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The distribution of ($^{234}U/^{238}U$) activity ratios in river sediments

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Abstract :

Uranium (U) isotopes can be used to estimate the comminution age of sediments, i.e. the time elapsed from sediment production on continents, via weathering and physical erosion, to deposition in the sedimentary record. The calculation of this comminution age is based on measured (234U/238U) activity ratios in river sediments, and inferred time-dependent recoil effect, which leads to the preferential release of 234U from mineral lattices during erosion processes. In this study, we report on a large-scale (234U/238U) investigation of modern river sediments worldwide, with the aim to determine the extent to which parameters such as grain size, lithology, weathering, climate and geomorphology may influence the distribution of U isotopes in fine-grained sediments. Our extensive dataset (N=64) includes U isotopic measurements for many of the world's largest rivers, but also rivers draining particular climatic and geological settings. Our results indicate that sediments collected from river basins draining mostly igneous, metamorphic or volcanic rocks often display (234U/238U) ratios >1, with clay-size fractions (<4 µm) being less depleted in 234U (higher 234U/238U) than corresponding silt-size fractions (4-63 µm). In contrast, sediments derived from multi-lithological basins or draining sedimentary rocks are typically characterized by (234U/238U) ratios <1, with clays generally exhibiting more depleted 234U signatures than silts. Taken together, these observations suggest that the formation of secondary clay minerals in soils from basins draining mostly igneous, metamorphic is accompanied by partial incorporation by recoil injection of 234U initially released during weathering processes, possibly from U-rich minerals, such as sphene or apatite. Instead, in multi-lithological catchments draining sedimentary rocks, we propose that the erosion of recycled sediments having experienced several cycles of weathering, possibly over glacial-interglacial timescales, could explain the much lower (234U/238U) ratios observed in clay-size fractions. While no direct relationships can be identified between sediment (234U/238U) ratios and lithology, weathering intensity, climatic or geomorphic parameters in corresponding river basins, we show that the catchment size probably plays an important role in controlling the distribution of (234U/238U) in river sediments, through its direct influence on the sediment residence time. Finally, a multiple regression analysis of our data, combining various environmental parameters for the lithology, climate and geomorphology of studied river basins, indicates predicted (234U/238U) values that are very similar to measured values (with R2 ~ 0.8). This finding provides further support for the usefulness of (234U/238U) ratios in the sedimentary record for reconstructing past landscape changes and their effect on sediment transport and residence time in river basins.

1. INTRODUCTION

The morphology of the Earth's surface results from complex interactions between climate, tectonic and weathering (e.g. Dixon et al., 2009; West et al., 2005). Understanding the factors

affecting landscape evolution is essential to predict the consequences of future climate change on soil resource availability and sediment discharge to the oceans. Additionally, the study of sediment transport processes and their timescales can be useful to quantify soil production and denudation on continents. Over the past decades, the development and application of novel geochemical proxies to river materials has significantly improved our understanding of the sedimentary and weathering processes occurring in river catchments (Négrel et al., 1993; Gaillardet et al., 1997, 1999; Vigier et al., 2001; Clift et al., 2002; Bindeman et al., 2019; Bayon et al., 2016, 2018, 2020). However, the quantification of the timescale of these processes, such as the sediment residence time, still remains challenging (DePaolo et al., 2006; Romans et al., 2016). On the continents, the fractionation of uranium (U) isotopes is initiated in the Critical Zone, associated with the preferential loss of ^{234}U relative to ^{238}U, whenever chemical weathering and rock fracturing start affecting the bedrock. In theory, the degree of U-series fractionation in sediments can provide direct temporal constraints on the timing of sediment formation in soils and subsequent storage within the catchment and the sedimentary system (Chabaux et al., 2003; DePaolo et al., 2006), which can be used to determine the so-called *comminution ages*, corresponding to the time elapsed since the production of small mineral grains (typically <63 μm).

In detail, the preferential depletion of ^{234}U relative to ^{238}U in mineral grains occurs during the decay of ^{238}U to ^{234}Th, where the daughter is recoiled (and subsequently decayed into ^{234}U) within the outer ~10-30 nm periphery of the grains (Chabaux et al., 2008; DePaolo et al., 2006; Dosseto et al., 2010; Hashimoto et al., 1985; Kigoshi, 1971; Lee et al., 2010; Maher et al., 2006a). As a consequence, soil solutions and river water are typically enriched in ^{234}U, exhibiting $(^{234}U/^{238}U) > 1$ (parentheses denote activity ratio), whilst fine-grained residual sediments in soils and suspended particulate loads in rivers display values <1 (Carl, 1987; DePaolo et al., 2006;

Maher et al., 2004, 2006b; Moreira-Nordemann, 1980; Plater et al., 1988). The measurement of ^{234}U depletion is only significant (i.e. measurable) in fine-grained (<63 μm) sediments (DePaolo et al., 2006; Dosseto et al., 2006a, 2008a) where the surface/volume ratio is large enough so that the thin outer layer through which preferential ^{234}U loss occurs is significant compared to the rest of the grain (which experiences no net loss of ^{234}U) (DePaolo et al., 2006). In theory, the comminution age method is based on two main assumptions: 1) the time-integrated recoil effect represents the main factor controlling ($^{234}U/^{238}U$) in fine-grained sediments (DePaolo et al., 2006); and 2) the pristine (unweathered) rocks on continents are at secular equilibrium with regard to U-series (i.e. with activity ratios = 1) (DePaolo et al., 2012). However, a few studies have reported evidence that weathering processes can locally influence the degree of U-series disequilibrium in fine-grained sediments (Li et al., 2016), or that the bedrock may depart from the secular equilibrium (Handley et al., 2013a), hence casting doubt on the validity of the above-mentioned assumptions. In fact, despite recent increasing interest for the use of U isotopes for tracing Earth surface processes (DePaolo et al., 2006, 2012; Dosseto et al., 2008a, 2010, 2014, 2015; Handley et al., 2013b; Lee et al., 2010; Li et al., 2017a), the controls on the ($^{234}U/^{238}U$) of river sediments are yet to be fully understood. To date, all previous studies focusing on U-series as tracers of continental erosion were dedicated to the case study of specific river basins, often yielding contrasted comminution ages and inferred sediment transport times. For instance, investigations conducted at the scale of the Amazon Basin resulted in a large range of sediment residence time from the Andean tributaries (3-4 kyr; Dosseto et al., 2006b, 2008) and the lowland tributaries (100-500 kyr; Dosseto et al., 2006b). The Ganges tributaries have also been investigated in detail (Chabaux et al., 2006, 2012; Granet et al., 2007, 2010), with inferred sediment residence times ranging between 30-350 kyr. Other case studies include the Mackenzie River (Vigier et al., 2001)

and smaller river systems in East Asia (Zhuoshui River, Lanyang River; Li et al., 2016), yielding sediment transfer times of ~25 kyr and ~110 kyr, respectively. More recently, Li et al., (2016) also determined relatively long sediment transfer times (from 250 to 600 kyr) for the Changjiang (Yangtze) River basin. These latter sediment residence times are surprisingly long considering the observed variability of the provenance of the sediment exported from the Yangtze River over the past 400 kyr, which implicitly suggests a much shorter storage time within the watershed (Beny et al., 2018).

