<sup>2</sup>/yr was as POC. However, the study

considered in-stream losses only for DOC and DIC but did not include POC. Worrall et al. (2014) updated the POC flux estimates and found that POC loss from terrestrial sources (all terrestrial sources, not just soil erosion) was 4.6 tonnes  $C/km^2/yr$  with the equivalent of 1.1 tonnes  $C/km^2/yr$  being lost in-stream.

128 The trade-off between replacement within catchment, transfer to the stream network,129 loss in stream, and eventual, permanent burial can be simply expressed as:

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131 
$$P_{replace} \ge P_{deliver}(1 - P_{burial})$$
 (i)

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where:  $P_{replace}$  = the proportion of organic carbon that is replaced following erosion; 133 134  $P_{deliver}$  = the proportion of eroded organic carbon delivered to the stream network; and  $P_{burial}$  = the proportion of POC flux that is buried in marine sediment or other permanent burial. Note 135 that Equation (i) applies for any eroded particulate organic carbon and not just from soils and 136 not just from agricultural land. The term  $(1 - P_{burial})$  represents the loss to the atmosphere in 137 transit. However, this equation does not hold for greenhouse gases because replacement will 138 be as CO<sub>2</sub> but loss within stream could be as CO<sub>2</sub>, CH<sub>4</sub> or N<sub>2</sub>O. Therefore, Equation (i) 139 should be re-written as: 140

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$$K_{CO2}P_{replace} \ge P_{deliver}(K_{CO2}P_{CO2} + K_{CH4}P_{CH4} + K_{N20}P_{N20})$$
(ii)

$$1 - P_{buried} = (K_{CO2}P_{CO2} + K_{CH4}P_{CH4} + K_{N20}P_{N20})$$
(iii)

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145 where:  $K_x$  = the greenhouse gas warming potential of x where x is CO<sub>2</sub>, CH<sub>4</sub> or N<sub>2</sub>O; and P<sub>y</sub> 146 = the proportion of loss of particulate organic matter that is lost as y, with y as CO<sub>2</sub>, CH<sub>4</sub> or 147 N<sub>2</sub>O. The aim of this study is to assess the complete greenhouse gas impact of soil erosion of organic matter from erosion to burial and so derive a more complete emission factor for this process. If a realistic emission factor for soil erosion can be derived, then the benefit of preventing soil erosion can be better understood.

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#### 153 2. Approach

This study considers the fluvial system as outlined in Figure 1, whereby eroded organic 154 matter (gross erosion) can be deposited within the terrestrial source area or delivered to the 155 156 stream channel (net erosion). The organic matter removed in gross erosion can (eventually) be replaced. Replacement can be by a number of processes (as outlined above) but in terms of 157 greenhouse gases, the carbon is drawn down from the atmosphere as CO<sub>2</sub> via photosynthesis 158 159 to primary production. Primary production does not draw down carbon from the atmosphere 160 as CH<sub>4</sub> or draw down nitrogen as N<sub>2</sub>O. The amount of POM that reaches the stream network is referred to as the net erosion and will be less than the gross erosion due to the internal (on-161 slope) redistribution. Several studies have suggested that on-slope storage of gross erosion 162 deposited within, for example, a field can prevent mineralisation of the deposited organic 163 matter by a number of processes (e.g. rapid burial - Berhe et al. 2007). 164

On entering the stream network, POM is subject to a number of physical and biogeochemical removal processes. POM can be mineralised within the stream network to all three of the greenhouse gases (CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O). POM can also be permanently buried by in-channel storage or by overbank sedimentation on to floodplains. It is assumed that POM inputs to estuaries and oceans will also be prone to mineralisation to CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O or to permanent burial.

171 The processes affecting each step presented in Figure 1 are quantified using data from 172 the literature, previous results of the authors' own work and newly available data from UK monitoring programmes. For each important process and storage compartment, the range was estimated. It was not always possible to give a consistent measure of uncertainty for each input variable or parameter: for some measures a range was not available while for others the distribution was known such that a mean and standard deviation could be calculated. The expected value and measure of variation were detailed and the total carbon budget and total greenhouse gas warming potential were calculated as a stochastic combination of the estimated ranges. When not known, a distribution was taken to be uniform.

Wherever possible, the assessment was considered over a 100-year window which in turn meant that values of  $K_x$  - the greenhouse gas warming potential (GWP) of x - in Equations (ii) and (iii) were then 3.67, 24, and 292 for CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O respectively. By convention, fluxes of carbon, or greenhouse gases, are considered relative to the atmosphere and therefore sinks to land are negative.

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#### 186 **3. Methods**

In line with the approach above this study estimated: replacement ratio; enrichment ratio; mineralisation rate of internally-redistributed sediment; the ratio of net erosion leaving the site of erosion (e.g. an agricultural field) to the stream network and the gross erosion within the site of erosion (e.g. the field) (net to gross ratio – Quinton et al., 2006); fluvial flux of POM; C/N of POM; proportion of loss as  $CH_4$  and  $N_2O$ ; estuarine removal; shelf sea removal; and, terrestrial burial efficiency.

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#### 194 *3.1. Replacement rate*

A key component of the argument that terrestrial erosion represents a net carbon sink is the extent to which the eroded carbon can be replaced. Van Oost et al. (2007) took the replacement rate to be between 0.19 and 0.58 with a preferred value of 0.26, i.e. 26% of the eroded carbon is replaced. However, several studies (e.g. Quinton et al., 2006) have assumedreplacement rates as high as 1. Therefore, we assumed a range of 0.19 to 1.

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#### 201 3.2. Enrichment ratio

Gross erosion of soil has been commonly observed to be enriched in carbon relative to the bulk soil; this fractionation is expressed as an enrichment ratio – the proportion of the carbon in the eroded particle relative to that in the bulk surface soil. Quinton et al. (2006) identified enrichment ratios as between 1.2 and 4.7 for UK soils. We have assumed that enrichment ratios observed for soil erosion in agricultural soils are true for erosion for other settings.

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# 208 3.3. Mineralisation rate of internally redistributed sediment

209 Most studies have assumed this to be zero (e.g. Van Oost et al., 2007) but we believe it is unreasonable to assume that no redistributed carbon can be mineralised. Van Hemelryck et al. 210 (2010) measured the mineralisation rate of redistributed soil particles; applying their derived 211 equations over a 100-year window, i.e. the same window of time for which the GWP values 212 are derived, gives a value of 0.75. In other words, over a 100-year period, 25% of the 213 internally redistributed will be mineralised to CO<sub>2</sub> and 75% will remain. Therefore, for this 214 study we took the range of the proportion of internally re-distributed organic matter that is 215 stored over a 100-year window as between 0.75 to 0.95. 216

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#### 218 3.3. Net to gross ratio

Gross erosion (e.g. the erosion rate within the field) will not necessarily reach the fluvial network and can instead be deposited on-slope. Thus the net erosion (that which leaves the field and enters the channel network) is less than or equal to the gross erosion. One reason for the difference between the net and the gross erosion rates is the storage of sediment within field and before reaching the stream network. Quinton et al. (2006) reviewed net to gross ratio for UK settings and gave values of between 0.47 and 0.65, i.e. at its maximum, the net erosion was 65% of the gross erosion.

