The flux of suspended sediment from the UK 1974 to 2010

Fred Worrall[1][1], Tim P.Burt[2], and Nicholas J.K. Howden[3].

1. Dept of Earth Sciences, Science Laboratories, South Road, Durham, DH1 3LE, UK.
2. Dept. of Geography, Science Laboratories, South Road, Durham, DH1 3LE, UK.
3. Dept. of Civil Engineering, University of Bristol, Queens Building, Bristol, UK.

Abstract

The suspended sediment flux has been estimated for 270 catchments across Great Britain from 1974 to 2010: a total of 6026 catchment-years. The fluxes were corrected for inconsistencies in sampling frequencies between catchments and years and combined by a regional, area-weighted technique to give the national flux from 1974 onwards. Soil-cover, land-use and hydro-climatic variables were derived for 192 catchments for which annual average suspended sediment flux could be calculated between 2001 and 2010. The results show that:

i) Suspended sediment concentrations in UK rivers significantly declined from 1974 to 2010, but this does not cause a decline in suspended sediment fluxes: this suggests declines in concentration were mainly at low flows;
ii) Suspended sediment exports have a median of 22.2 tonnes /km^2/yr with a 5th percentile = 5.4 tonnes/km^2/yr and 95th percentile = 107.7 tonnes/km^2/yr: giving a national flux between 2,199 and 27,550 ktonnes/yr.

[1] Corresponding author: Fred.Worrall@durham.ac.uk; tel. no. +44 (0)191 334 2295; fax no. +44 (0)191 334 2301.

It was not possible to estimate the turnover of suspended sediment through the watersheds but the results indicate that the total fluvial flux of carbon from the UK terrestrial biosphere is at least 22.2 tonnes C/km^2/yr and 9.1 tonnes N/km^2/yr for the total fluvial flux of nitrogen – more than twice the previous estimates.

Keywords: rivers, nutrients, carbon.

1. Introduction

The flux of suspended sediments from rivers to marine waters is important for a number of reasons: it represents denudation rates of the land surface, but it also represents a supply of nutrients (eg. Seitzinger et al., 2005); the transport of pollutants (eg. Kroon et al., 2012); and changes in light penetration (eg. Stramski et al., 2004). Transfer of carbon in particulate organic matter can represent a sink of carbon in ocean sediment with only an estimated 20% being recycled to the atmosphere (eg. Galy et al., 2007, Masiello, 2007). Worrall et al. (2007) estimated the fluvial carbon flux from England and Wales including the flux of particulate organic carbon at the tidal limit – scaling this to the UK would give a value of 554 Mtonnes/yr (equivalent to 2.3 tonnes C/km^2/yr). However, the study of Worrall et al. (2007) used particulate fluxes published by the OSPAR Commission (OSPAR, 2007), and indeed, later studies of UK nutrient fluxes have been limited to the OSPAR Commission results (eg. Worrall et al., 2009). The OSPAR Commission is the Europe-wide body that controls the member nations reporting under the Oslo and Paris treaties that were signed to monitor the fluxes of a range of determinands and contaminants to the European continental shelf. In the UK, the nation's requirement under the Oslo and Paris treaties has been met by establishing a nationwide monitoring programme for major rivers entering coastal seas and this programme is referred to as the Harmonised Monitoring Scheme (HMS). There are several problems with this approach. Firstly, a flux of suspended solids is not a flux of particulate organic carbon and the composition of the suspended sediment had to be assumed from published studies from across the UK of suspended sediment composition (eg. Hillier, 2001). Secondly, OSPAR Commission results do not rescale their estimates for the unsampled area, even when fully reporting, HMS catchments only cover 63% of the country (Worrall et al., 2009). Thirdly, OSPAR must base its published fluxes on low frequency data which in the UK is based mainly on sampling only once a month and such low frequency sampling has been associated with considerable underestimation of fluxes. Cassidy and Jordon (2011) degraded a high-frequency record of phosphorus concentration in streamwater to show that, with decreasing sampling frequency bias of the flux estimate rose to 60% with monthly sampling (in this case estimates were 60% lower than the true value). Moater et al. (2012) considered the precision and bias of differing sampling frequencies upon the estimation of the suspended sediment flux and suggested that bias of the order of a factor of 2 would be true for monthly sampling.

A wide range of methods have been proposed for calculating river fluxes from concentration and flow data (e.g. De Vries and Klavers 1994, Littlewood 1995). These methods differ between interpolation methods (e.g. Webb et al., 1997) and extrapolation methods (e.g. Duan, 1983). When considering suspended sediment, Webb et al. (1997) considered 5 interpolation and 2 extrapolation methods and found that for suspended sediment flux estimation extrapolation methods gave the least biased results, and bias increased with decreased sample frequency. Several studies have recommended or considered adaptive strategies. Kronvang and Bruhn (1996) suggested taking samples “hydrologically” rather than on a regular basis and a number of studies (Cooper and Watts, 2002; Skarbøvik et al., 2012) have suggested including flood samples alongside regular sampling. However, the use of extrapolation and adaptive strategies is impossible when considering a dataset from a monitoring such as the HMS monitoring network in the UK where sampling is regular rather than adaptive, and often infrequent, typically monthly.