An important pre-requirement for quantifying the timescale of sediment transport in continental watersheds using U isotopes, is to assess whether the distribution of ($^{234}U/^{238}U$) in river suspended loads and sediments can be affected by other parameters, such as grain-size, lithology, climate and tectonic settings. Early works pointed out at the importance of removing non detrital sediment fractions prior to analysing sediment samples (e.g. Lee, 2009; Martin et al., 2015; Francke et al., 2018) and demonstrated the influence of grain size on U activity ratios and calculated sediment transfer times (Dosseto et al., 2014; Granet et al., 2010). More recently, Bosia et al. (2018) also emphasize the importance of analysing separate mineral phases. While it can be difficult to separate different mineral fractions from fine-grained sediments, one alternative option is to focus instead on particular grain-size fractions. In this study, we report ($^{234}U/^{238}U$) measurements for both clay (<4μm) and silt (4-63μm) size detrital fractions extracted from an extensive set of river sediment samples worldwide. This study builds upon previous investigations of the same suite of sediment samples, which have proven particularly useful for identifying the various external factors (e.g. climate, lithology, weathering regime) that control the distribution of geochemical proxies in sediments (Bayon et al., 2015, 2016, 2018, 2020; Bindeman et al., 2019).

Such a proxy evaluation study is of utmost importance prior to applying U isotopes in ancient sedimentary records for reconstructing past sediment transfer dynamics through time.

2. METHODS

2.1 Samples

A total of 64 sediment samples were analysed during this study, collected from river catchments, estuaries, or submarine deltas near the mouth of rivers (Bayon et al., 2015, 2016, 2018, 2020). All studied samples correspond to modern or relatively recent sediments presumably deposited during the last few centuries. A few sediment samples (n=17) come from igneous/metamorphic terranes from the Precambrian cratons of North America (Canadian Shield), northern South America (Guiana Shield), Africa (West African and Congo Shields), Fennoscandia, northwest Ireland and a small river from the Hercynian Armorican Massif in France (Elorn River). Seven samples are derived from rivers draining both modern (Indonesia) and ancient (British Tertiary, Northern Ireland) volcanic provinces. The rest of studied samples (n=40) correspond to rivers draining 'mixed/sedimentary' formations including some of the world's major rivers (e.g. Amazon, Congo, Mississippi, Nile, Niger, Yangtze, Mackenzie, Volga, Murray, Orinoco), plus rivers draining sedimentary basins (e.g. Adour, Shannon).

The studied river catchments can also be classified according to their climate setting (Bayon et al., 2016, 2018), depending on their mean annual temperature (MAT), and mean annual precipitation (MAP). A total of 10 samples are representative of cold and dry regions with MAT < 8°C and MAP < 800mm (e.g. Don, Fraser, Lule). Other sediments correspond to temperate and warm dry environments (e.g. Murray-Darling, Yellow River; n=18), with MAT > 10 °C and MAP < 800 mm, or from temperate and humid regions (n=18), with 8 °C < MAT < 16 °C and MAP >

1000 mm (e.g. Yangtze, Rhine). Finally, the rest of the studied samples come from sub-tropical humid regions, with MAT > 20 °C and MAP < 1500 mm (e.g. Niger, Mekong, n=6) or from tropical regions with MAT > 20 °C and MAP > 1500 mm (e.g. Amazon, Congo, Red River; n=12).The studied sediments can also be grouped using physical basin characteristics, such as lowlands, characterized by a maximum elevation of 500 m (e.g. Clarence, Swilly; n=21), uplands, with a maximum of elevation between 500 and 800 m (e.g. Severn, Shannon; n=6), mountainous regions (n=19 ; e.g. Loire, Fortescue), with maximum elevation between 800 and 3000 m, and high mountains (n=18), with maximum elevation >3000 m (e.g. Ganges, Danube).

2.2 Analytical procedures

Three distinct U pools can be distinguished in any sediment sample (Martin et al., 2015): 1) a non-detrital pool hosted by various carbonate minerals, Fe-Mn oxyhydroxide phases, and organic compounds, associated typically with enriched (($^{234}U/^{238}U$) >1) signatures (Plater et al., 1988; Andersson et al., 1998, 2001; Maher et al., 2006a); 2) a depleted (($^{234}U/^{238}U$) <1) detrital pool corresponding to the outer part of detrital minerals that have experienced partial dissolution and preferential loss of ^{234}U due to recoil effect; and 3) a pristine detrital pool at secular equilibrium (($^{234}U/^{238}U$) = 1), which corresponds to the inner part of detrital grains that has not experienced ^{234}U loss.

In this study, sequential leaching was conducted on all samples in order to remove any biogenic, authigenic and organic components prior to preparation for U isotopic measurements. First, the <63 μm fraction of the bulk sediment was recovered by wet sieving. About ~3 g of dry bulk sediment were treated successively with diluted acetic acid (AA), a mixed solution of 15% (v/v) AA and 0.1 M hydroxylamine hydrochloride (HH), and diluted hydrogen peroxide, in order to remove any carbonate, Fe-Mn oxyhydroxide and organic phases, respectively (Bayon et al.,

2002). Finally, clay- (<4 μm) and silt- (4-63 μm) size fractions of the residual detritus were separated by centrifugation using the two-step protocol reported in Bayon et al. (2015).

A series of experiments was conducted on different sediment samples in order to assess the reliability of our leaching protocol for efficiently removing the radiogenic non-detrital component of the sediment characterized by high ($^{234}U/^{238}U$) ratios. To this purpose, following the approach previously developed by Lee (2009), the effect of each leaching step of our protocol was successively assessed by measuring U isotopic ratios in both leachates and corresponding residual phases from two sediment samples from the Loire and Clarence rivers (Fig. 2). To assess whether the leaching was completed, an additional final leaching step termed "carbonate emphasized" was performed at the end of this series of experiments. The validity of our sequential leaching protocol was also further assessed by analysing two sediment samples (1B-26H-cc and 1B-17H-cc) previously studied by Francke et al. (2018) using a different leaching method. Finally, two certified reference rock materials (BCR-2, BHVO-2) were processed using the same experimental protocol used in this study, to investigate whether sequential leaching could also lead to any particular U isotopic fractionation.

Both clay- and silt-size residual detrital fractions were digested by alkaline fusion (Bayon et al., 2009a), to ensure complete dissolution and provide reassurance that the observed ($^{234}U/^{238}U$) differences between silt and clay samples cannot be possibly caused by the partial dissolution of any accessory minerals. About 50 mg dry powdered sediment were placed into a glassy carbon crucible, after addition of ^{236}Uspike, and digested at 650°C (12 min) together with NaOH and Na_2O_2. Subsequent addition of ultrapure water to the obtained melt results in the formation of iron oxide precipitates, leading to trace element scavenging (including U). Note that the good homogenization of the sediment powder and the spike during sample treatment is shown by the

validity of measured U isotopic compositions in both rock standard and certified materials. After centrifugation, U-bearing Fe-oxyhydroxide phases were rinsed twice in ultrapure water and dissolved in 2 mL of 7.5 M HNO_3.

Uranium was isolated by ion exchange chromatography (AG1X-8 resin) using a protocol adapted from Bayon et al. (2009b) and Edwards et al. (1986), which was repeated once to ensure complete U purification. The analyses were performed at the Pôle Spectrométrie Océan (Brest) on a Neptune Multi-Collector Inductively-Coupled Plasma Mass Spectrometer (MC-ICP-MS), using an APEX HF desolvating system. Instrumental mass correction was performed by standard bracketing of IRMM 184 standard solutions analysed every two samples.

The external reproducibility and accuracy of measured uranium isotopic ratios were assessed through repeated analyses of various reference (NBL CRM U005-A) and rock (BCR-2, BHVO2, JSd-2) standard solutions, with results being in good agreement (within the error range) with references from the literature (Table 1). The uncertainty on measured ($^{234}U/^{238}U$) due to sediment sampling and processing was further assessed by analysing three sediment samples collected at distinct locations in the Loire Estuary (Table 2), yielding a mean ($^{234}U/^{238}U$) value 0.959 ± 0.001 (2σ error) and 0.953 ± 0.001 (2σ error) for silt- and clay-sized fractions, respectively. Total procedural blanks were systematically <65 pg, hence negligible compared to the amount of analysed U for each studied sample (~40 ng).