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#### 227 3.4. Fluvial flux of POM

Worrall et al. (2014) examined all 35,490 records of POM concentration from 1974 to 2010 228 from all 270 catchments with a mean discharge > 2  $m^3/s$  included in the UK's Harmonised 229 Monitoring Scheme. Applying the rule that for a flux calculation within any monitored 230 231 catchment there would have to be a minimum of 12 samples per year, this gave 2,808 flux estimates from 111 sites. When only the flux estimates for sites at the tidal limit of the 232 catchment were considered and only those with data between 2001 and 2010, then the flux of 233 234 POM from 80 catchments across the UK for the years 2001 to 2010 could be calculated. Instream losses were then estimated by comparing these catchments, and allowing for 235 differences in hydro-climatic and land-use factors. This approach is based upon net POM 236 fluxes from these catchments and so includes all sources of POM to the fluvial network and 237 not just from one specific source or process. The 80 catchments covered a range of 238 landscapes in England and Wales where the soil type varied from 0 to 100% of 3 soil classes 239 (mineral, organo-mineral and organic soils as defined by Hodgson, 1997). Land use in the 80 240 catchments varied from: 0.1 to 70% arable; 2 to 78% grassland; and 0.4 to 36% urban. 241

In-stream losses can be due to turnover of the organic matter or burial in channel or in the floodplain. To calculate the proportion of the in-stream loss due to floodplain sedimentation, it is necessary to estimate the proportion of time that the discharge in any UK river is greater than bankfull discharge, and therefore the proportion of time in which there is flow and sediment delivery to the floodplain. Nixon (1959) found that 29 English rivers were at or exceeded their bankfull discharge between 0.1 and 2.9% of the time, i.e. overbank

sedimentation would be occurring between 1 day every 3 years and 11 days per year. Days of 248 overbank sedimentation are likely to be days of high flow in the main channel and thus days 249 of considerable sediment flux. Therefore, using the POM flux data from Worrall et al. (2014), 250 251 we assumed the highest flows each year were overbank flows and that any sediment flux they carried was lost to overbank storage. The number of days of flux lost to overbank 252 sedimentation was varied from the lowest to the highest value as measured by Nixon (1959) 253 (0.33 to 11 days per year) with the assumption that the first day of overbank sedimentation 254 was the day with the highest flux of POM in that year and then, for each further day of the 255 256 overbank sedimentation, it was assumed that these were days of progressively lower POM flux. It was then assumed that all the POM flux on days of overbank sedimentation was lost 257 to overbank sedimentation. The POM flux lost each year to overbank sedimentation was 258 259 expressed as a percentage of the total POM flux from the catchment for that year. Because all 260 available comparator data were for overbank storage of suspended sediment and not POM, the calculation was also performed for suspended sediment based upon flux values given by 261 262 Worrall et al. (2013b). It was assumed that POM flux lost to overbank sedimentation was permanently stored and not mineralised to the atmosphere within the time frame considered. 263 The limitations of these assumptions are discussed later on. 264

Collins and Walling (2007) gave values of in-channel storage as between 18% and 57% of the outlet flux for two UK lowland streams but they noted that most of this storage was transient. Indeed, Walling et al. (2002) noted that permanent in-channel storage was only between 2 and 5% of the catchment-outlet flux of suspended sediment and, given that Worrall et al. (2014) showed that on average suspended sediment in the UK was 30% POM, then for this study it is assumed that in-channel storage of was 1 - 2% of the incoming POM flux.

#### 273 3.5. C/N of POM

Worrall et al. (2014) estimated POC and PON from POM given that the organic carbon 274 content of organic matter was between 45 and 50% and that the average C/N ratio of 275 276 suspended sediment in the UK was  $8.1 \pm 5.2$  (n=13: Hillier, 2001). Here, we used POM data collected as part of the LOIS project (Neal and Davies, 2003) compared to literature values 277 (Table 1). The LOIS project collected 2,484 samples for POM across 5 years for the Humber 278 Basin (26,109 km<sup>2</sup>; 17% of the UK catchment area). Analysis of variance (ANOVA) was 279 used to test whether C/N of POM varied from between sampling site, sampling month and 280 281 sampling year. To comply with the assumptions of ANOVA, the data were tested using the Levene and the Anderson-Darling tests and transformed as required. Differences between the 282 levels of significant factors were tested using the post hoc Tukey test. Data from the LOIS 283 284 project were augmented and compared with data from the literature. There was no information in the LOIS data on the concentration of particulate nitrogen or particulate 285 carbon so we followed Worrall et al. (2014) and used a range of 45 to 50%. 286

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#### 288 **3.6.** Proportion of $CH_4$ and $N_2O$

POM lost in the fluvial network, estuaries or shelf seas can be lost as  $CO_2$ ,  $CH_4$  or as  $N_2O$ . Therefore, this study examined the literature (Table 2 & 3) to find ranges of the loss of  $CH_4$ as a proportion of C loss in aquatic systems and likewise the loss of  $N_2O$  as a proportion of N loss.

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#### 294 3.7. Estuarine removal

Estuaries will remove sediment and with it POM. Tappin et al. (2003) reported a POC budget for the Humber estuary (17% of UK's runoff drains through this estuary with a residence time of 2-3 months) and found that for 3 years (1994 -1996) the flux of POC from the estuary 298 varied between 16 and 43% of the fluvial POC flux into the estuary and that burial rate was 4% of input with the remainder of the fluvial POC flux input to the estuary being mineralised 299 (between 36 and 54%). The estimates from Tappin et al. (2003) are given without further 300 301 estimates of any uncertainty and the ranges given were used with the additional caveat that there must be mass balance. The proportion of organic matter buried was taken such that 302 mass balance was met, i.e. proportion of the incoming flux that was buried may be greater 303 than 4% given the variation in the proportions lost to mineralisation and the exported from 304 the estuary. That is: 305

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307 
$$P_{burial}^{estuary} = 1 - P_{mineral}^{estuary} - P_{transfer}^{estuary}$$
 (iv)

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where:  $P_x^{estuary}$  = the proportion of the x with x as buried (burial), mineralised to greenhouse gases (mineral) or transiting through the estuary to the shelf (transfer).

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#### 312 3.8. Shelf-sea processes

Galy et al. (2007) reported very high burial efficiencies (approx. 100%) of fluvially-derived 313 carbon in the Ganges-Brahmaputra fan, which they ascribed to rapid burial, but these 314 sediments also have remarkably small POC contents ( $0.6 \times 10^{12} \text{ mol C/yr from 1 } \times 10^9 \text{ tonnes}$ 315 of suspended sediment, equivalent to less than 1% C content - Frances-Lenard and Derry, 316 1997), and therefore the Ganges-Brahmaputra has an export equivalent to 4.4 tonnes 317 C/km<sup>2</sup>/yr compared to the 3.5 tonnes C/km<sup>2</sup>/yr that the UK exports at its tidal limit. Equally, 318 the estimate of 100% burial, and therefore a large carbon sink due to the Ganges-319 320 Brahmaputra fan, has neglected to account for the in-stream losses of carbon from particulates before reaching the sea. For other rivers, Burdige (2005) suggested a removal 321 rate from source to ocean sediment of 70% based upon a measured burial efficiency in ocean 322

323 sediment of 30%. Burdige (2005) presented no new data but quoted data from Aller (1998) who in turn quoted Canfield (1994) who included data from Middleburg (1991) and Reimer 324 et al. (1992), but relied mostly upon data from Canfield (1989). In this study we supplement 325 326 this information further with data from Meyer et al. (2007), Weijers et al. (2009), Li et al (2013) and Hung et al. (2012). The data from these sources are for the carbon sedimentation 327 rate but not the sedimentation rate of terrestrial organic carbon. Therefore, the following 328 sources were used to estimate the burial rate of the terrestrial organic carbon compared to the 329 sedimentation rate in shelf seas: Burdige (2005), Weijers et al. (2009), Li et al. (2013), Hung 330 et al. (2012) and Meyer et al. (2007). These data were compiled to relate the carbon 331 sedimentation rates to the burial efficiency of the terrestrials organic carbon. With respect to 332 the UK, there are several studies of the sediment and carbon budgets of the North Sea 333 334 (Brockman et al., 1990; de Hass et al., 1997a, b).