Finally, rivers are not passive conduits of carbon and nutrients, they are themselves sources and sinks. The river represents a finite travel time and the flux is being measured at the end of the transport period and not at its beginning, i.e. at the source for the majority of the carbon or nutrient in the river system. By calculating the flux at the river output, previous estimates of particulate nutrient and carbon fluxes have not been able to account for in-stream losses and have not been able to assess losses from the terrestrial biosphere. Rivers and lakes are known to be sources of CO_2 to the atmosphere (Kempe 1982, 1984) and part of that CO_2 will come from the turnover of dissolved and particulate organic carbon (DOC and POC). Similarly, nitrogen can be immobilised or released to the atmosphere. Kroeze et al. (2003) reviewed N retention in surface waters found that fluvial N retention is typically between 11 and 50% of N input. Worrall et al. (2012a and b) compared fluxes of DOC and dissolved nitrogen species from different size catchments and, by allowing for differences in catchment soil cover, land use and hydro-climatic properties, it was possible to measure the net watershed loss of DOC or dissolved nitrogen, thus the loss at source from the terrestrial biosphere could be calculated. The net watershed losses of DOC was 70% of the flux coming from the terrestrial biosphere, for the UK that loss of carbon to the atmosphere would represent 3% of the UK's greenhouse gas inventory (Cannell et al., 1999). The decline in sediment yield through a catchment has often been expressed as a sediment delivery ratio (eg. Walling et al., 1983). This decline in sediment yield has been associated with storage of suspended sediment in channel (eg. Collins and Walling, 2007) and on floodplains (eg. Walling and Owens, 2003). Although studies have considered spiralling of organic matter (as defined by ref) they have not considered loss by turnover (eg. Young and Huryn, 1997, Griffiths et al., 2012).

Within the context of improving carbon and nutrient fluxes the aim of this study is, therefore, to estimate the particulate flux from low frequency, long-term, spatially extensive datasets in order to appraise suspended sediment flux at a national scale.

2. Methodology

The approach of this study was to calculate the flux of suspended sediment for a given catchment. The flux from across the UK is then calculated in two ways. Firstly, the time series of suspended sediment from the UK was calculated as the area-weighted average of exports from the catchments for which a flux could be calculated in that year. Second, the times series of the suspended sediment flux was calculated by comparing the flux from individual catchments to physical characteristics of that catchment. By comparing suspended sediment flux to its catchment properties it is possible to compare across catchments to assess what is important in controlling the flux and by allowing for differences in land use and soil type it becomes possible to compare flux from different size catchments.

2.1. The Flux of Suspended Sediment

The study used data from the Harmonised Monitoring Scheme (HMS - Bellamy and Wilkinson, 2001). There are 56 HMS sites in Scotland and 214 sites in England and Wales (Figure 1). Rivers for monitoring were selected as the tidal limit of rivers with an average annual discharge over 2 m^3/s; in addition, any tributaries that have an average annual discharge above 2 m^3/s are also sampled. These criteria mean that there is good spatial coverage of the coast of England and Wales, but in Scotland many of the west coast rivers are too small to warrant inclusion in the HMS. No data were available from Northern Ireland. This study only considered sites where monitoring was coincident with flow monitoring, otherwise a flux calculation would be impossible. Among the monitoring agencies, sampling frequencies vary, ranging from sub-weekly to monthly or even less frequently. Annual data were rejected at any site where there were less than 12 samples in that year with the samples in separate months, in this way it was hoped that a range of flow conditions would be sampled, in general, this was the sampling scheme being followed within these national monitoring schemes.

Littlewood and Marsh (2005) proposed an interpolation method that accounts for differing sampling frequencies as sampling frequencies from the minimum this study accepted (12 samples in separate months to a maximum frequency of daily sampling):

$$F_y = K\sum_{i=1}^{n_y} n_x C_i Q_i \qquad \text{(i)}$$

$$n_x = \frac{A_y}{n_y} \qquad \text{(ii)}$$

Where: F = the annual flux at the site; C_i = the measured concentration at the site at time i; Q_i= the river discharge at time i; K = a conversion factor which takes into account the units used; n_y = the number of samples at the site in that year; and A_y = the number of days in that year, i.e. this can vary with a leap year. This approach assumes that each sample taken at a site is equally likely to be representative of an equal proportion of the year as any other sample.

The quality of methods and sampling frequencies used to calculate flux need to be considered in two ways. Firstly, the accuracy can be considered as the difference between the true load and estimated load and represents the systematic bias. Secondly, the precision of the method represents the spread of the load estimates about a certain value, in other words the consistency of the load estimates. In many studies that discuss uncertainty in flux estimation due to changing method or sampling frequency, it is the precision that is described and not the bias or accuracy. An example of this is Littlewood et al., (1998) who could only trace precision with changing sampling frequency with "indicative" curves but could not discuss accuracy of methods because there was no "true" value available. Johnes (2007) considered 17 catchments where there was daily measurement of phosphorus but had no sub-daily data and had to assume that "method 5" (Littlewood, 1995) was the true value and only considered precision but not bias. The lack of a "true" value with which to compare bedevils the assessment of precision of changing method of sampling frequencies. Cassidy and Jordan (2011), with sub-daily measurement of phosphorus, considered both bias and precision in their approach and thus showed bias with decreasing sampling frequency, with bias of up to 60% on monthly sampling, and large uncertainty for all sampling frequencies except for near continuous monitoring. Therefore, it is clear that for the type of low-frequency data available to this study, there could be considerable sampling bias, most likely leading to underestimation. In the absence of detailed, high frequency data from which a "true" value of suspended sediment flux could be calculated then this study used an alternative approach to assess how biased the annual flux estimates from this study were.