3. RESULTS

3.1 Leaching experiments

The measurements of ($^{234}U/^{238}U$) in the Clarence and Loire samples show that the residual detrital ($^{234}U/^{238}U$) compositions decreased progressively during the course of the sequential

leaching procedure (from 1.0 to 0.8; Fig. 2), especially for the case of Clarence River sample, indicating that the non-detrital ^{234}U-enriched phases have been efficiently removed during our leaching procedure. Importantly, the final leaching steps were not accompanied by any ($^{234}U/^{238}U$) increase in the residual detrital fractions, suggesting negligible dissolution of pristine detrital minerals at secular equilibrium. Furthermore, the comparison of the ($^{234}U/^{238}U$) values obtained for the two test samples using our protocol agree well (>5% of difference) with the data obtained using the procedure of Francke et al. (2018) (Table 1). Finally, the ($^{234}U/^{238}U$) ratios determined on the geological reference materials following our leaching procedure also agree well with the recommended values (Table 1).

3.2 Uranium in river sediments

Measured U concentrations in studied clay- and silt-sized fractions range from 0.37 ppm (Glenariff) to 9.90 ppm (Lule), and from 0.22 ppm (Glenariff) to 10.11 ppm (Benue), respectively (Fig. 3, Table 3). In both silt and clay size fractions, the mean concentration values (~3.0 ppm and 3.3 ppm respectively) are higher than average estimate values for the upper crust continental – UCC (from 2.2 to 2.8 ppm, Condie, 1993; McLennan, 2001). The great majority of studied river sediment samples display slightly lower U concentrations in silts than in corresponding clay fractions. Note that no particular relationships were observed between U concentrations and the various parameters (e.g. climate, weathering, tectonic settings) that will be discussed in the sections below.

The ($^{234}U/^{238}U$) ratios in clay and silt fractions range from 0.819 (Rhine) to 1.340 (Swilly) and from 0.897 (Mackenzie) to 1.152 (Murchinson), respectively (Fig. 3). Our data indicate a much larger ($^{234}U/^{238}U$) variability amongst studied clay-sized fractions than in corresponding silts with a standard deviation of 0.14 and 0.05 respectively (Fig. 3, Table 3). The mean ($^{234}U/^{238}U$) value is

lower in silt than cay size fraction, 0.987 ± 0.002 (2σ error) and 1.001 ± 0.003 (2σ error) respectively. Interestingly, many sediment samples (n=21 silts; n=27 clays) display ($^{234}U/^{238}U$) values above secular equilibrium (>1).

4. DISCUSSION

4.1 Influence of grain size on ($^{234}U/^{238}U$) ratios of detrital sediments

As modelled by DePaolo et al. (2006), the recoil loss factor is directly proportional to grain size, leading to separate time-dependent evolution of ($^{234}U/^{238}U$) ratios in separate size fractions of the sediment. The influence of grain size on ($^{234}U/^{238}U$) was also assessed by Dosseto et al. (2014), who analysed various size fractions of river suspended particulates from the Murray-Darling Basin in south-eastern Australia, ranging from the dissolved load (<10 kilo-daltons - kDa) to coarse silts (>25 μm). The above-mentioned studies showed that sedimentary ($^{234}U/^{238}U$) ratios typically decrease with increasing grain size. To some extent, this 'grain-size' effect could explain the observed large range of sediment residence time determined in different river systems (Granet et al., 2010).

In the studied sediment samples, the silt fraction generally represents between 60 and 95% of the bulk (< 63μm) detrital fraction (mean ~ 82 ± 23% 2SE; Bindeman et al., 2019), meaning that the ($^{234}U/^{238}U$) values of the silt-size fractions can be taken, to a first approximation, as representative of the bulk ($^{234}U/^{238}U$) composition of the fine-grained detrital sediments investigated in this study.

A striking feature of our results is the fact that many clay-sized fractions are characterized by activity ratios higher than >1, often exhibiting higher values than in corresponding silts. As mentioned above, this observation is opposite to the grain-size effect predicted theoretically by

DePaolo et al. (2006). In previous studies, samples exhibiting activity ratios >1 were generally discarded, because they were thought to be affected by the presence of radiogenic authigenic components (Dosseto et al., 2006b; Granet et al., 2010; Martin et al., 2019; Vigier et al., 2001, 2006). However, we are confident here that our clay-sized fractions solely correspond to detrital material, previously leached from any possible carbonate, oxide and/organic components. On this basis, we propose that measured activity ratios above 1 could reflect partial incorporation of ^{234}U previously released from incongruent silicate weathering into neoformed clays (Dequincey et al., 2002; Plater et al., 1992).

4.2 The effect of weathering, climate and erosion on ^{234}U-^{238}U fractionation

The disequilibrium between ^{234}U and ^{238}U in soils mainly reflects the recoil effect during water/rock interactions (Osmond and Ivanovich, 1992; Riotte et al., 2003) and the preferential leaching of ^{234}U relative to ^{238}U embedded in recoil tracks (Fleischer, 1980, 1982). It is important to investigate to which extent weathering could influence the distribution of ($^{234}U/^{238}U$) ratios in river sediments and eventually lead to an over- or underestimation of the sediment residence time in watersheds. In theory, bulk mineral dissolution should not lead to any significant fractionation between ^{234}U and ^{238}U (Chabaux et al., 2003), so that chemical weathering intensity is not expected to have direct influence on sediment ($^{234}U/^{238}U$). However, Li et al. (2016) reported various correlations between ($^{234}U/^{238}U$) and chemical weathering indices in leached river sediments (<50 μm), suggesting a link between U-isotope fractionation patterns and the type of weathering regime. Lower ($^{234}U/^{238}U$) values (<0.90), indicative of presumably longer sediment residence times, were determined in sediments having experienced intense chemical weathering in transport-limited regimes. In contrast, in weathering- (or kinetically) limited weathering regimes associated with high denudation rates and fast sediment transfer, Li et al. (2016) proposed that high mineral

dissolution rates could result in sedimentary ($^{234}U/^{238}U$) ratios being close to 1. To some extent, this latter observation is counter-intuitive, because enhanced physical erosion in high mountain environments and other kinetically-limited weathering regimes could be alternatively associated with sediments having lower ($^{234}U/^{238}U$) ratios, as a result of enhanced production of small grain-size sediments and associated ^{234}U recoil loss. For these reasons, and because the chemical weathering signature of the sediments can be partly inherited from previous weathering cycles and sediment recycling (Dou et al., 2016), but also because the ($^{234}U/^{238}U$) ratio only records the last million years of chemical weathering, Li et al. (2016) remained cautious with the interpretation of the relationships between weathering indices and ($^{234}U/^{238}U$).

In our study, the ($^{234}U/^{238}U$) composition of both silt-and clay size fractions does not display any relationships weathering indices, such as the Chemical Index of Alteration (CIA; Nesbitt and Young, 1982; Fig. 4A) or the Chemical Index of Weathering (CIW; Harnois, 1988); Fig. 4B). Considering only small catchments ($<30 \times 10^3$ km^2), in order to exclude any effect related to alluvial storage (which could further modify $^{234}U/^{238}U$ activity ratios), no relationship was observed between ($^{234}U/^{238}U$) and the CIA in river sediments (Appendix 1), while a weak correlation was identified with CIW in silt size fractions ($R^2 = 0.56$; Appendix 2), with lower ($^{234}U/^{238}U$) values being associated with higher CIW. Overall, our data indicate that chemical weathering intensity does not exert a first-order control on ($^{234}U/^{238}U$) in river sediments, although enhanced weathering most likely results in preferential leaching of ^{234}U, in agreement with the findings of Li et al. (2016).

In addition to weathering, climate could also directly influence the observed degree of ^{234}U-^{238}U fractionation in river sediments. For instance, one would expect to observe an higher degree of ^{234}U-^{238}U fractionation in dry regions due to slow sediment transport (Kronfeld et al., 2004).