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#### **336 4. The net effect of soil erosion on GHG emissions**

#### 337 4.1. Fluvial flux of POM

338 The best-fit multiple regression equation reported by Worrall et al. (2014) was:

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$$POM_{flux} = 3827 + 6.70rgmin + 8.10rg + 7.5Grass - 2.4Area$$
(v)

(2.2)

341

(2.6)

(842)

342  $r^2 = 0.5, n=80, p < 0.05$ 

343

where: Orgmin = the area of organo-mineral in the catchment (km<sup>2</sup>); Org = the area of organic soils in the catchment (km<sup>2</sup>); Grass = the area of grazed land within the catchment (km<sup>2</sup>); and *Area* = the area of the catchment (km<sup>2</sup>). The values in brackets represent the standard errors in the coefficients. Equation (v) can be interpreted as an export coefficient

(3.3)

(1.4)

348 model where each regression coefficient is interpreted as an export coefficient. Thus, Equation (v) predicts that 1 km<sup>2</sup> of organo-mineral soil would export 6.7  $\pm$  2.6 tonnes/km<sup>2</sup>/vr 349 of POM where the range denotes the coefficient's standard error. This interpretation suggests 350 351 the biggest source of POM is organic soil; POC fluxes are commonly reported for peatcovered catchments where the extent of degradation and vegetation cover control the loss of 352 POC and fluxes can be as high as 195 tonnes C/km<sup>2</sup>/yr (Evans et al., 2006). Most 353 contemporary studies of soil erosion in the UK and especially those concerned with the 354 carbon or greenhouse gas emissions have focused upon mineral soils and arable land (e.g. 355 356 Rickson, 2014), but the results from this study imply there is no significant flux of POM from a catchment with only arable or urban land use on mineral soils. Most studies have focused 357 on the production of sediments from soil erosion as opposed to the study of POM more 358 359 generally (e.g. Quinton et al., 2006); Equation (v) implies that certain land uses are not that important in the contemporary flux of POM from the UK. Equation (v) has a y-intercept 360 value which predicts that any catchment will have a minimum export of  $3817 \pm 842$ 361 362 tonnes/yr; this may relate to erosion unrelated to land use, e.g. bank erosion. Equation (v) can be applied across the UK given knowledge of land use and soil distribution (Defra, 2005, 363 Lilly et al., 2009) to give the flux of POM from the UK at the tidal limit: the average value 364 for the period 2001 to 2010 was  $1195 \pm 308$  ktonnes/yr. 365

Equation (v) includes a significant loss term with catchment area which implies that, for every additional 1 km<sup>2</sup> of catchment area, 2.4 tonnes/yr of POM are lost. Given the loss rate, the amount of POM lost in transit through UK rivers would be  $594 \pm 206$  ktonnes/yr which in turn means a loss of POM at the soil source of  $1854 \pm 238$  ktonnes/yr. This gives an in-stream removal rate for POM of  $33.5 \pm 11.2\%$ .

371 Using the estimates of days of bankfull discharge and the highest daily fluxes gives an372 estimate that, for one day each year, the loss to floodplains of suspended sediment flux is

2.5% of the total flux leaving the catchment and therefore the maximum percentage lost to 373 overbank sedimentation would be 27.5% (i.e. for 11 days per year of bankfull or greater 374 discharge). The same analysis for POM flux shows that only 0.97% of the annual flux is lost 375 376 per day of overbank flow, in which case after 11 days of overbank flow only 10.9% of the POM flux would be removed. Note that the percentage losses are less for POM than for 377 suspended sediment, i.e. POM is not fractionated into overbank storage relative to suspended 378 sediment. In the UK case, the proportion of suspended sediment that is organic matter 379 decreases as flow increases. Walling et al. (1999) estimated overbank sedimentation for the 380 Yorkshire Ouse as 30% of the outlet flux (23% of influent suspended sediment flux) and as 381 40% of the outlet flux (29% of influent flux) for the River Tweed. Erkens (2009) gave a long-382 term, Holocene accumulation rate of total sediment in the Rhine floodplain as 27% of the 383 384 upstream input, but this was not a measure of the organic carbon storage. Hoffmann et al. 385 (2009) suggested that the long-term storage of carbon on the Rhine floodplain was equivalent to the downstream flux of POC at the catchment outlet. In contrast, Gomez et al. (2003) 386 387 found only 4% POC storage in a New Zealand floodplain.

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#### 389 4.2. C/N of POM

Across 5 years (1994 - 1998) and 16 sites across 13 rivers (Rivers Aire, Calder, Derwent, 390 Don, Great Ouse, Nidd, Yorkshire Ouse, Swale, Trent, Tweed, Ure, Wear and Wharfe -391 Robson and Neal, 1997), the median POC/PON C/N ratio was 11.5 with an interquartile 392 range of 9.2 to 14.3. The Anderson-Darling test showed the data were log-normally 393 distributed and so ANOVA was performed on log-transformed data; the Box-Cox 394 transformation removed 7 out of 2,477 data. ANOVA showed that all factors were 395 significant. By far the most important factor was the difference between months with a 396 minimum in C/N value in June and a maximum in February; the seasonal cycle is remarkably 397

symmetrical. The second most important factor is the difference between years, although *post hoc* testing indicated that the main difference lay between the years 1993 ( $13.8 \pm 1$ ) and 1995 ( $10.1 \pm 1$ ). The year 1995 was a drought year in the UK (1 in 33 year drought – Worrall et al. 2008). The difference between sites explained only 4.8% of the original variance and was between only 3 out of the 16 sites Calder (Methley Bridge 12.7), Swale (Catterick Bridge 13.0) and Trent (Cromwell Lock 10.1).

The review of literature data allowed an estimation of mean and standard deviation 404 for 47 catchments (Table 1). The geometric mean of the catchments, not including any of the 405 406 UK data, was 10.3. The discharge-weighted average from the review of Ittekkot and Zhang (1989) was 10.7 and for the LOIS data the geometric mean was 11.7. Therefore, applying this 407 to UK values of POM flux, the flux of POC at the tidal limit would be  $601 \pm 152$  ktonnes 408 409 C/yr. The POC flux at the terrestrial source would be  $877 \pm 102$  ktonnes C/yr meaning that 410 the carbon lost through the streams of the UK would be  $290 \pm 96$  ktonnes C/yr. Equally, the flux of PON at the tidal limit would be  $52 \pm 13$  ktonnes N/yr. The PON flux at the soil source 411 412 would be  $74 \pm 14$  ktonnes C/yr meaning that the nitrogen lost through the streams of the UK would be  $23 \pm 9$  ktonnes N/yr. 413

414

# 415 *4.3. Shelf-sea processes*

Burdige (2005) suggested a figure of 30% for burial efficiency of terrestrial organic matter into permanent burial in shelf sediments; Weijers et al. (2009), Li et al. (2013), Hung et al. (2012), and Meyer et al. (2007) had values between 30 and 79% with a median value of 59% (n=13, Figure 2). Given the median value of terrigenous input of the organic sedimentation rate means that the best-fit equation is for burial efficiency ( $B_{tom}^{eff}$ ):

422 
$$B_{tom}^{eff} = 49 + 5.7 log_e(S_{om})$$
 n = 25, r<sup>2</sup> = 67.4%, p < 0.05 (vi)

- (3.0) (0.8)
- 424

425 where:  $S_{om}$  = the sedimentation rate of organic matter (g C/m<sup>2</sup>/yr).;

Brockman et al. (1990) estimate that the POC flux to the North Sea basin is 4 426 Mtonnes C/yr. Haas et al. (1997a, b) give the carbon budget for the basin itself. On the North 427 Sea plateau the accumulation is limited by scouring currents to 100 ktonnes C/yr with most 428 accumulation in the deep channels to the east of the sea (Norwegian channel and Skaggerak) 429 at 1 Mtonnes C/yr and another 0.1 Mtonnes C/yr exported off the shelf into the Norwegian 430 431 Sea (de Haas et al., 1997a,b): this would give a burial efficiency of 30%. de Haas et al. (1997a) suggest that 20% of the accumulated sediment is of terrestrial origin and they 432 measured a series of sedimentation rates varying from 0.05 to 0.35  $g/m^2/yr$  which, given the 433 434 relationship presented in Equation (vi), gives a range of burial efficiency from 31 to 43% with a 95% confidence interval of 27 to 49%. However, for the UK, the North Sea is but one 435 shelf sea and de Hass et al (2002) note that for the Celtic Sea the sea bed is entirely made of 436 437 re-worked Pleistocene sediment which meant that there was no accumulation on the shelf but there are no measurements of the export off the shelf to the ocean. Therefore, in the case of 438 UK, figures of burial efficiency for the North Sea must be viewed as the upper range of 439 possible values. 440

441

#### 442 4.4. Proportion of loss as CH<sub>4</sub>

Included in the analysis is the range of values for reservoirs (Guerin et al., 2006). Of the 12
measurements that are detailed in Table 2, the range used in this study was taken as the 5<sup>th</sup> to
95<sup>th</sup> percentile range (0.64% to 2.2%) with a median value of 0.97%.