Using analysis of covariance (ANCOVA) the sampling frequency for all catchment-year combinations was compared to a flow weighted flux estimate (i.e. $\frac{F_y}{\sum Q}$). For the ANCOVA, sampling frequency was considered as a factor with four levels (sampling frequency ≤ 1 per week, ≤ 2 weeks, ≤ 3 weeks, and ≤ 1 per month). The annual water yield for each catchment-year combination ($\sum Q$) was used as the covariate. The normality of the data was tested using the Anderson-Darling test (Anderson and Darling, 1952); if the test failed at a 5% probability of the data not being normally distributed, then the data were transformed and the distribution re-tested. If there was a significant effect due to sampling frequency, *post hoc* testing using the Tukey test was used to identify where significant differences lay between factor levels. Where significant differences were found, a correction factor for that sampling frequency could be derived by comparing the mean for that level of sampling frequency relative to the other factor levels, i.e. relative to other sampling frequencies. Such a correction factor was then applied to the suspended sediment flux for each catchment-year combination given its sampling frequency. Such correction factors adjust the interpolation method results for the inconsistencies in sampling frequency for the UK and adjusts them to higher frequency observations. To illustrate this approach the catchment-year combination with the highest sampling frequency was analysed - River Rother, Hardham, Sussex, where in 1987 there was daily sampling. The time series of 365 concentration-river flow pairs was randomly degraded to give 100 time series of based upon 7, 14, 21 and 28 day sampling was considered. The annual flux from the daily sampling was then compared to estimates of the annual flux based upon the 7, 14, 21 and 28 day sampling frequency without and with the use of the correction factors derived above. As a further test of this approach the two catchments in the available dataset with the greatest contrast in base flow index (BFI – Gustard et al., 1992) were selected to give the biggest contrast in hydrological behaviour. For these two catchments - Rivers Test (BFI – 0.9) and River Thurso (BFI = 0.3) – the flux was calculated using an extrapolation method (Ferguson, 1986) based on all the available suspended sediment concentration and flow data for that catchment given that the data were made stationary over the time series of their sampling period. The fluxes calculated by extrapolation were then compared back to those calculated by interpolation with and without correction for sampling frequency derived above.

From the flux for each HMS site in each year, as adjusted by the derived correction factor, the export rate was calculated as the flux per unit catchment area per year. The flux from Great Britain was then calculated using an area-weighted average of export rates. A total regional flux was then calculated from the area weighted average export and the regional area (Figure 1). The flux from all the regions was summed to give the national flux. This regional approach better represents regional hot spots without biasing the national value due to uneven spatial distribution of available records, while at the same time using all site information in the calculation of national-scale flux. This area-weighted upscaling removes the problem of estimation of flux for the unsampled area. When considering the suspended sediment flux using the HMS records, there were no years, for any region, that were completely devoid of annual flux information. This analysis was only performed for Great Britain as no suspended flux data were available for Northern Ireland. However, the land area of Northern Ireland was known and so the results for Great Britain could be upscaled to give an estimate of the flux from the whole of the UK.

2.2. Catchment characteristics

The catchment properties considered by this study included soil, land use and hydrological characteristics. The dominant soil of each 1 km^2 grid square in Great Britain was classified into mineral, organo-mineral and organic soils based upon the classification system of Hodgson (1997) derived from nationally-available data (Smith et al., 2007, Lilly et al., 2009); note that by this definition, peat soils are a subset of organic soils. The land use for each 1 km^2 of Great Britain was classified into: arable, grass and urban based upon the June Agricultural Census for 2004 (Defra, 2005). In addition, the number of cattle and sheep in each 1 km^2 were counted within this census. The catchment area to each monitoring point for which suspended sediment flux information was available was calculated from the CEH Wallingford digital terrain model which has a 50m grid interval and a 0.1 m altitude interval. The soil and land-use characteristics based upon 1 km^2 grid square were summed across the catchment areas to the monitoring points for which flux information was available. It was then possible to express both soil and land-use properties as percentages of the catchment. For livestock, the “equivalent sheep per hectare” were calculated based upon published nitrogen export values of the respective livestock (Johnes et al., 1996) and giving a ratio of 3.1 sheep per cow. In addition, a range of hydrological characteristics for each catchment were calculated, these were: the BFI, the average actual evaporation, and the standard average annual rainfall for each catchment for which suspended sediment flux data were available from the National Water Archive (www.ceh.ac.uk). The study did not directly use the average annual total riverflow for each catchment as the difference between average annual rainfall and the average actual evaporation would be an estimate of the annual runoff for each catchment assuming reasonable water balance closure: if total riverflow is important it will be apparent from the importance of these other variables.

2.3. Statistical Modelling

Principal component analysis (PCA) was used to assess whether groups or clusters of catchments existed in the data that could invalidate interpretations of any multiple linear relationships found within the data. The PCA was carried out using percentage land use and soil characteristics, so that the influence and collinearity with catchment area was minimised. The data included in the PCA were not normalised, standardized or transformed prior to analysis and so correlation rather than covariance analysis was used. Only the principal components with an eigenvalue > 1 and the first with an eigenvalue < 1 were considered. On the basis of the results of the PCA, it was apparent that the catchments for which data were available could be divided into two groups, to understand the separation between the two groups of data logistic regression analysis was used.