Additionally, in cold arid regions, freeze-thaw physical weathering of rocks, which typically results in the formation of very fine sediments (Anderson, 2005), could be accompanied by enhanced recoil effect and, in turn, low ($^{234}U/^{238}U$) values in the residual fine-grained sediment (DePaolo et al., 2006). While glacial weathering in subarctic environments also drives intense rock physical weathering through glacial abrasion (Pedersen and Egholm, 2013), resulting possibly in enhanced recoil effect and low ($^{234}U/^{238}U$) ratios in corresponding sediments, Vigier et al. (2001) reported ($^{234}U/^{238}U$) close to secular equilibrium in the suspended particulate loads of the Mackenzie River. In tropical regions with high rainfall, the degree of preferential ^{234}U loss from detrital grains will be mostly dependent on the time elapsed in soils and associated recoil effect, which should lead to sediment ($^{234}U/^{238}U$) close to secular equilibrium (Kronfeld et al., 2004; Robinson et al., 2004), as the sediment should be exported faster from regions experiencing heavy rainfall than from dryer catchments. However, high levels of precipitation could also drive high rates of physical erosion, which would increase mineral breakdown processes and hence result in lower ($^{234}U/^{238}U$) (Dosseto et al., 2008b).

In this study, we investigated the potential relationship between climate and ($^{234}U/^{238}U$) using the classification used by Bayon et al. (2016), which categorized studied river basins into distinct climate zones (see section 2.1; Fig. 5A). A t-test comparison for each climatic zone indicates that there is statistically no significant ($^{234}U/^{238}U$) variations (with all p-values >0.05) between each climatic zones for both silt and clay fractions (Fig. 5A), hence suggesting that climate does not play a major role in controlling the distribution of ($^{234}U/^{238}U$) of river sediments. In order to separate any possible effect related to the size and lithology of studied river basins, we also investigated the influence of climate on ($^{234}U/^{238}U$) in small catchments (Appendix 3) and igneous/metamorphic or sedimentary basins (Fig. 5), without identifying any clear relationships

too. Additionally, no particular correlations were observed between ($^{234}U/^{238}U$) in both silt and clay size fractions and MAT and MAP, regardless of the size and/or lithology of studied river basins (Fig. 5D; Appendix 4), hence supporting the view that ($^{234}U/^{238}U$) in river sediments is not controlled by climate.

Physical erosion could also potentially play a role in controlling the U isotopic composition of detrital sediments. High mountain regions are typically subject to substantial denudation rates and rapid sediment export, while erosion is much reduced in floodplains and coastal areas (von Blanckenburg, 2006; Burbank et al., 1996; Larsen and Montgomery, 2012; Summerfield and Hulton, 1994). High-elevation watersheds are generally characterized by "weathering-limited" regimes, where the rate of supply of detrital sediments is more rapid than the rate of silicate mineral dissolution. Sediment ($^{234}U/^{238}U$) in such environments could remain close to 1, since there is limited time for substantial ^{234}U loss from detrital grains. However, enhanced physical weathering in high mountain environments can also produce abundant fine-grained material with a high surface/volume ratio, thus being more prone to ^{234}U loss due to recoil effect. Li et al. (2016) observed that ($^{234}U/^{238}U$) ratios in Changjiang (Yangtze) and Zhuoshui rivers sediments increase with elevation in weathering-limited highlands, illustrating the relationship between U isotopes and erosion since higher elevations are also characterized by higher erosion rates. Under reduced physical erosion rates, such as in floodplains and lowlands, the residence time of sediments is relatively long, and this typically leads to more intense mineral dissolution. In such "transport-limited" weathering systems (Carson and Kirkby, 1972; Stallard and Edmond, 1983), the supply of fresh minerals is generally limited by the degree of soil removal in upper soil sequences (Riebe et al., 2004; West et al., 2005). As a result, the long duration of weathering is expected to result in low ($^{234}U/^{238}U$) in fine-grained material. Li et al. (2017b) identified a direct relationship between

erosion rates and ($^{234}U/^{238}U$) in sediments, showing that sedimentary ($^{234}U/^{238}U$) ratio are generally lower in river basins characterized by low denudation rates, reflecting a long residence time of the sediment in soils.

Here, there is no clear relationship between ($^{234}U/^{238}U$) in silt and clay size fractions and the maximum elevation of the river basins (Fig. 6A). Focusing on small catchments (Appendix 5) or grouping catchments according to their lithology yield similar observations for silt size fraction (Fig. 6B). In crystalline catchments (Fig. 6C), the ($^{234}U/^{238}U$) of clay size fractions slightly decrease with increasing maximum elevation ($R^2 = 0.46$). This suggests that the effect of physical weathering on producing fine grained sediments and thus promoting ^{234}U loss, dominates over the role of physical erosion on reducing sediment residence time (and thus limiting ^{234}U loss).

4.3 The role of lithology

The degree of rock weathering and erosion on continents is strongly dependent on lithology (Bluth and Kump, 1994; Meybeck, 1987). For instance, the evidence that sedimentary rocks weather much faster than crystalline igneous or metamorphic rocks could possibly impact the distribution of ($^{234}U/^{238}U$) in sediments.

In basins underlain by carbonaceous sedimentary rocks, the combination of both high weathering and high physical denudation rates (Bluth and Kump, 1994; Suchet et al., 2003) (Palumbo et al., 2010) could result in short sediment residence time and as a consequence to reduced degrees of ^{234}U-^{238}U fractionation in corresponding sediments. In such sedimentary basins, ($^{234}U/^{238}U$) in detrital grains could be expected to be close to secular equilibrium, as long as the source sedimentary rocks are initially in secular equilibrium (e.g. Sarin et al., 1990). In contrast, weathering is slower in river basins dominated by igneous or metamorphic rocks, and

this could result in lower sediment ($^{234}U/^{238}U$) ratios. With regard to U isotopes, multi-lithological catchments could behave similarly to sedimentary basins, as marls and other carbonaceous sedimentary rocks typically weather more rapidly than igneous and metamorphic rocks, hence possibly resulting in ($^{234}U/^{238}U$) near secular equilibrium in erosional products.

To determine the role of lithology on ($^{234}U/^{238}U$) ratios in river sediments, we compared ($^{234}U/^{238}U$) of both silt and clay fractions with the percentage of sedimentary, igneous, and volcanic rocks for corresponding river catchments (Bayon et al., 2020; Table 3). Interestingly, while there is no correlation with the percentage of igneous or volcanic rocks, we observe a broad relationship between ($^{234}U/^{238}U$) and the percentage of carbonate rocks (silt: $R^2 = 0.42$; clay: $R^2 = 0.50$; Figure 7B). Additionally, the possible effect of the lithology can be also assessed by comparing measured ($^{234}U/^{238}U$) ratios to corresponding neodymium (Nd) isotopic compositions for the same sediment fractions (Bayon et al., 2015, 2020) (expressed here as ε_{Nd} values; Fig. 7A). In contrast with U isotopes, sedimentary neodymium isotopic signatures from the initial rocks are preserved during all weathering, transport and depositional processes (e.g. Goldstein et al., 1984), and hence can be used as sediment provenance tracers (e.g. Goldstein and Hemming, 2003). To a large extent, the distribution of ε_{Nd} values in river-borne sediments reflects the mean age of average source rocks in corresponding drainage basins (Goldstein and Jacobsen, 1987), and hence can be used, to a first approximation, as a lithological tracer for discriminating between sediments derived from 'old' igneous/metamorphic provinces (characterized by low ε_{Nd} values typically below -14; Bayon et al., 2015), and 'young' volcanic basins (with ε_{Nd} values typically > -5). Large river systems draining a large diversity of continental rocks generally display intermediate ε_{Nd} values (Bayon et al., 2015; Goldstein et al., 1984), which make Nd isotopes less suitable as lithological tracers. In this study, there is no direct relationship between ε_{Nd} and ($^{234}U/^{238}U$) in both size fractions (Fig.