446

# 447 4.5. Proportion of loss as $N_2O$

The ranges reported in the literature are given in Table 3. The IPCC guidelines say that the N<sub>2</sub>O yield would be 2.5% of leached N, where leached N is calculated as 30% of applied fertiliser and manure N when runoff is greater than 50% of pan evaporation (IPCC, 2007). Clearly such an approach has no application to PON. Baulch et al. (2011) found a consistent N<sub>2</sub>O yield of 0.75% across 72 watersheds in the US. The present estimate of N<sub>2</sub>O flux from UK rivers is 24 ktonnes N<sub>2</sub>O/yr, based on IPCC guidelines.

454

# 455 4.6. Stochastic modelling of GHG emissions factor

456 The ranges of the input parameters used are given in Table 4. Given the stochastic combination of the ranges developed above, the total greenhouse gas flux due to particulate 457 erosion, flux and burial from and in the UK leads to a median emission of 3853 ktonnes 458  $CO_{2eq}/yr$  with a 5<sup>th</sup> to 95<sup>th</sup> percentile range of 2970 to 7807 ktonnes  $CO_{2eq}/yr$ . The median 459 greenhouse gas flux of the replacement of -2490 ktonnes  $CO_{2eq}/yr$  with a 5 to 95<sup>th</sup> percentile 460 range of -1260 to -5822 ktonnes  $CO_{2eq}$ /yr, which is 56.9% of the emissions (ranging between 461 462 25.3 and 137.9%). Given the input ranges, there is only a 12.8% chance that erosion in the UK is a greenhouse gas sink. When the individual greenhouse gases are considered, then CO<sub>2</sub> 463 represents 74% of the greenhouse gas warming potential (59 to 89% - 5<sup>th</sup> to 95<sup>th</sup> percentile); 464  $CH_4$  represents 3.8% of the greenhouse gas warming potential (2.1 to 6.5% - 5^{th} to 95^{th} 465 percentile); and N<sub>2</sub>O 22% of the greenhouse gas warming potential (5 to 37% - 5<sup>th</sup> to 95<sup>th</sup> 466 percentile). The largest loss of greenhouse gases is from the shelf seas (48%), then estuaries 467 (30%), with the least from the rivers (19%). The distribution of the greenhouse gas fluxes and 468 POM fluxes are summarised in Figure 3. When overall flux of greenhouse gases is compared 469 470 to flux of particulate carbon from the soil, then 1 tonne of particulate carbon entering the fluvial network gives a median emission factor of 5.5 tonnes CO<sub>2eq</sub>/tonnes C/yr with a 3.3 to 471 9.9 as the  $5^{\text{th}}$  to  $95^{\text{th}}$  percentile. 472

For 1 tonne of C released as particulate matter to the fluvial network, then it is possible to give ranges of what this may mean in terms of gross erosion. Given the ranges discussed above, 1 tonne of fluvial organic particles will have come from 0.47 to 1 tonne of gross erosion of soil carbon, which will in turn have been replaced by between 0.19 and 1 tonne C. The emission factor of 1 tonne of gross carbon erosion is then:

479 
$$GWP_{0e} = GWP_{Ne} - \frac{N_{Ce}}{G_{Ce}}RMK_{CO_2}$$
 (vii)

480

where:  $GWP_{Ne}$  = the emission factor of net erosion of organic particles (tonnes  $CO_{2eq}/yr$ ); 481  $N_e/G_e$  = the net to gross erosion ratio; R = replacement rate; M = mineralisation rate of 482 internally redistributed carbon particles;  $K_{CO_2}$  = the greenhouse gas warming potential of CO<sub>2</sub> 483 (3.67). Given the ranges quoted above, the median value of the sink due to replacement and 484 internal deposition is a sink of -1.1 tonnes  $CO_{2eq}/yr$  with a 5<sup>th</sup> to 95<sup>th</sup> percentile range of -0.6 485 to -2.3 tonnes CO<sub>2eq</sub>/yr. Given this value for the sink due to replacement and internal 486 redistribution, then the emission factor of 1 tonne of carbon (GWP<sub>Oe</sub>) from gross erosion 487 would be a source of 4.4 tonnes  $CO_{2eq}/yr$  with a 5<sup>th</sup> to 95<sup>th</sup> percentile range 1.6 to 8.9 tonnes 488 CO<sub>2eq</sub>/yr. 489

Many authors have noted that eroded soil is enriched in organic carbon compared to topsoil; Quinton et al. (2006) gave a range of enrichment ratios of between 1.2 and 4.7 for UK soils. The majority of mineral soils in the UK have been 2 and 4% soil organic carbon for a range of land uses (Bell et al., 2010). Assuming that the supply of POM to the fluvial network is predominantly supplied from soil erosion, then 1 tonne of gross mineral erosion represents 0.09 tonnes of C in gross erosion (with a range 0.03 to 0.16 tonnes C) meaning that the emission factor for 1 tonne of gross erosion is 0.30 (5<sup>th</sup> to 95<sup>th</sup> percentile range of 0.11 to 497 0.66) tonnes  $CO_{2eq}/yr$ . Erosion rates in the UK have been reviewed by Defra (2005b) which 498 indicated median values of net soil loss from arable fields in England as 410 tonnes/km<sup>2</sup>/yr 499 and from English grasslands as 60 tonnes/km<sup>2</sup>/yr. Boardman (2013) reviewed soil erosion in 500 Britain and gives values as high 4500 tonnes/km<sup>2</sup>/yr for a bare sandy loam compared to 501 values as low as 30 tonnes/km<sup>2</sup>/yr for clay soil under cereals, i.e. limits suggested on gross 502 erosion estimated above are very low compared to those observed in the UK..

For 1 tonne C released to the fluvial network, the gross erosion can be calculated given the range of values of  $\frac{N_e}{G_e}$  as discussed above; the amount of GHG this removes is

505 3.67R. Therefore, for gross soil erosion to be a net sink of GHG the following must be true:

506

507 
$$\frac{K_{CO_2}}{GWP_{Ne}}R > \left[\frac{N_{Ce}}{G_{Ce}} + \left(1 - \frac{N_{Ce}}{G_{Ce}}\right)M\right]$$
(viii)

508

509 Given the ranges used in this study, for a GHG sink this can only be achieved if R > 0.7 with 510  $\frac{N_e}{G_e} < 0.47$ .

511

# 512 4.7. Stochastic modelling of GHG emissions factor

513 When just carbon is considered, and not all GHGs, there is a median emission of 1099 514 ktonnes C/yr with a 5<sup>th</sup> to 95<sup>th</sup> percentile range of 697 to 1575 ktonnes C/yr. The median 515 carbon flux of the replacement is 656 ktonnes C/yr with a 5<sup>th</sup> to 95<sup>th</sup> percentile range of 343 516 to 1587 ktonnes C/yr, which is 63.7% of the emissions (with a 5<sup>th</sup> to 95<sup>th</sup> percentile between 517 34 and 140%). Given the input ranges, there is only a 17.9% chance that erosion in the UK is 518 a carbon sink.