Multiple linear regression was used to compare the average annual flux for the period 2001 to 2010 to catchment characteristics and was performed with both explanatory variables and the response variable untransformed and log-transformed. Normality of transformed and untransformed variables was tested using the Anderson-Darling test (Anderson and Darling, 1952). Variables were only included in the model if they were statistically significant of being different from zero at least at the 95% probability. Models were chosen both on the basis of model fit, as assessed by the correlation coefficient (r^2), and the physical-interpretability of the model. Of particular interest were models containing only those soil and land-use characteristics that could be mapped across Great Britain. Regression analysis was used to assess the relationship between average flux and the size of the catchment, to discover, if there were significant net watershed losses, this should be discernible from the relationship between total flux and catchment area. If the best-fit model included catchment area, the model was then recalculated excluding catchment area and the residuals of that model were compared to the catchment area. In using regression to filter the data for effects other than that of catchment area, care was taken to consider information that was a proxy or collinear with catchment area, e.g. area of arable land in a catchment is a collinear with catchment area. For any statistically significant model derived from the multiple linear regression, an analysis of residuals was performed where a standardised residual (residual divided by its standard deviation) greater than 2 was considered an outlier and worthy of further investigation. As further analysis of fit of preferred models, the residuals after model fitting were analysed to test for their normality using the Anderson-Darling test.

3. Results

3.1. Suspended sediment concentration

With respect to the suspended sediment concentration, there were 103,162 measurements with 91,604 measurements for which there were also measured flow and they came from 270 catchments and across all years from 1974 to 2010. The median suspended sediment concentration = 9 mg/l with a 5th percentile = 2 mg/l, and a 95th percentile = 65 mg/l. The ANOVA shows that all factors were significant explaining 56% of the original variance, with

Region being the most important factor in itself explaining 47% of the original variance. When ANCOVA was considered, log-transformed river flow was found to be a significant variable but there was only a slight increase in the amount of the original variance explained – 51%, the role of the river flow is at the expense of the proportion of the variance explained by the Region factor with the river flow explaining 4% of the original variance.

The ANCOVA indicates that a significant multiple linear regression model for suspended sediment concentration was possible:

$$ln[sedt] = \underset{(0.5)}{21.4} + \underset{(0.002)}{0.132}lnflow - \underset{(0.0002)}{0.01}year + \underset{(0.004)}{0.07}sin\left(\frac{m\pi}{6}\right) - \underset{(0.004)}{0.818}cos\left(\frac{m\pi}{6}\right)$$

$r^2 = 0.34$, n = 103,162 (iii)

Where: flow = the river flow at the time of sampling (m^3/s); year = year; and m = month number in the calendar year (1 = January to 12 = December). The implication of equation (iii) is that there is a significant decline in suspended sediment concentration with time since 1974. However, examination of the annual average for all records shows that the decline is not pronounced and shows a shift from approximately 7 mg/l before 1989 to an average annual suspended sediment concentration of approximately 6 mg/l after that. The ANOVA shows that the difference between years represents only 1.3% of the original variance and similarly the partial regression analysis shows that the variation due to secular trend represents only 1.5% of the original variance.

3.2. Catchment suspended sediment fluxes

The annual suspended sediment flux for all 270 catchments in the HMS scheme but out of a possible 9,472 catchment-year combinations a flux calculation was possible for 6026 catchment –year combinations (66%). Dividing the 6026 site-year combinations by their sampling frequency, there were 223 ≤ 1 per week, 1174 ≤ 2 weeks, 836 ≤ 3 weeks, and 3793 ≤ 1 per month. On the basis of the Anderson-Darling test, the flow-weighted annual fluxes were log-transformed before ANCOVA. Both the sampling frequency and the water yield were found to be significant, although they collectively only explained 7.5% of the original variance as there was no allowance made for differences between catchments or changes over time. The *post hoc* tests showed that within the sampling frequency factor there were no differences between ≤ 1 per week and ≤ 2 weeks but there were significant differences between these two levels and sampling at ≤ 3 weeks, and ≤ 1 per month. This pattern of significance means that there was no inconsistency for sampling frequencies up to 14 days, but that there was a significant bias for sampling frequencies greater than every 14 days. Given the *post hoc* differences, it was possible to create a correction factor for each class of sampling frequency by comparing the average for each class of sample frequency to that for sampling frequency of less than 1 week. In this way the correction factors were: $f \leq 1$ per week = 1.00, 1 week < $f \leq 2$ weeks = 1.05, 2 < $f \leq 3$ weeks = 1.34, and 3 weeks < $f \leq 1$ per month = 1.48. The result suggests that for sampling frequencies of 12 per year ($f \sim 1$ per month) the result would be 67% of the true value. Cassidy and Jordan (2011) for phosphate suggested for one site that monthly sampling was 40% of the true value and Worrall et al. (in press) found for DOC flux that monthly sampling was 48% of the true value. Moatar et al. (2012) when considering suspended particulate matter suggested monthly sampling was as little as 50% of the true value. The bias correction suggested by Philips et al. (1999) gave correction factors of the order of 10 to 20 depending upon the interpolation method used.