7A). A t-test indicates a statistically significant difference between sediments originating from crystalline basins ($-15 < \varepsilon_{Nd}$ or $\varepsilon_{Nd} > -5$), which have ($^{234}U/^{238}U$) mostly >1 and sediments that are from sedimentary basins ($-15 > \varepsilon_{Nd} < -5$), which generally have ($^{234}U/^{238}U$) below 1, with a p-value between these two groups of 0.0015 (<0.05).

The role of lithology on ($^{234}U/^{238}U$) can be further explored considering the case study of rivers from Northern Ireland. As discussed in Bayon et al. (2018), rivers from Northern Ireland drain watersheds characterized by distinct geological formations, including Cenozoic basaltic rocks (River Bush, River Maine, Six-Mile Water), old Proterozoic metamorphic terranes (River Foyle, River Swilly, Moyola), and Paleozoic/Mesozoic sedimentary rocks (Blackwater, Lough Erne, Lower River Bann). Northern Ireland was completely covered by the British-Irish ice sheet during the last glacial period (Clark et al., 2012). As a consequence, the fine-grained particulate loads transported by Northern Irish rivers are most likely derived from the erosion of late glacial deposits and/or post-glacial soil sequences that developed in the region after the retreat of the ice-sheet (i.e. paraglacial processes; Church and Ryder (1972)), hence from soils having similar ages of formation (Dempster et al., 2013). In addition, all studied river systems are characterised by very similar climatic conditions. Therefore, any observed ($^{234}U/^{238}U$) variation amongst studied sediment samples is likely to reflect lithological effects, rather than differences in sediment transfer, soil age or climate. Rivers draining the British Tertiary volcanic province carry clay- and silt-sized detrital fractions both characterized by ($^{234}U/^{238}U$) > 1 (Fig. 7B). The three rivers draining Proterozoic metamorphic rocks display clays with ($^{234}U/^{238}U$) > 1 and coarser silt fractions with activity ratios close to 1 (Fig. 7C). The sediments from the Paleozoic/Mesozoic sedimentary rocks have ($^{234}U/^{238}U$) < 1 in both silt and clay size fraction. Thus, in the case of Northern Ireland, river sediments derived from sedimentary basins also appear to exhibit ($^{234}U/^{238}U$) ratios lower than

those from crystalline basins. For such sedimentary basins, the presence of relatively low ($^{234}U/^{238}U$) signatures in river sediments could reflect a residual ($^{234}U/^{238}U$) composition <1 in corresponding source rocks, although it remains unclear how any significant ^{234}U-^{238}U disequilibrium could possibly occur in rocks older than 1 Ma . A more plausible explanation is that the lower ($^{234}U/^{238}U$) values determined in sediments from sedimentary basins reflect that the fact that sedimentary rock weathering most likely results in finer particle sizes compared to the alteration and erosion of crystalline silicate rocks. However, the t-test comparison between grain sizes from each lithological group indicates that there is no statistically significant variation of ($^{234}U/^{238}U$) with p-values >0.05 (0.94) for sediments from sedimentary and mixed lithologies, and close to 0.05 (0.06) for sediments from igneous & metamorphic rocks. The observed ($^{234}U/^{238}U$) > 1 in sediments derived from crystalline basements could reflect the fact that neo-formed clays produced from crystalline silicate rocks are more prone to subsequent re-adsorption of ^{234}U on secondary clays, although further investigation will be required to test this hypothesis. To summarize, while the lack of any strong relationship between U and Nd isotopes suggests that the lithology does not represent a major control on the distribution of ($^{234}U/^{238}U$) ratios of river sediments, the apparent correlation observed between ($^{234}U/^{238}U$) and the percentage of carbonate rocks in river catchments implicitly suggests that the degree of U isotope fractionation in fine-grained sediments is partially dependent on the lithology of source rocks, probably reflecting the fact that they are being eroded more rapidly (and presumably into finer particle sizes) than crystalline silicate rocks.

4.4 The role of catchment size and sediment residence time on sedimentary ($^{234}U/^{238}U$) ratios

As mentioned in the Introduction, the sediment transfer time within any given watershed is expected to exert a major control on sedimentary U isotopic ratios. The residence time of sediments in river basins is mainly influenced by the possibility of storage along its pathway from the hillslopes to the alluvial plain or the nearby ocean margin. Sediment transport along river systems is generally complex and mostly related to geomorphic parameters (Harvey, 2002). Several studies highlighted the relationships between sedimentary fluxes and both external (climate, sea-level) and internal factors (drainage area, relief, lithology) (Milliman and Syvitski, 1992; Syvitski et al., 2003). In particular, the drainage area and relief are the main parameters that govern the quantity of sediments delivered downstream of alluvial plains (Hovius, 1998; Milliman and Syvitski, 1992; Syvitski et al., 2003; Walling and Webb, 1996).

In general, the capacity of sediment storage in river basins is controlled by their geomorphological characteristics. On hillslopes, storage is dependent on soil thickness and the mean slope of the watershed, with thick soil sequences being typically associated with reduced erosion and vice versa. In this study, no global correlations were identified between ($^{234}U/^{238}U$) and mean soil thickness (Fig. 8A), even when considering small catchments only (Appendix 6). However, when investigating separately the relationship between ($^{234}U/^{238}U$) and soil thickness in igneous/metamorphic and sedimentary basins (Fig 8B & C), we can notice a slight decrease of the ($^{234}U/^{238}U$) ratios as the average depth to bedrock decreases in sedimentary watersheds. In contrast, clays separated from crystalline basins display a trend exhibiting decreasing ($^{234}U/^{238}U$) ratios when the depth to bedrock increases (Fig. 8C), in agreement with the fact that sediments will spend more time in thicker soil sequences (thus $^{234}U/^{238}U$ activity ratios will decrease accordingly). Ideally, sediment samples collected at the bottom of hillslopes would be best suited to investigate the role of hillslope storage on the ($^{234}U/^{238}U$) of sediments, rather than at the river mouth, where

the effect of alluvial storage adds complexity to the interpretation. This may explain the apparent lack of clear control of hillslope storage on the ($^{234}U/^{238}U$) of sediments.

In river basins where alluvial storage is significant, the sediment residence time could be a function of the size of the alluvial plain and its storage efficiency. Previous studies have shown that specific sedimentary fluxes decrease with the size of the drainage area (Milliman and Syvitski, 1992; Syvitski and Milliman, 2007), whenever storage efficiency is increasing. No strong relationship is apparent when looking at the variation of clay- and silt-size ($^{234}U/^{238}U$) depending on the size of the catchment (Fig. 8D, Table 4), except when considering the rivers draining igneous & metamorphic rocks, in which activity ratios decrease as the size of the catchment increases (Fig. 8E & 8F). These observations clearly suggest that the size of the catchment and inferred sediment residence time exerts a major role in controlling the degree of ^{234}U-^{238}U fractionation in river sediments. The observed slight increase of ($^{234}U/^{238}U$) in river sediments from the largest catchments could indicate that incorporation of ^{234}U into neo-formed clays occurs during the storage of the sediments along the alluvial plain. It is interesting to note that no trend is visible for sediments derived from basins draining sedimentary rocks and mixed lithologies. One hypothesis to explain this absence of relationship could be that the weathering in sedimentary rocks create grains with inherited shapes, by breaking the cement linking them, whereas in crystalline basins silicate weathering generates new grains with fresh mineral surfaces prone to dissolution. The fractionation of ^{234}U over ^{238}U being influenced by the shape of the grain, this could explain the observed difference ($^{234}U/^{238}U$) and the drainage area in sedimentary and crystalline basins.