519

#### 520 **5. Discussion**

521 Given the data used in this study, it is possible to update estimates of the GHG fluxes from UK rivers. This study has not updated the carbon budget estimates for UK rivers provided by 522 Worrall et al. (2014) but has updated the estimates of the GWP of those fluxes by considering 523 not just the turnover of DOC and POC to CO<sub>2</sub> but the turnover of POM and DOM to CO<sub>2</sub>, 524 CH<sub>4</sub> and N<sub>2</sub>O. Table 5 updates the estimates of the GWP and, based upon median values of 525 the ranges used above, the median value of GWP of UK rivers is 14254 ktonnes CO<sub>2ea</sub>/yr; 526 this equates to an additional 58 tonnes  $CO_{2eq}/km^2/yr$  for the UK land area lost from the fluvial 527 network. 528

529 This estimate of the GHG loses from the UK fluvial network has been based upon the assumption that any storage in the fluvial system is a permanent sink, i.e. there is no loss of 530 organic matter from the in-channel or floodplain storage on the time scales of decades. 531 532 Hoffman et al. (213b) have shown that over the last 7500 years the floodplains of European rivers have acted as a net carbon sink because they represent an environment which has a 533 high preservation potential for carbon and that, when this material erodes and is replaced by 534 primary productivity, then the floodplain is still a net sink of carbon. However, even with this 535 mechanism a proportion of POM stored on floodplains would be mineralised and returned to 536 the atmosphere and a further proportion of the POM stored on floodplains would be lost to 537 the atmosphere as the stored POM is eroded and returned to the fluvial network. Therefore, 538 even if the fate of POM within fluvial storage is a net sink, this study has made the even more 539 540 conservative assumption that all of the fluvial storage is acting as a sink. Secondly, it should also be noted that Hoffman et al. (2013b) show their result for carbon and not for greenhouse 541 gases. The relative high standing water tables of floodplains mean that the proportion of 542 543 organic matter lost maybe smaller but the proportion of the deposited POM lost as the more powerful greenhouse gases (CH<sub>4</sub> and N<sub>2</sub>O) maybe greater (Pinay et al., 2007). Thirdly, it 544 should be pointed out that the proportion of the POM lost as storage in the fluvial network is 545

almost 6 times smaller than that lost directly to the atmosphere via turnover in the river and,
furthermore, the greatest proportion of the loss of POM is not in the river network but in the
estuaries and shelf seas. Therefore, the estimates given here are likely to be underestimates.

549 The estimates made in this study show the critical importance of the interplay of the net to gross erosion and the replacement of soil organic matter. In this study we have 550 551 assumed a broader range of replacement rates than assumed by Van Oost et al. (2007) but there is no information on what controls replacement rates and how this relates to gross 552 erosion. It could be argued that the ability of a soil to replace carbon lost as erosion will be 553 akin to its rate to turnover soil organic carbon (SOC). An arable soil in southern England has 554 a surface SOC residence time of 22 years (Jenkinson and Raynor, 1977). If a soil has between 555 2 and 4% SOC, then the plough layer (depth of 20 cm) of an arable field contains between 5.2 556 and 10.4 kg of organic carbon  $/m^2$  (assuming an average bulk surface soil density of 1300 557 kg/m<sup>3</sup>) and the organic carbon is turning over at a rate of between 0.24 and 0.48 kg C/m<sup>2</sup>/yr. 558 Given an enrichment ratio (1.2 to 4.7) and the range in %SOC (2 to 4%) used in this study, 559 then this turnover rate would be equivalent to the amount of carbon exported in a gross 560 erosion of between 3.9 (5<sup>th</sup> to 95<sup>th</sup> percentile range of 0.5 to 8.6) kg C/m<sup>2</sup>/yr equivalent to a 561 median gross soil erosion of 30.8 tonnes/km<sup>2</sup>/vr (5<sup>th</sup> to 95<sup>th</sup> percentile between 2.7 to 91.9 562 tonnes/km<sup>2</sup>/yr). As noted above, grassland soils in the UK would tend to have the higher 563 %SOC and the lower gross soil erosion rates which suggests that for many mineral soils 564 565 under certain land uses (e.g. grassland), the replacement rate could be close to 1 but that the replacement rate (R) would decrease rapidly for arable fields where %SOC is naturally lower 566 and gross soil erosion is commonly higher. It should also be repeated that the evidence for the 567 UK is that mineral soils under arable usage are not the most important sources of POM 568 leaving the UK. 569

570 Given the values of the emissions factors estimated above, then it is possible to reconsider the impact of soil erosion at the national and global scale. Quinton et al. (2006) 571 estimated that between 0.2 and 0.76 Mtonnes C/yr were released by soil erosion in England 572 and Wales (rescaled to the UK this would be 0.3 to 1.2 Mtonnes C/yr) which we can now 573 equate to 3.9 Mtonnes  $CO_{2eq}/yr$  (with a 5<sup>th</sup> to 95<sup>th</sup> range of 1.0 to 8.3 Mtonnes  $CO_{2eq}/yr$ ). 574 When added to the river loss, it suggests that, considering the total fate of carbon from soil 575 source to burial at sea, then the equivalent loss of greenhouse gas to the atmosphere for the 576 UK is 17254 ktonnes  $CO_{2eq}/yr$  (71 tonnes  $CO_{2eq}/km^2/yr$ ). 577

578 At a global scale, Van Oost et al. (2007) estimated that between 470 and 610 Mtonnes C/yr were eroded from agricultural land worldwide and, given the emissions factors 579 presented above, meant that global, agricultural soil erosion was a median net source of 3.5 580 581 Gtonnes CO<sub>2eq</sub>/yr (1.4 to 8.4). Lal (2003) estimated that global soil erosion was 75 Gtonnes/yr of which he estimated that 15-20 Gtonnes were lost to rivers and that between 4 582 and 6 Pg C/yr of carbon were lost to the oceans with 20% mineralisation of the carbon, 583 suggesting that 0.8 to 1.2 Gtonnes C was released to the atmosphere each year. Lal (2003) did 584 not consider replacement or enrichment ratio and assumed a %SOC of eroded sediment in 585 rivers of between 2 and 3% while for the UK a median value of 16% was found (Worrall et 586 al. (2014). 587

When studies argue that soil erosion can lead to carbon storage, the obvious conclusion is that soil erosion must be allowed – even encouraged - in order to store more and more carbon. Quinton et al. (2006) found that slope contouring significantly decreased, by 33%, gross soil and carbon erosion losses; other techniques were explored and, although having positive results, there were not such significant effects. Rickson (2014) reviewed 73 studies of soil erosion mitigation from the UK of which 43 quantified the effectiveness of the intervention. Rickson (2014) concluded that none of the 18 techniques were significantly different from each other and so it was impossible to select one mitigation technique over another; nevertheless, taken together, all 18 techniques decreased the soil erosion typically by 40%. Given that this study has concluded that soil erosion is likely a net source of GHGs, any decrease in soil erosion immediately represents a GHG saving. Thus, soil conservation can be shown to prevent GHG emissions to the atmosphere as well as protected terrestrial and aquatic ecosystem services.

601

#### 602 **5. Conclusions**

603 This study has shown that:

i) It is unlikely that soil erosion in the UK represents a net sink of carbon, let alone ofgreenhouse gases;

606 ii) Losses of greenhouse gases to the atmosphere are dominated by CO<sub>2</sub> (74%) followed by 607 N<sub>2</sub>O (22%) and then CH<sub>4</sub> (4%);

608 iii) The emission factor for 1 tonne of net carbon erosion is between 3.3 and 9.9. tonnes 609  $CO_{2eq}/yr$ ;

iv) The emission factor for 1 tonne of gross carbon erosion is between 1.6 and 8.9 tonnes
CO<sub>2eq</sub>/yr;

- v) The emission factor for gross soil erosion is estimated to be between 0.11 and 0.66 tonnes
   CO<sub>2eq</sub>/yr for every 1 tonnes of gross erosion;
- 614 vi) Gross soil erosion can only represent a net sink of carbon and greenhouse gases in 615 circumstances where the replacement is high (R> 0.7), the net to gross erosion rate is low 616 (<0.47) and the gross erosion rate is very low (< 91 tonnes/km<sup>2</sup>/yr).
- 617 Our results indicate that soil conservation measures are required to protect the atmosphere as 618 well as land and water. Whilst it is possible in some circumstances that soil erosion can 619 produce a net carbon sink, in most cases the effect of erosion is detrimental to the atmosphere

in terms of GHG emissions. Our study finds little evidence therefore to support a more
relaxed approach to soil erosion, indeed quite the converse. Thus, conserving soil organic
carbon provides a more extensive range of ecosystem services than might previously have
been thought, with protection not only of terrestrial, freshwater and marine ecosystems, but
also of the GHG composition of the atmosphere.