For the individual site –year combination with the highest sampling frequency (1987 on the River Rother at Hardham) comparing the median estimated suspended sediment fluxes from each of the sampling frequencies with that calculated from the daily sampling gives the following biases: 7 days = 0.97; 14 days = 0.79; 21 days = 0.75; and 28 days = 0.76, i.e. by sampling monthly the estimated suspended sediment flux would 76% of the value estimated from daily sampling. When the correction factors were applied the comparison became: 7 days = 0.97; 14 days = 0.83; 21 days = 1.00; and 28 days = 1.13, i.e. by sampling monthly the estimated suspended sediment flux would be a 13% overestimate compared to daily sampling, however, applying the correction factors did improve the consistency of estimates across all sampling frequencies

The second test was to compare the flux estimates after correction to those from an extrapolation approach. For the River Test the 10 year average suspended sediment flux the results were: 4820 tonnes/yr for extrapolation method; 3179 tonnes/yr for interpolation method; and 4705 tonnes/yr for the corrected interpolation method, i.e. the correction method used here gave a result that was 98% of that from an extrapolation method. For the River Thurso the 10 year average suspended sediment flux the results were: 1302 tonnes/yr for extrapolation method; 2427 tonnes/yr for interpolation method; and 5270 tonnes/yr for the corrected interpolation method. The reason for the low estimate from the extrapolation method is that rating curve for this catchment shows two distinct trends even once it had been made stationary, i.e. extrapolation methods can be unreliable for this type of data.

For catchment-year combinations that met the criteria of $f \leq 1$ month the bias corrected suspended sediment exports have a median of 22.2 tonnes /km^2/yr with a 5th percentile = 5.4 tonnes/km^2/yr and 95th percentile = 107.7 tonnes/km^2/yr. Without the sample frequency correction the suspended sediment exports have a median of 10.5 tonnes /km^2/yr with a 5th percentile = 1.5 tonnes/km^2/yr and 95th percentile = 107.7 tonnes/km^2/yr. The bias corrected results can be compared to other results for suspended sediment yield from across the UK (Table 2), and shows that the results of this study fall within the range reported for other UK catchments.

3.3. National suspended sediment flux

The area of each HMS region covered by catchments draining to sampling sites varies between 23 and 85% of the individual regions, with Scotland having the lowest area covered – mainly due to the poor representation of west coast catchments within the HMS. Conversely, the best coverage is achieved where the region is dominated by a single large river (e.g. River Thames in the Thames region – Figure 1). From 1974 to 2010 the lowest number of catchments for which a flux could be calculated in any one year was 95 catchments in 1974 to a maximum of 218 catchments in 2010. The error associated with upscaling from catchment export estimates to the regional and national scales was estimated as half the percentage difference between the values estimated from the 5th and 95th percentile exports for each region. This gives an error due to upscaling of ±15%.

The upscaling to national level shows that the flux of suspended sediment of sediment from Great Britain peaked at 27,550 ktonnes/yr in 1978 and had a minimum of 2,199 ktonnes/yr in 2003 (Figure 2) – this is equivalent to an export of between 9.6 and 119.8 tonnes/km^2/yr. Figure 2 shows the national flux without the correction for sampling inconsistencies and on average the uncorrected flux is 56% of the corrected flux, i.e. once the fluxes have been scaled up the correction factor would be approximately 2 – the same as that proposed by Moatar et al. (2012). Despite there being a significant temporal trend in the suspended sediment concentration there is no significant trend with time for the suspended sediment flux. The temporal trend in the suspended sediment concentration is relatively weak and the decrease in concentration may have been focused upon low flows which are of less importance in considering the flux. Equally, there appears to be no particular association between peaks in suspended sediment flux and wet or dry years: during the course of the monitoring the three driest years were: 1976, 1995 and 2003, with the wettest years being 1980 and 2000.

Between the years 2001 and 2010 it was possible to calculate a flux for 192 catchments for which complete land use, hydroclimatic and soil characteristics could be obtained (Figure 3). The PCA gave three principal components with an eigenvalue > 1 and the first component with an eigenvalue < 1 (Table 3). An examination of the loadings on the principal components shows that principal component 1 (PC1) has an even distribution of loadings across all variables which simply illustrates that suspended sediment flux increase with increasing catchment area: with increasing catchment area, all land use and soil areas also increase. Principal component 2 (PC2) has a high positive loading for the suspended sediment flux and area of organic soils in contrast to high negative loadings for the area of mineral soils and grass area. The 3rd principal component (PC3) is dominated by a large negative loading for grass area but not for mineral soils. Plotting PC1 vs. PC2 shows that all the suspended sediment flux data can be bounded by two trends (OA and OB – Figure 4) and such a pattern shows that analysing the dataset as a single linear equation, all be it multivariate, would be doomed to fail as it would always have to be compromise between the two bounding trends. Therefore this study considers the data can be divided into two groups as suggested by Figure 4. To understand and to separate the two groups logistic regression was applied. *A priori* the catchment data were divided as group 1 PC2 > 0 and group 2 PC2 < 0: logistic regression was then applied to find the best-fit discriminator between the two groups:

$$ln\left(\frac{\theta}{1-\theta}\right) = 1.2 - 0.027OrgMin - 0.042Org + 0.05Arable + 0.007Area \qquad \text{(iv)}$$

(0.5) (0.007) (0.009) (0.01) (0.003)

Where: θ = the probability of being in group 1; OrgMin = the area of organo-mineral soils in the catchment (km^2); Org = area of organic soils in the catchment (km^2); Arable = area of arable land within the catchment; and Area = the area of the catchment (km^2). Only variables found to be significantly different from zero at the 95% level are included in Equation (iv), the numbers in brackets beneath the equation are the standard errors in the coefficients. The odds ratios of the variables included in Equation (iv) show that no one variable is more important than any other. Equation (iv) shows 98.3% concordance with the data and indeed if the criterion for defining the groups were shifted to PC2 = 0.002, the concordance was 100%. By setting a probability of 50%, it is possible to rearrange Equation (iv) to give an inequality by which it is possible to classify an area as being more likely to be from group 1 than group 2:

$$0.027OrgMin + 0.042Org < 1.2 + 0.05Arable + 0.007Area \quad \text{(v)}$$

Mapping the group membership across the country and examining Equations (iv and v) shows that the groups are distinguished by catchments dominated by organic soils and grassland with those dominated by mineral soils and arable land use (Figure 5). As catchments increase in size they would tend to become members of group 1. For Great Britain, 165,344 km^2 was in group 1, and 78656 ~~164,655~~km^2 was in group 2. Maps of the distribution show that group 1 catchments are predominantly in the south and east of the country; conversely group 2 catchments are predominantly in the north and west. Use of PCA therefore suggests that multivariate analysis is best applied separately to these two groups of catchments as defined by the logistic regression (Equation iv).