Coarse-grained sediments are generally stored more efficiently in watersheds, hence being associated with greater transfer time, while in contrast, fine-grained particles such as clays are more rapidly exported from river systems (Russell, 1955). This time-integrated differential

behaviour between coarse and fine particles possibly accounts for the observed relationship in Fig. 7G, showing that in small sedimentary basins (area <30 x10^3 km^2), the ($^{234}U/^{238}U$) activity ratio is lower in silt-size fractions compared to clays (Fig. 8H). As silt-size particles reside for a presumably longer time than clays in river systems, they are likely to be comparatively depleted in ^{234}U, leading to lower ($^{234}U/^{238}U$) signatures than in corresponding clays (Fig. 9). Conversely, in larger sedimentary systems, the difference in ($^{234}U/^{238}U$) between clay and silt fractions is null or slightly negative (Fig. 8H), which could suggest that extensive storage in the alluvial system can buffer differences in transport rate between the two size fractions (Fig. 9).

4.5 Complex interactions of environmental controls on sediment residence time

In a sedimentary system, many external and internal parameters interact together to result in a variety of different landscape morphologies. Thus, after having shown in the above discussion that the distribution of ($^{234}U/^{238}U$) in river sediments is not controlled by any single parameter , it is interesting to investigate the combined influences of several parameters on measured ($^{234}U/^{238}U$) ratios. For this purpose, we used a multiple regression analysis between ($^{234}U/^{238}U$) in both silt and clay fractions and all the environmental parameters that were discussed above: MAP and MAT for the climatic parameters; CIA for the degree of chemical weathering; the percentage of sedimentary rocks outcropping in river catchments and ε_{Nd} for the lithology; the depth to bedrock, the catchment size and maximum elevation for the physical basin characteristics. The obtained predicted ($^{234}U/^{238}U$) agree well with measured ($^{234}U/^{238}U$) values, with $R^2 = 0.68$ (n = 30 ; Figure 10) for silt-size fractions. As a consequence, this implies that about 70% of the variability of U activity ratios in river sediments can be explained by the sum of these different environmental parameters. Note however that the obtained p-value for each individual variable is only significant for the

percentage of sedimentary rocks within the basin, suggesting that all the resulting relationship need to be considered with caution. For the clay-size fractions, the same parameters can predict 80% of ($^{234}U/^{238}U$) (n=28; $R^2 = 0.80$), with p-values being only statistically significant for MAP (p-value = 0.01 < 0.05), the maximum elevation of the basin (p-value = 0.02 < 0.05) and the depth to bedrock (p-value = 0.01 < 0.05). Despite inherent uncertainties, this multiple regression analysis indicates that many environmental factors govern the distribution of ($^{234}U/^{238}U$) and the sediment residence time in river catchments.

5. CONCLUSIONS

Our large-scale investigation of river sediments worldwide demonstrates a strong grain size decoupling of U isotopes. A significant proportion of fine-grained (clay-size) sediments in river basins are characterized by ($^{234}U/^{238}U$) values above secular equilibrium (>1), especially in crystalline basins regions dominated by volcanic and igneous/metamorphic rocks, possibly reflecting the preferential incorporation of dissolved ^{234}U into secondary clays upon weathering. Neither weathering intensity, climate, erosion nor lithology appear to directly govern the distribution of ($^{234}U/^{238}U$) values in river sediments, although the erosion of sedimentary rocks most likely results in a higher degree of ^{234}U-^{238}U fractionation compared to crystalline silicate rocks, possibly reflecting the fact that it can generate finer sediments that are more prone to ^{234}U loss through recoil effect. Overall, the size of the catchments is identified as being one of the main parameters explaining the observed distribution of ($^{234}U/^{238}U$) in river sediments, via its impact on the sediment residence time. In catchments draining crystalline basement rocks, the difference between ($^{234}U/^{238}U$) in silt- and clay-size fractions is best explained by the fact that clays are exported more rapidly than silts, resulting in different size-dependent ($^{234}U/^{238}U$) signatures. In

large river basins, sediment storage in alluvial plains also impact the distribution of ($^{234}U/^{238}U$) in river sediments, causing significant ^{234}U loss regardless of grain size. Taken together, our findings confirm that U isotopes are sensitive tracers of the sediment residence time in river basins, providing further evidence for their utility in the sedimentary record to reconstruct past variations in sediment residence time and their links to landscape evolution.

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FIG. CAPTIONS

Fig. 1: World map with the location of studied sediment samples, the sediments derived from igneous & metamorphic rocks are in blue whereas those from sedimentary and mixed lithologies are in orange. The sediments from large catchments (>30x10^{3}km^2) are symbolised by a triangle and those from small basins (<30x10^{3}km^2) by a circle. The digital elevation model is derived from the ETOPO1 Global relief Model (https://www.ngdc.noaa.gov/mgg/global/global.html).

Fig. 2: Depletion of (^{234}U/^{238}U) following the successive leaching steps for Clarence (dark grey) and Loire (light grey) sediments for silt (triangles) and clay (diamonds) size fractions. 2σ errors are smaller than the symbol size. The dashed line represents the secular equilibrium (^{234}U/^{238}U) = 1.

Fig. 3: Histogram of (A) uranium concentrations and (B) (^{234}U/^{238}U) activity ratios in silt (n=65, light grey) and clay (n=65, dark grey) size fractions; the dashed lines represent the mean values for silt (light grey) and clay (dark grey), and the blue rectangle represents the values of the upper continental crust (UCC - Condie, 1993; McLennan, 2001).

Fig. 4: (^{234}U/^{238}U) activity ratio as function of (A) the Chemical Index of Alteration (CIA) in silt (n=40, light grey) and clay (n=34, dark grey) size fraction fractions; (B) the Chemical Index of Weathering (CIW) in silt (n=40, light grey) and clay (n=34, dark grey) size fraction fractions. 2σ errors are smaller than the symbol size. No clear relation emerged between (^{234}U/^{238}U) and weathering indices.

Fig. 5: ($^{234}U/^{238}U$) activity ratios as a function of climatic zones as defined in Bayon et al. (2018): catchments are classified into five different climatic zones, defined according to the following arbitrary criteria: (1) 'SAr': cold and dry regions, with MAT < 8 °C and MAP < 800 mm; (2) 'Dry': Temperate and warm dry environments, with MAT > 10 °C and MAP < 800 mm; (3) 'Te.H': Temperate and humid regions, with 8 °C < MAT < 16 °C and MAP > 1000 mm; (4) 'Wa.H': Tropical regions with humid conditions, with MAT > 20 °C and MAP < 1500 mm; and (5) 'Tr.W': Tropical wet regions, with MAT > 20 °C and MAP > 1500 mm, depending on (A) grain size fractions (silt :light grey and clay: dark grey) or lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in silt (B) and in clay (C) size fractions ; ($^{234}U/^{238}U$) activity ratios as a function of Mean Annual Temperature (MAT) of the catchment depending on (D) grain size fractions (silt :light grey and clay: dark grey) or lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in silt (E) and in clay (F) size fraction. There is no apparent climate control on the ($^{234}U/^{238}U$) activity ratios.

Fig. 6: ($^{234}U/^{238}U$) activity ratios as function of maximum elevation of the catchment depending on (A) grain size fractions (silt :light grey and clay: dark grey) ; lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in (B) silt and in (C) clay size fraction. There is no clear relationships between maximum elevation and ($^{234}U/^{238}U$), except in crystalline catchments where ($^{234}U/^{238}U$) of clay size fractions slightly decrease with increasing maximum elevation.

Fig. 7: ($^{234}U/^{238}U$) activity ratios as a function of (A) ε_{Nd} values (from Bayon et al., 2015 ; 2020) in sediments (diamonds: clay, triangles: silt (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) for all studied rivers (silt, n=51; clay, n=40), (B) the percentage of carbonate rock inside the basin (silt :light grey and clay: dark grey) (C) ε_{Nd} values for Northern Ireland rivers (silt, n=9; clay, n=8). There is no direct control of the global lithology on the variation of ($^{234}U/^{238}U$), however the percentage of sedimentary rocks inside the basin influence ($^{234}U/^{238}U$).