625

626

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630

## 631 **References**

Alewell C. Schaub M. Conen F. 2009. A method to detect soil carbon degradation during soil
erosion. *Biogeosciences* 6: 11: 2541-2547

Aller RC. 1998. Mobile deltaic and continental shelf muds as suboxic, fluidized bed reactors,
 *Marine Chemistry* 61: 143–155.

Aufdenkampe, A.K., Mayorga, E., Raymond, P.A., Mclack, J.M., Doney, S.C., Alm, S.R.,

Aalto, R.E., Yoo, K. 2011. Riverine coupling of biogeochemical cycles between land,
oceans and atmosphere. *Frontiers in Ecology* 9(1): 53-60.

Bao HW. Ying Z. Jing D. Bing HQ. 2015. Composition and flux of suspended organic matter

640 in the middle and lower reaches of the Changjiang (Yangtze River) - impact of the Three

- 641 Gorges Dam and the role of tributaries and channel erosion. *Hydrological Processes* 28:
  642 3: 1137 1147
- Baulch HM. Schiff SL. Maranger R. Dillon PJ. 2011. Nitrogen enrichment and the emission
- 644 of nitrous oxides from streams. *Global Biogeochemical Cycles* 25: GB4013.

- 645 Battin TJ. Kaplan LA. Findlay S. Hopkinson CS. Marti E. Packman AI. Newbold JD. Sabater
- T. 2009. Biophysical controls on organic carbon fluxes in fluvial networks. *Nature Geosciences* 1: 95-100,
- Bauer JE. Cai W-J. Raymond PA. Bianchi TS. Hopkinson CS. Regnier PAG. 2013. The
  changing carbon cycle of the coastal ocean. *Nature* 504, 7478, 61-70
- Beaulieu JJ. Arango CP. Hamilton SK. Tank JL 2008. The production and emission of
  nitrous oxide from headwater streams in the Midwestern United States. *Global Change Biology* 14: 4: 878-894
- 653 Bell MJ. Worrall F. Smith P. Bhogal A. Black H. Lilly A. Barraclough D. Merrington, G.
- 2011. UK land-use change and its impact on SOC: 1925-2007. Global Biogeochemical
  Cycles 25: GB4015
- Berhe AA. Harte J. Harden JW. Torn MS. 2007. The significance of the erosion-induced
  terrestrial carbon sink. *Bioscience* 57: 4: 337-346
- Boardman, J. 2013. Soil erosion in Britain: updating the record. Agriculture 3, 418-442.
- Bouillon S. Yambele A. Spencer RGM. Gillikin DP. Hernes PJ. Six J. Merckx R. Borges AV.
- 660 2012. Organic matter sources, fluxes and greenhouse gas exchange in the Oubangui
  661 River (Congo River basin). *Biogeosciences* 9: 6: 2045-2062
- Brockman UH. Laane RWPM. Postma H. 1990. Cycling of nutrient elements in the North
  Sea. Netherlands Journal of Sea Research 26: 2-4: 239-264.
- Burdige DJ. 2005. Burial of terrestrial organic matter in marine sediments: a re-assessment.
   *Global Biogeochemical Cycles* 19: GB4011, 1-7.
- 666 Canfield, D.E., 1989. Sulfate reduction and oxic respiration in marine sediments: implications
- 667 for organic carbon preservation in euxinic environments. Deep-Sea Res., 36: 121-138.
- 668 Canfield DE. 1994. Factors influencing organic matter preservation in marine sediments,
- 669 *Chemical Geology* 114: 315–329

- 670 Cauwet G. 2002. In"Biogeochemistry of marine dissolved organic matter"Hansell DA.
  671 Carlson CA. Ed., Academic Press, New York.
- Collins AL. Walling DE. 2007. Fine-grained bed sediment storage within the main channel
  systems of the Frome and Piddle catchments, Dorset, UK. *Hydrological Processes* 21:
  11: 1448-1459.
- 675 Crawford JT. Lottig NR. Stanley EH. Walker JF. Hanson PC. Finlay JC. Striegl RG. 2014.
- 676 CO<sub>2</sub> and CH<sub>4</sub> emissions from streams in a lake-rich landscape: Patterns, controls, and 677 regional significance. Global Biogeochemical Cycles 28: 3: 197-210
- 678 Crawford JT. Striegl RG. Wickland KP. Dornblaser MM. Stanley EH. 2013. Emissions of
- 679 carbon dioxide and methane from a headwater stream network of interior Alaska. Journal
- 680 of Geophysical Research Biogeosciences 118: 2: 482 494
- DEFRA 2005, Agriculture in the United Kingdom 2004. Department of Environment, Food
  and Rural Affairs, HMSO, London, 2005..
- DEFRA. 2005. Documenting soil erosion rates on agricultural land in England and Wales Part 2 Final Report, SP0413.
- de Haas H. Boer W. van Weering TCE. 1997a. Recent sedimentation and organic carbon
  burial in a shelf sea: the North Sea. Marine Geology144: 1-3: 131-146.
- de Haas H. van Weering TCE. 1997b. Recent sedimentation accumulation organic carbon
  burial and transport in the northeastern North Sea. Marine Geology 136:173-187.
- de Haas H. van Weering TCE. de Stieger H. 2002. Organic carbon in shelf seas: sinks or
  sources, processes and products. *Continental Shelf Research* 22: 5: 691-717
- 691 Erkens G. 2009. Sediment dynamics in the Rhine catchment: Quantification of fluvial
- response to climate change and human impact. *Netherlands Geographical Studies* 388,
- 693 Utrecht, The Netherlands.

- Evans, M.G., Warburton, J., Yang, J., 2006. Eroding blanket peat catchments: global and
  local implications of upland organic sediment budgets. *Geomorphology* 79: 45-57.
- Frances-Lenard C. Derry LA. 1997. Organic carbon burial forcing of the carbon cycle from
  Himalayan erosion. *Nature* 390: 65-67.
- Galy V. France-Lanord C. Beyssac O. Faure P. Kudrass H. Palhol F. 2007. Efficient organic
  carbon burial in the Bengal fan sustained by the Himalayan erosional system. *Nature*450: 7168,: 407-409.
- Gomez B. Trustrum NA. Hicks DM. Rogers KM. Page MJ. Tate KR. 2003. Production,
   storage and output of particulate organic carbon: Waipaoa River basin, New Zealand.
   *Water Resources Research* 39: 6: WR001619.
- Guerin F. Abril G. Richard S. Burban B. Reynouard C. Seyler P. Delmas R. 2006. Methane
   and carbon dioxide emissions from tropical reservoirs: Significance of downstream
   rivers. *Geophysical Research Letters* 33: 21: L21407
- Guo, L. Cai Y. Belzile C. Macdonald RW. 2012. Sources and export fluxes of inorganic and
   organic carbon and nutrient species from the seasonally ice-covered Yukon River.
   *Biogeochemistry* 107: 1-3: 187-206
- 710 Harden JW. Sharpe JM. Parton WJ. Ojima DS. Fries TL. Huntington TG. Dabney SM. 1999.
- 711 Dynamic replacement and loss of soil carbon on eroding cropland, *Global*712 *Biogeochemical Cycles* 13: 885-901.
- Higgins TM. McCutchan JH. Jr. Lewis WM. Jr. 2008. Nitrogen ebullition in a Colorado
  plains river. *Biogeochemistry* 89: 3: 367-377.
- 715 Higueras M. Kerherve P. Sanchez-Vidal A. Calafat A. Ludwig W. Verdoit-Jarraya M.
- 716Heussner S. Canals M. 2014. Biogeochemical characterization of the riverine particulate
- 717 organic matter transferred to the NW Mediterranean Sea. *Biogeosciences* 11: 1: 157 –
- 718 172