For group 1 there are 110 catchments and the best-fit equation is:

$$SS_{flux} = 7700 + 27.1OrgMin + 35.8\ Grass \qquad r^2 = 0.42,\ n = 110 \qquad \text{(vi)}$$

$$\quad (2870) \qquad (9.0) \qquad (10.3)$$

Where: SS_{flux} = the average annual suspended sediment flux (tonnes /yr); Grass = the area of grass land in the catchment (km^2); and other variables defined as previously. Only variables that were found to be significant at least at the 95% probability of being greater than zero (5% probability of being equal to zero) were included in equation (vi), the numbers in the brackets are again the standard errors of each coefficient. The Anderson-Darling test suggested that it was not necessary to log-transform the response variable prior to construction of the linear models and log-transformation of the explanatory variables did not improve the fit of the model.

For group 2 the best-fit equation was:

$$SS_{flux} = 3000 + 20.8Org + 30.6Grass \qquad r^2 = 0.67,\ n == 82 \qquad \text{(vii)}$$

$$\quad (1100) \qquad (5.7) \qquad (4.2)$$

For neither Equation (vi) nor (vii) was the catchment area significant. Worrall et al. (2012a and b) did find a catchment area effect when considering the flux of DOC and dissolved N using the same approach for the UK and interpreted this as net watershed loss. A lack of a negative correlation with catchment area could be interpreted as no significant net watershed loss of suspended sediment. Alternatively, if only Area was considered in the correlation analysis, and no other variables were included, the following significant equations were found:

$$SS_{flux} = 7374 + 22.6Area \quad r^2 = 0.37, n== 110 \qquad (viii)$$

$$\qquad (3082) \quad (2.9)$$

$$SS_{flux} = 11.5Area \quad r^2 = 0.49, n== 82 \qquad (ix)$$

$$\qquad (1.3)$$

Equations (vi) and (vii) can be interpreted as an export coefficient type of model. Equation (vii) predicts that suspended sediment export from 1km^2 of organic soils would be 20.8 ± 5.7 tonnes /km^2/yr where the quoted error is the standard error in the coefficient. However, an export coefficient interpretation of Equations (vi) and (vii) means the predicted export for mineral soils; and urban or arable land use would be zero. As with Equations (viii) and (ix) it is possible to exclude the variables found to be significant in Equations (vi) and (vii) and only include the other land use and soil variables. When this was done, there was still no significant effect for arable or urban land use. For Group 2 there was no significant effect due to other soil types but when forced as for group 1, the following equation was significant:

$$SS_{flux} = 10240 + 47.3Min + 24.3Org \qquad r^2 = 0.32, n== 110 \qquad (x)$$

$$\qquad (3271) \quad (10.6) \quad (7.4)$$

Where: Min = area of mineral soils in the catchment (km^2); and all other variables defined as above.

For all but Equation (ix), there is a significant constant term. For example, the constant term in Equation (vi) implies that when a catchment has no organo-mineral soil or grass area, the flux of suspended sediment from the catchment would be 7700 tonnes/yr and 3000 tonnes/yr for group 2 catchments. Since there are no catchments where there is a complete absence of grass area, the constant term implies a baseline of suspended sediment flux that all catchments have. However, the smallest catchment in the study was only 32 km^2 (group 1) and 4 km^2 (group 2), i.e. the constant term could represent the flux from catchments smaller than in the study, i.e. 241 and 825 tonnes/km^2/yr respectively.

The residual analysis of Equation (vi) shows the residuals to be approximately normally distributed about zero, and applying a critical absolute magnitude to the standardised residual value (greater than 2), suggests that there are 6 catchments, 5 of which are over-predicted and 1 under-predicted. These catchments are all the largest catchments in group 1 and so it appears that Equation (vi) fits best to the medium-sized catchments. For Equation (vii) there are 3 catchments that have large standardised residuals and in each of these cases Equation (vii) over predicts. In all cases the distinguishing feature of these catchments was having less than 3 km^2 of organic soils.

Using Equation (iii) it is possible to classify each 1 km^2 grid square of Great Britain into group 1 or group 2 and then either Equation (vi) or (vii) is applied accordingly (Figure 6). Figure 6 represents the contribution of each 1 km^2 grid square to the suspended sediment flux at the tidal limit (rather than the flux leaving each grid square). Given this caveat, the map shows that the more highly grazed areas of northern England and Wales have the highest export, rather than the highlands of Scotland where grazing intensities are lower.

3.4. Suspended sediment export

The export of suspended sediment can also be compared to the catchment characteristics and the following significant relationship was found:

$$log_{10}(SS_{export}) = 2.5 - 0.76log_{10}(Area) + 0.18log_{10}(Orgmin) + 0.25log_{10}(Grass)$$

(0.2) (0.09) (0.04) (0.07)

$r^2 = 0.29$, n== 188 (xi)

This Equation implies a very similar result to those for suspended sediment flux, i.e. grassland and organo-mineral soils act as significant sources and there is a default sediment export at catchment areas less than that in the study set – in this case the default suspended sediment export at the 1 km^2 would be 316 tonnes/km^2/yr. the export decreases with increasing catchment area but at this rate of export decline the actual flux increases with catchment area.