Fig. 8: ($^{234}U/^{238}U$) activity ratios as a function of Depth To Bedrock (DTB; in cm) (A) depending on grain size fractions (silt: light grey and clay: dark grey) (B) lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in silt and (C) in clay size fraction ; ($^{234}U/^{238}U$) activity ratios as a function of drainage area (in km^2) (D) depending on grain size fractions (silt :light grey and clay: dark grey) (E) lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in silt and (F) in clay size fraction ; (G) difference of ($^{234}U/^{238}U$) between clay and silt size fractions, as a function of the basin drainage area, (H) grouped in two lithological categories (blue for igneous & metamorphic rocks and beige

for sedimentary and mixed lithologies). The ($^{234}U/^{238}U$) activity ratio is mainly dependent of the size of the drainage area.

Fig. 9: Schematic evolution of ($^{234}U/^{238}U$) in silt and clay fractions as a function of the catchment size In small catchments, the clay fraction may be exported too rapidly to develop significant ^{234}U depletion, with ($^{234}U/^{238}U$) remaining close to the secular equilibrium, while a slower transport for silts results in more pronounced ^{234}U depletion and ($^{234}U/^{238}U$) ratios <1. In large river systems, alluvial storage buffers the difference in the transport rates of clays and silts, implying that both fractions show significant ^{234}U depletion.

Fig. 10: ($^{234}U/^{238}U$) activity ratio observed in river sediments compared with ($^{234}U/^{238}U$) activity ratio predicted with a multi-regression analysis while considering: MAP, MAT, CIA, the percentage of carbonate rocks, ε_{Nd}, the depth to bedrock, the Area and the maximum of elevation of the catchment, (silt :light grey and clay: dark grey). 2σ errors are smaller than the symbol size. The fractionation of uranium isotopes is dependant of multiple factors that control sedimentary processes, which reflect the complexity of sedimentary system.

APPENDIX

Appendix 1: Relationships between ($^{234}U/^{238}U$) and the chemical index of alteration (CIA)(A) as categories or (B) as continuous values (C) and in small area (<30 x10^3 km^2) in silt (n=40, light grey) and clay (n=34, dark grey) size fraction fractions ; depending on two lithology groups (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in respectively silt and clay size fractions (D)(G) as categories or (E)(H) as continuous values and (F)(I) in small area (<30 x10^3 km^2).

Appendix 2: Relationships between ($^{234}U/^{238}U$) and the chemical index of weathering (CIW)(A) as categories or (B) as continuous values (C) and in small area (<30 x10^3 km^2) in silt (n=40, light grey) and clay (n=34, dark grey) size fraction fractions ; depending on two lithology groups (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in respectively silt and clay size fractions (D)(G) as categories or (E)(H) as continuous values and (F)(I) in small area (<30 x10^3 km^2).

Appendix 3: ($^{234}U/^{238}U$) activity ratios as a function of Mean Annual Temperature (MAT) of the catchment depending on (A) grain size fractions (silt :light grey and clay: dark grey) ; lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in (B) silt and (C) in clay size fraction for all the studied river and in the same way for sediment from small catchments (D),(E) and (F) respectively ; ($^{234}U/^{238}U$) activity ratios as function of (A) climatic zones as explained in Fig. 4 for sediments from small catchment groups depending on (G)

grains size fraction (silt - light grey and clay - dark grey) and lithologies (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in (H) silt and (I) clay.

Appendix 4: ($^{234}U/^{238}U$) activity ratios grouped based on grain size (silt :light grey and clay: dark grey) as a function of Mean Annual Precipitation (MAP) (A) as categories, (B) continuous values) for all the studied sediments, (C) for sediments from small catchments (Area <30,000 km^2); grouped based on two lithological categories (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in silt size fraction (D) as categories, (E) continuous values) for all the studied sediments, (F) for sediments from small catchments (Area <30,000 km^2); and in clay size fraction respectively (G), (H) and (I).

Appendix 5: ($^{234}U/^{238}U$) activity ratios as a function of maximum elevation of the basin depending on (A) grain size fractions (silt :light grey and clay: dark grey) ; lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in (B) silt and (C) in clay size fraction for all the studied river and in the same way for sediment from small catchments (D),(E) and (F) respectively

Appendix 6: ($^{234}U/^{238}U$) activity ratios as a function of Depth To Bedrock (DTB) of the basin depending on (A) grain size fractions (silt :light grey and clay: dark grey) ; lithology (blue for igneous & metamorphic rocks and beige for sedimentary and mixed lithologies) in (B) silt and (C) in clay size fraction for all the studied river and in the same way for sediment from small catchments (D),(E) and (F) respectively

Table 2
U concentration and activity ratio for silt and clay of Loire River Sediments

Sample sites	grain size fraction	U (ppm)	($^{234}U/^{238}U$)	2s.e.
Cordemais	silt	3.84	0.960	*0.001*
	clay	3.48	0.983	*0.001*
Donges	silt	3.82	0.958	*0.002*
	clay	2.83	0.938	*0.001*
Port Lavigne	silt	4.43	0.959	*0.001*
	clay	2.95	0.937	*0.002*

Table 3 . Uranium concentration and activity ratio of silt and clay size fractions in river sediments and corresponding basin parameters

	River (*n*= *64*)	Main drained lithology	Country	Latitude	Longitude	clay U (ppm)	silt U (ppm)	clay ($^{234}U/^{238}U$)	*2s.e.*	silt ($^{234}U/^{238}U$)	*2s.e.*	Clays εNd*	Silts εNd*	Clays CIA	Silts CIA	Clays CIW	Silts CIW	Climatic zone	MAT (°C)	MAP (mm)	Maximum elevation (m)	DTB (cm)	Area (10^3 km^2)
1	Adour	Mixed/sedimentary formations	France	43.49	-1.47	2.7	3.7	**0.891**	*0.002*	**0.969**	*0.001*	-11.0	-11.6	83.4	68.5	96.8	86.7	Te.H	13	1260	2800	1717	16
2	Amazon	Mixed/sedimentary formations	Brazil	3.10	-49.50	3.2	3.0	**0.917**	*0.001*	**0.956**	*0.001*	-10.5	-10.7	88.0	75.6	99.0	93.2	Tr.W	27	2030	5500	2342	6300
3	Betsiboka	Igneous/metamorphic terranes	Madagascar	-15.52	45.72	1.6	2.0	**1.049**	*0.002*	**1.122**	*0.002*							Tr.W					
4	Blackwater	Mixed/sedimentary formations	Ireland	54.51	-6.58	3.2	2.5	**0.927**	*0.002*	**0.957**	*0.002*	-11.6	-12.6	86.7	66.0	97.9	83.6	Te.H	9	1000	360	976	1.1
5	Brantas	Volcanic Rocks	Indonesia	-7.44	112.46	0.6	0.6	**0.980**	*0.003*	**0.988**	*0.003*		4.1					Tr.W	25	2982	3480	3235	11
6	Chao Phraya	Mixed/sedimentary formations	Thailand	13.57	100.58	3.2	3.2	**0.972**	*0.004*	**0.991**	*0.002*	-8.4	-9.8	86.5	77.3	97.7	93.3	Tr.W	28	1500	2500	6378	160