- Hillier S. 2001, Particulate composition and origin of suspended sediment in the R. Don,
  Aberdeenshire, UK. *Science of the total Environment* 265: 281-293.
- Hilton, R.G., Galy, A., Hovius, N., Horng, M-J., Chen, H. 2015. Efficient transport of fossil
   organic carbon to the ocean by steep mountain rivers: an orogenic carbon sequestration
   mechanism. *Geology* 39: 71-74.
- Hodgson, J.M. 1997. Soil Survey Field Handbook: Describing and Sampling Soil Profiles.
  Soil survey Technical Monograph No. 5. Soil Survey and Land Research Centre, Silsoe.
- England.
- Hoffmann T. Glatzel S. Dikau R. 2009. A carbon storage perspective on alluvial sediment
  storage in the Rhine catchment. *Geomorphology* 108: 1-2: 127-137.
- Hoffmann, T., Mudd, S.K., van Oost, K., Verstraeten, G., Erkens, G., Lang, A., Middelburg,
- H., Boyle, J., Kaplan, J.O., Willenbring, J., Aalto, R., 2013a. Humans and the missing Csink: erosion and burial of soil carbon through time. Earth Surface Dynamics 1: 45-52.
- Hoffmann, T., Schlummer, M., Notebaert, B., Verstraeten, G., Korup., O. 2013b. Carbon
  burial in soil sediments from the Holocene agricultural erosion, Central Europe. *Global*
- 734 Biogeochemical Cycles 27: 828-835.
- Houghton JT. Meira-Filho LG. Callender BA. Harris N. Kattenberg A. Maskell K. 1995.
- 736 Climate change 1995: The science of climate change. Cambridge, University Press. 339737 pp.
- Hung JJ. Yeh Y-T. Huh C-A. 2012. Efficient transport of terrestrial particulate carbon in a
  tectonically-active marginal sea off southwestern Taiwan. *Marine Geology* 315: 29-43.
- 740 IPCC. 2007. Contribution of Working Group I to the Fourth Assessment Report of the
  741 Intergovernmental Panel on Climate Change. Solomon, S., D. Qin, M. Manning, Z.
- 742 Chen, M. Marquis, K.B. Averyt, M. Tignor, and H.L. Miller (eds.). Cambridge
- 743 University Press, Cambridge, United Kingdom and New York, NY, USA.

- 744 Ittekkot V. Zhang S. 1989. Pattern of particulate nitrogen transport in World rivers. *Global*745 *Biogeochemical Cycles* 3: 383-391.
- Jenkinson DS. Rayner JH. 1977, Turnover of soil organic matter in some of Rothamsted
  classical experiments. *Soil Science* 123: 298-305.
- 748 Jha, PK. Masao M. Factors Affecting Nutrient Concentration and Stable Carbon and Nitrogen
- 749 Isotope Ratio of Particulate Organic Matter in the Ishikari River System, Japan. *Water*750 *Air and Soil Pollution* 224: 5: 1551
- Lal R. 2003. Soil erosion and the global carbon budget. *Environment International* 29, 4,
  437-450.
- Li, XX. Bianchi, TS. Allison, MA. Chapman, P. Yang, GP. 2013. Historical reconstruction of
   organic carbon decay and preservation in sediments on the East China Sea shelf. Journal
   of Geophysical Research Biogeosciences 118: 3: 1079-1093
- Lilly A. Hudson G. Bell JS. Nolan AJ. Towers W. 2009. National soil inventory of Scotland
  (NSIS\_1): Site location, sampling and profile description protocols. (1978-1988).
  Technical bulletin. Draft Version 1 2009. Macaulay Land Use Research Institute.
- Ludwig W. Probst JL. Kempe S. 1996. Predicting the oceanic input of organic carbon by
  continental erosion. *Global Biogeochemical Cycles* 10: 1: 23-41.
- Martinotti W. Camusso M. Guzzi L. Patrolecco L. Pettine M. 1997. C, N and their stable
  isotopes in suspended and sedimented matter from the Po estuary (Italy). *Water Air and Soil Pollution* 99: 1-4: 325-332
- Mayer LM. Schick LL. Allison MA. Ruttenberg KC. Bentley SJ. 2007. Marine vs.
   terrigenous organic matter in Louisiana coastal sediments: The uses of bromine : organic
   carbon ratios. *Marine Chemistry* 107: 2: 244-254
- 767 Meybeck M. 1993. Riverine transport of atmospheric carbon sources, global typology and
- budget. *Water, Air and Soil Pollution* 70: 443-463.

- Middelburg JJ. 1991. Organic carbon, sulphur and iron in recent, semi-euxinic sediments of
  Kau Bay, Indonesia. *Geochimica Cosmochimica Acta* 55: 815-828.
- Neal C. Davies H. 2003. Water quality fluxes for eastern UK rivers entering the North Sea: a
  summary of information from the Land Ocean Interaction Study (LOIS). Scienc e of the
  Total Environment 314-316: 821-882.
- Ni J. Yue Y. Borthwick AGL. Li T. Miao C, He X. 2012. Erosion-induced CO<sub>2</sub> flux of small
  watersheds. *Global and Planetary Change* 94-95:101-110
- Nixon M. 1959. A study of the bank full discharge of rivers in England and Wales.
   *Proceedings on the Institute of Civil Engineers* 12: 2: 157-174.
- Petrone KC. 2010. Catchment export of carbon, nitrogen, and phosphorus across an
  agro-urban land use gradient, Swan-Canning River system, southwestern
  Australia. *Journal of Geophysical Research-Biogeosciences* 115: G01016
- 781 Pinay G. Gumiero B. Tabacchi E. Gimenez O. Tabacchi-Planty AM. Hefting MM. Burt TP.
- Black VA. Nilsson C. Iordache V. Bureau F. Vought L. Petts GE. Decamps H. 2007.
- 783 Patterns of denitrification rates in European alluvial soils under various hydrological
- regimes. *Freshwater Biology* 52: 2: 252-266.
- 785 Quinton JN. Catt JA. Wood GA. Steer J. 2006. Soil carbon losses by water erosion:
- experimentation and modelling at field and national scale in the UK. *Agriculture*, *Ecosystems & Environment* 112: 87-102.
- Quinton JN. Govers G. Van Oost K. Bardgett RD. 2010. The impact of agricultural soil
  erosion on biogeochemical cycling. *Nature Geoscience* 3: 5: 311-314
- Reimers CE. Jahnke RA. McCorkle DC. 1992. Carbon fluxes and burial rates over the
  continental slope and rise off Central California with implications for the global carbon
  cycle. Global Biogeochemical Cycles 6: 199-224.

- Rickson RJ. (2014). Can control of soil erosion mitigate water pollution by sediments? *Science of the Total Environment* 468: 1187-1197.
- Robson, A.J., Neal, C. (1997). A summary of regional water quality for Eastern UK
  rivers.Science of the Total Environment 194-195, 15-37.
- Seitzinger SP. 1988. Denitrification in freshwater and coastal marine ecosystems ecological
   and geochemical significance. *Limnology and Oceanography* 33: 4: 702-724
- Seitzinger SP. Kroeze C. 1998. Global distribution of nitrous oxide production and N inputs
  in freshwater and coastal marine ecosystems. *Global Biogeochemical Cycles* 12: 1: 93113
- Silvennoinen H. Liikanen A. Torssonen J. Stange CF. Martikainen PJ. 2008. Denitrification
   and nitrous oxide effluxes in boreal, eutrophic river sediments under increasing nitrate
   load: a laboratory microcosm study. *Biogeochemistry* 91: 2-3: 105-116
- 805 Silvennoinen H. Liikanen A. Rintala J. Martikainen PJ. 2008. Greenhouse gas fluxes from the
- 806 eutrophic Temmesjoki River and its Estuary in the Liminganlahti Bay (the Baltic Sea).
  807 *Biogeochemistry* 90: 2: 193- 208
- Smith SV. Sleezer RO. Renwick WH. Buddemeier R. 2005. Fates of eroded soil organic
  carbon: Mississippi basin case study. *Ecological Applications* 15: 6: 1929-1940
- Stallard RF. 1998. Terrestrial sedimentation and the carbon cycle: Coupling weathering and
  erosion to carbon burial. *Global Biogeochemical Cycles* 12: 231-257.
- Striegl RG. Dornblaser MM. McDonald CP. Rover JR. Stets EG. 2012. Carbon dioxide and
  methane emissions from the Yukon River system. *Global Biogeochemical Cycles* 26:
  GB0E05
- Syvitski JPM. Vorosmarty CJ. Kettner AJ. Green P. 2005. Impact of humans on the flux of
  terrestrial sediment to the global coastal ocean. *Science* 308: 5720: 376-380