The relatively poor fit of Equation (xi) can be explained by examining the plot of export versus catchment area. Figure 7 shows that there is not one trend of declining export with area but rather all the study catchments are bound by two trends. Trend A and B are distinguished by their difference in the rate of decline of export with increasing catchment size but both have a common point at point O. The equations of the bounding trends are:

Trend A:

$$log_{10}(SS_{export}) = 6.5 - 2.63log_{10}(Area) \quad \text{(xii)}$$

Trend B

$$log_{10}(SS_{export}) = 3.29 - 0.44log_{10}(Area) \quad \text{(xiii)}$$

Note that from Figure 7 Trend B only applies for catchment area > 30 km^2 while trend A applies the across the entire range available. Trend A implies a suspended sediment export of

1,950 tonnes/km^2/yr at source and both trend A and trend B imply that the greatest change in export occurs over the first 70 to 100 km^2 of a catchment.

4. Discussion

Has this been a reasonable approach to upscaling suspended sediment fluxes? Firstly, it is a better estimate than previous attempts (OSPAR Commission, 2004). Worrall and Burt (2007) have already pointed out that the reported fluxes are an underestimate of fluxes because they do not allow for the unsampled catchments and in the UK the sampled catchments of the HMS only represent just over 60% of the UK land area and no correction for river flows from the other 40% was made: this study used an area-weighted approach to correct for that error. It would be possible to suggest that many of the fluxes calculated under these schemes are less than 50% of the true value because of sampling bias that arises from such low frequency sampling. Secondly, the correction factors derived from the statistical analysis above are similar magnitude to those reported elsewhere (eg. Cassidy and Jordan, 2011; Moatar et al. 2013). Thirdly, the results for individual catchments are similar to those reported from previous studies (Table 2). Fourthly, when compared to the highest frequency data the correction method performs well and as well, if not better, than extrapolation methods. This is not to say that further improvements could not be made and each individual catchment could be studied to find the best solution for that particular record and for that particular catchment given its inherent behaviour and sampling frequency, although such an approach may not have any objective criteria for assessing which is the correct approach for any given catchment. Equally, there is no doubt that should sub-daily data become available for each of the 270 catchments in this record then it would provide a better estimate, but very high frequency sampling is not yet available for more than a few intensively studied catchments and then there would be the question of how applicable it would be for data back in the

1970s. Therefore, we think we are justified in claiming this is an improved method that produces consistent suspended sediment fluxes when amalgamated for these large catchments and at the national scale and certainly an improved method for dealing with sparse, low frequency and historical monitoring records. A limitation of this study is that it is only suspended sediment flux that is being estimated and no estimate is made of bedload transport and flux. In the relatively low gradient rivers of the UK bedload flux is likely to be a small proportion of total sediment flux. Foster and Walling (1994) measured bedload flux to be 21% of the total sediment flux for a lowland grass-dominated catchment while Labadz et al. (1991) found a proportion upto 14% of the total sediment load for eroding Peatlands in northern England.

The fluxes of suspended sediment from the UK are generally considered low with respect to other areas of the world (Foster and Lees, 1999). Beusen et al. (2005) reviewed suspended sediment exports from 124 catchments from around the world with the largest export for the River Hai Ho in China (245,000 km^2) at 2687 tonnes/km^2/yr. For catchments of equivalent size to the area of the Great Britain (200,000 to 300,000 km^2) the review of Beusen et al. (2005) shows exports between 2 and 2,687 tonnes/km^2/yr. It is perhaps not surprising that the UK generally has a low sediment export relative to more vulnerable locations like the Chinese loess plateau (eg. River Hai Ho).

Given the new estimates of the suspended sediment flux from the UK, how has this changed the estimates of nutrient and carbon fluxes? Over the period 1990 to 2002, Worrall et al. (2007) used data from the OSPAR Commission (OSPAR Commission, 2004) to give the flux of suspended solids for England and Wales; and rescaling the value for UK gives values of the suspended sediment flux of between 1,828 – 4,874 ktonnes/yr (equivalent to between 7.9 and 21.2 tonnes/km^2/yr) which is even lower than the range predicted for the fluxes estimated without sampling frequency correction. The reason for the very low values estimated for OSPAR (2004) is because they do not re-scale their estimates for the unsampled area, and even when fully reporting, HMS catchments only cover 63% of the country (Worrall et al., 2009). The suspended sediment concentration can vary in its organic carbon content; for British rivers, Hope et al. (1997) give a preferred value of 14%, while Hillier (2001) measuring the carbon content of suspended sediment from throughout the River Don in Scotland (catchment area = 1,320 km^2) reported values that varied between 6.9 and 14.1%. Neal (2003) studied sediment from rivers with catchment areas from 373 to 8,231 km^2 and reported organic carbon contents to vary from 5 to 17%. As the latter study is the most comprehensive in terms of area and river type, this study will calculate POC flux considering a range of organic carbon content of suspended sediment of 5 – 17% with a median value of 11% being taken as the preferred value. Given the assumption of carbon content of between 5 – 17% as stated above, previous studies have estimated that the POC flux in 2002 was between 375 and 1,088 ktonnes/yr (equivalent to between 1.63 and 4.73 tonnes C/km^2/yr - Table 4), with a preferred value of 2.27 tonnes C/km^2/yr based upon a 11% carbon content. Given the assumed values of carbon content of the suspended sediment, this study would calculates that POC flux from the UK would range between 242 and 3,031 ktonnes C/yr (equivalent to between 1.1 and 13.2 tonnes C/km^2/yr) with a median value of 1,042 Ktonnes C/yr (4.3 tonnes C/km^2/yr) and that there has been no change in POC flux from the UK since 1974. Summarising data for 2002 (Table 4) and using the most recent estimates from Worrall et al. (2012a) suggests that the loss of carbon from the terrestrial biosphere via the fluvial pathway is almost 5 Mtonnes C/yr at an export of 22.2 tonnes C/km^2/yr.