817	Tappin AD. Harris JRW. Uncles RJ. 2003. The fluxes and transformations of suspended
818	particles, carbon and nitrogen in the Humber estuarine system (UK) from 1994 to 1996:
819	results from an integrated observation and modelling study. Science of the Total
820	Environment 314: 665-713.
821	Tranvik, L.J., Downing, J.A., Cotner, J.B. Lotselle, S.A., Striegl, R.G., Ballatore, T.J., Dillon,
822	P., Finlay, K., Fortino, K., Knoll, L.B., Kortelainen, P.L., Kutser, T., Larsen, S., Laurion,
823	L., Leech, D.M., McCallister, S.L., McKnight, D.M., Melack, J.M., Overbolt, E., Porter,
824	J.A., Prairie, Y., Renwick, W.H., Roland, F., Sherman, B.S., Schindler, D.W., Sobek, S.,
825	Tremblay, A., Vanni, M.J., Verschoor, A.M., von Wachenfeldt, E., Weyhenmeyer, G.A.
826	2009. Lakes and reservoir regulators of carbon cycling and climate. Limnology &
827	Oceanography 24: 2298-2314.
828	Van Hemelryck H. Fiener P. Van Oost K. Govers G. Merckx R. 2010. The effect of soil
829	redistribution on soil organic carbon: an experimental study. Biogeosciences 7: 12: 3971-
830	3986
831	Van Oost K. Quine TA. Govers G. De Gryze S. Six J. Harden JW. Ritchie JC. McCarty GW.
832	Heckrath G. Kosmas C. Giraldez JV. da Silva JRM. Merckx R. 2007. The impact of
833	agricultural soil erosion on the global carbon cycle. Science 318: 5850: 626-629.
834	Walling DE. Owens PN. Leeks GJL. 1999. Rates of contemporary overbank sedimentation
835	and sediment storage on floodplains of the main channel systems of the Yorkshire Ouse
836	and River Tweed, UK. Hydrological Processes 13: 993-1009.
837	Walling DE. Russell MA. Hodgkinson RA. ZhangY 2002. Establishing sediment budgets for
838	two small lowland agricultural catchments in the UK. Catena 47: 4: 323-353.
839	Wang XC. Chen RF. Gardner GB. 2004. Sources and transport of dissolved and particulate
840	organic carbon in the Mississippi River estuary and adjacent coastal waters of the
841	northern Gulf of Mexico. Marine Chemistry 89: 1-4: 241-256

- Weijers JWH. Schouten S. Schefuss E. Schneider RR. Damste JSS. 2009. Disentangling
  marine, soil and plant organic carbon contributions to continental margin sediments: A
  multi-proxy approach in a 20,000 year sediment record from the Congo deep-sea fan.
  Geochimica Cosmochimica Acta 73: 1: 119-132
- Worrall F. Burt TP. Adamson JK. 2006. Long-term changes in hydrological pathways in an
  upland peat catchment recovery from severe drought? *Journal of Hydrology* 321: 5-20.
- Worrall F. Guillbert T. Besien T. 2007. The Flux of Carbon from rivers: the case for flux
  from England and Wales. *Biogeochemistry* 86: 63-75.
- Worrall F. Burt TP. Howden NJK. 2013a. The flux of suspended sediment from the UK 1974
  to 2010. *Journal of Hydrology* 504: 29-30.
- Worrall F. Clay GD. Masiello CA. Mynheer G. 2013b. Estimating the oxidative ratio of the
  global terrestrial biosphere carbon. *Biogeochemistry* 115: 1-3: 23-32.
- Worrall F. Burt TP. Howden NJK. 2014. The fluvial flux of particulate organic matter from
  the UK: quantifying in-stream losses and carbon sinks. *Journal of Hydrology* 519: 611625.
- Yan W, Laursen AE. Wang F. Sun P. Seitzinger SP. 2004. Measurement of denitrification in
  the Changjiang River. *Environmental Chemistry* 1: 2: 95- 98.

860 Table 1. The summary of C/N data used within this study.

Ref.	Region	Min.	Max.
Bao et al. (2014)	Yangtze	6.4	9.2
Higueras et al. (2014)	Rhone	2.8	14.7
Bouillon (2012)	Congo	7	10.9
Petrone et al. (2010)	W.Australia	2.8	14.7
Jha and Masao (2013)	Japan	5.3	17.8
Guo et al. (2012)	Yukon	19	35
Wang et al. (2004)	Mississippi	10	17
Martinotti et al. (1997)	Ро	6.7	8.2
This study	UK	6.7	21.4

Table 2. The percentage of the C lost to the atmosphere that was lost as CH<sub>4</sub>.

Ref.	Region	%C loss as CH <sub>4</sub>
Striegl et al. (2012)	Yukon	0.72
Crawford et al. (2014)	Wisconsin	0.81
Crawford et al. (2013c)	Yukon	1.1
Silvennomen et al. (2008)	Finland	1.2
Guerin et al. (2008)	Reservoirs	1.4

Ref.	Region	Range
Higgins et al. (2008)	Colorado river	0.17 to 4
Beaulieu et al. (2008)	Midwest rivers	upto 20.7
Silvennoinen et al. (2008)	Finland	0.02
Yan et al. (2004)	Changjiang	0 - 0.14
Baulch et al. (2011)	USA	0.75

Table 3. The percentage of the C lost to the atmosphere that was lost as CH<sub>4</sub>.

Parameter	Range	Units
Replacement rate	0.19 - 1	dimensionless
Enrichment ratio	1.2 - 4.7	dimensionless
Mineralisation	0.75 - 0.95	dimensionless
Net to gross		
POM flux at source	887 - 1504	Ktonnes/yr
Over bank sedimentation	0.97	%
Percentage of year with	0.1 - 2.9	%
overbank sedimentation		
In-channel storage	2 - 5	% of incoming POM flux
C/N	9.2 - 14.3	dimensionless
C in POM	45 - 50	%
Proportion of C loss as CH <sub>4</sub>	0.64 - 2.2	%
Proportion of N loss as N <sub>2</sub> O	0.3 - 3.0	%
Estuarine transit	16-43	% of incoming POM flux
Estuarine removal	36 - 54	% of incoming POM flux
Burial efficiency	27 - 49	%

# 869 Table 4. The range of the input parameters

Pathway	Flux (ktonnes	Export (tonnes	Flux(ktonnes
-	C/yr)	$C/km^2/yr$ )	$CO_{2eq}/yr)$
РОМ	863	3.5	
POM loss	264	1.1	998
DOM	909	3.7	
DOM loss	2650	10.9	11062
Excess CO <sub>2</sub>	598	2.5	2194
Total loss at source	5020	21.8	
Total loss to atmosphere	3512	15.2	14254

Table 5. The summary of the fluvial carbon and GHG fluxes for the UK rivers.

Figure 1. Schematic diagram of the generation and fate of particulate organic matter as

877 considered in this study.

- Figure 2.The terrigenous organic matter burial efficiency compared to the sedimentation rate.
- 881 Figure 3. Summary of flux, sinks and sources of POM (values in **bold** ktonnes/yr) and
- greenhouse gases (*values italics* ktonnes  $CO_{2eq}/yr$ ) as estimated by this study. Ranges on
- these are provided in the text.