Similar to the discussion for fluvial fluxes of carbon, it is possible to update estimates of fluvial nitrogen fluxes. Hillier (2001) studied suspended sediment throughout the River Don catchment in Scotland (area = 1,320 km^2); the average C/N ratio was 8.1 with a range of 5.2 (n=13). Given these composition data, it is possible to suggest that the median value of PON flux from the UK would be 129 ktonnes N/yr (0.53 tonnes N/km^2/yr – Table 5) and that the total fluvial flux of nitrogen from the UK terrestrial biosphere would be equivalent to 2,222 ktonnes N/yr (equivalent to 9.1 tonnes N/km^2/yr). The OSPAR data (OSPAR Commission, 2004) as utilised in Worrall et al. (2007) for annual suspended sediment flux shows a significant upward trend between 1992 and 2002: no such significant trend was found in this study.

Calculating the export to the oceans did not aid in the estimation of losses within the fluvial system. Walling et al. (2002) working in 1.5 km^2 and 3.6 km^2 agricultural catchments on mineral soils in England showed soil erosion rates up to 466 tonnes/km^2/yr with 80% removal, or deposition, by the outlet of the catchment – a sediment delivery ratio of 20% (Walling et al., 1983). Defra (2005b) gave median values of net soil loss from arable fields in England as 410 tonnes/km^2/yr and from English grasslands as 60 tonnes/km^2/yr. Worrall et al. (2011) gave a value of 406 tonnes/km^2/yr for a bare peat plot in the South Pennines. These values are in line with several of the estimates of sediment loss to the stream network at source made above but would not support a value as high as 1,950 tonnes/km^2/yr (derived from Equation xiii). Given the ranges of erosion reported for the UK and, if we accept a value of 241 tonnes/km^2/yr for group 1 catchments and 825 tonnes/km^2/yr for group 2, we define a loss of suspended sediment at source as 93.2 Mtonnes/yr from the UK and a delivery ratio of between 2 and 29% with a median of 10%. This large apparent buffering capacity of the UK is one possible explanation of why no trend in suspended sediment flux has been observed for the UK since 1974. A lack of trend in suspended sediment flux does not support the view of Bellamy et al. (2005) that there were increased losses of soil organic carbon from the UK, a large part of which was proposed to be due to increased fluvial flux of carbon.

An improved estimate of the total flux of suspended sediment at the tidal limit is not an improved estimate of the loss within the watershed. Moody et al. (in press) have shown in laboratory experiments that between 3.8 and 8.7%/day of peat-derived POC was lost from a peat headwater over a 10 day period. Griffiths et al. (2009) found a rate of loss of 1.5%/day for coarse organic particles (maize leaves). If it is assumed that suspended sediment remains suspended throughout its travel time and that the in-stream residence time for the UK is approximately 1 day (Moody et al., 2013) then the loss of carbon to the atmosphere per year would be between 10 and 87 ktonnes C/yr (equivalent to 0.04 and 0.4 tonnes C/km^2/yr).

5. Conclusions

This study has shown that:

i) The flux of suspended sediment from the UK has varied between 2,199 ktonnes and 27,550 ktonnes/yr – equivalent to between 9.6 and 119.8 tonnes /km^2/yr. There was no significant trend with time since 1974 in the national flux of suspended sediment to the coastal shelf.

ii) The UK could be divided into two types of catchment dependent upon the catchment area, extent of land use and soil type.

iii) Within each group of catchments, the suspended sediment flux was controlled by the soil type and in particular the presence of grazing that leads to the highest being from grazed, organic soils.

iv) Compared to many parts of the world, the UK has low suspended sediment export and source erosion rates with a sediment delivery ratio to the coastal waters of 10%.

The loss of suspended sediment through the UK's fluvial network implies a fluvial export from the terrestrial biosphere of 22.2 tonnes C/km^2/yr and 9.1 tonnes N/km^2/yr.

Acknowledgements

The authors are grateful to Abby Lane and Sarah Wheater of the Environment Agency of England and Wales for supplying the HMS data.

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Figure 1. Location of monitoring points for which a suspended sediment export could be calculated for the period 1974-2010 separated by the regions used for area-weighted averaging of fluxes.

Figure 2. The annual suspended sediment flux from the UK from 1974 comparing the uncorrected and corrected time series.

Figure 3. The distribution of the average annual suspended sediment export for the country.

Figure 4. Comparison of principal component 1 vs. principal component 2 for the average suspended sediment flux from 2001 to 2010 in comparison to catchment properties.

Figure. 5. The distribution of catchments classified as group 1 (●) and group 2 (✚).

Figure 6. The project map of suspended sediment export to the tidal limit.

Figure 7. The comparison of the suspended sediment export and the catchment area.