7	Churchill	Igneous/metamorphic terranes	Canada	58.97	-94.10	3.3	1.5	**0.866**	*0.002*	**0.933**	*0.002*		-28.7							SAr	-3		800		1138	290
8	Clarence	Mixed/sedimentary formations	Australia	-29.43	153.25	3.2	5.1	**0.961**	*0.002*	**0.946**	*0.002*									Wa.H		15.00	250			0.132
9	Danube	Mixed/sedimentary formations	Romania	45.06	29.62	2.4	2.8	**0.872**	*0.002*	**0.954**	*0.001*	-8.5	-9.11		7 0.1		8 8.0	Dry	10	7 60.0	410 0	21 06	820
10	Don	Mixed/sedimentary formations	Russia	47.29	39.10	1.8	1.8	**0.993**	*0.009*	**0.985**	*0.006*	9.3	-1 1.0	82.5	6 1.1	96.3	8 1.3	SAr	7	5 8 0.0	180		420
11	Dordogne	Mixed/sedimentary formations	France	45.03	-0.59	2.7	3.3	**0.888**	*0.002*	**0.952**	*0.001*							Dry		8 00.0	170 0		24
12	Elbe	Mixed/sedimentary formations	Germany	53.54	9.81	2.4	2.8	**0.897**	*0.003*	**0.965**	*0.003*		-10.8					Dry	7		800		148.3
13	Elorn	Igneous/metamorphic terranes	France	48.40	-4.38	3.0	3.5	**0.941**	*0.002*	**0.961**	*0.002*	-10.9	-11.2	83.1	7 3.8	96.7	9 1.4	Te.H	11	11 20.0	340	12 89	0.33
14	Fitzroy river	Igneous/metamorphic terranes	Australia	-17.73	123.64	3.0	2.9	**1.098**	*0.002*	**0.973**	*0.002*		-18.6					Dry	19	7 60.0	490	13 05	86
15	Fly	Mixed/sedimentary formations	PNG	-8.67	144.00	2.6	2.3	**0.894**	*0.001*	**0.948**	*0.001*	-3.8	-4.99	82.9	7 5.0	95.5	8 9.5	Tr.W	26	28 50.0	400 0	24 56	76
16	Fortescue river	Igneous/metamorphic terranes	Australia	-21.29	116.14	2.6	2.1	**1.072**	*0.004*	**1.029**	*0.002*		-2 2.11					Wa.H	25		115 0	72 7	50
17	Foyle	Igneous/metamorphic terranes	Ireland	54.76	-7.45	3.5	3.1	**1.208**	*0.003*	**0.989**	*0.002*	-15.2	-16.0	78.9	6 4.7	96.6	8 4.2	Te.H	9	11 10.0	640	10 63	2.9

18	Fraser	Volcanic Rocks	Canada	49.16	-123.37	1.7		**0.948**	*0.004*			-4.2	-8.5	74.2	58.5	89.4	75.4		SAr	4	760	400	21	230
19	Ganges	Mixed/sedimentary formations	Bangladesh	23.17	90.47	4.8	2.8			**1.022**	*0.002*		-15.4						Te.H	18		700	30	1080
20	Gascogne	Igneous/metamorphic terranes	Australia	-29.83	113.77	3.3	2.2	**1.197**	*0.001*	**1.082**	*0.002*								Dry	22	235	500		79
21	Glenariff	Volcanic Rocks	Ireland	55.02	-6.11	0.4	0.2	**1.119**	*0.007*	**1.070**	*0.006*	3.7	3.7	94.5	58.2	96.9	71.8		Te.H	9	1000	410	29	0.1
22	Jamata	Igneous/metamorphic terranes	Nigeria	6.13	6.76	4.1	8.1	**1.052**	*0.002*	**0.932**	*0.001*								Wa.H	29		820		
23	Kymijoki	Igneous/metamorphic terranes	Finland	60.46	26.91	3.7	3.6	**0.961**	*0.003*	**0.972**	*0.002*	-19.8	-19.2	67.1	61.6	87.8	83.2		SAr	3	600	100		37
24	Lee	Mixed/sedimentary formations	Ireland	51.88	-8.27	2.9	4.6	**0.871**	*0.002*	**0.939**	*0.002*								Te.H					1.2
25	Loire	Mixed/sedimentary formations	France	47.28	-1.90	3.0	3.4	**0.937**	*0.002*	**0.959**	*0.001*	-7.9	-8.3	84.3	74.3	97.0	91.5		Dry	11	750	190	16	120
26	Lough Erne	Mixed/sedimentary formations	Ireland	54.30	-7.64	4.0	3.8	**0.908**	*0.002*	**0.976**	*0.003*		-14.8						Te.H	9		650	94.8	4.4
27	Lower Bann	Mixed/sedimentary formations	Ireland	54.86	-6.48	1.6	1.5	**0.935**	*0.003*	**0.966**	*0.002*	-8.9	-8.9	88.2	66.9	97.9	82.7		Te.H	9	1000	640	94.3	5.8
28	Lule	Igneous/metamorphic terranes	Norway	65.68	21.82	9.9	3.1	**1.317**	*0.002*	**1.076**	*0.002*	-20.4	-18.0		74.7		76.2		SAr	-3	700	100		25

29	Mackenzie	Mixed/sedimentary formations	Canada	69.26	-137.29	3.9	3.2	**0.848**	*0.002*	**0.897**	*0.002*	-12.2	-13.0	79.2	74.9	96.5	93.0	SAr	-4	380	3600	10990	1800
30	Mae Klong	Mixed/sedimentary formations	Thailand	13.43	99.95	4.1	3.6	**1.004**	*0.001*	**1.008**	*0.001*	-13.7	-14.3	86.2	82.1	97.8	97.6	Tr.W	28	1200	2200	670	31
31	Maine	Volcanic Rocks	Ireland	54.75	-6.32	0.6	0.3	**1.298**	*0.002*	**1.138**	*0.006*	0.6	0.1	92.8	63.7	96.3	76.4	Te.H	9	1000	4600	1032	0.29
32	Mayenne	Mixed/sedimentary formations	France	47.50	-0.55	3.5	2.6	**1.003**	*0.001*	**0.945**	*0.002*	-9.5	-9.6	84.1	74.4	96.7	91.6	Dry	12	630	4200	874	4.4
33	Mekong	Mixed/sedimentary formations	Cambodia	10.96	105.06	3.9	3.3	**1.008**	*0.002*	**1.051**	*0.002*	-8.6	-10.5	86.6	75.0	97.5	90.8	Wa.H	21	1270	5100	920	800
34	Mississippi	Mixed/sedimentary formations	USA	28.93	-89.49	2.8	2.8	**0.858**	*0.001*	**0.918**	*0.001*	-10.8	-12.3	84.3	71.3	97.2	90.1	Dry	13	760	3700	6200	3300
35	Moyola	Mixed/sedimentary formations	Ireland	54.75	-6.52	3.9	2.1	**1.104**	*0.002*	**0.989**	*0.001*	-16.1	-16.2	81.8	65.8	97.1	85.4	Te.H	9	1110	5400	1087	0.3
36	Murchison	Igneous/metamorphic terranes	Australia	-27.83	114.69	5.9	2.5	**1.239**	*0.002*	**1.152**	*0.002*		-23.6					Dry	22		5200	1442	82
37	Nalon	Mixed/sedimentary formations	Spain	43.56	-6.07	3.6	4.8	**0.892**	*0.002*	**0.947**	*0.001*							Dry			1400		3.7
38	Narva	Mixed/sedimentary formations	Estonia	59.54	27.58	4.9	3.1	**0.887**	*0.001*	**0.920**	*0.001*	-16.7	-16.0	71.1	63.0	93.4	88.5	SAr	6	640	3200	1941	56

39	Nelson river	Igneous/metamorphic terranes	Canada	57.39	-91.80	2.8	1.4	**0.823**	*0.002*	**0.932**	*0.002*		-25.6							Te.H	-3			3400	28	11
40	Niger	Mixed/sedimentary formations	Nigeria	3.20	6.68	4.0	8.7	**1.048**	*0.001*	**0.976**	*0.001*	-11.9	-11.9	85.8	8.3	94.5	7.0	Wa.H	2	11 4 0 6	820	12 47 4	22 00
41	Nile	Volcanic Rocks	Egypt	32.51	30.38	2.2	3.0	**0.947**	*0.001*	**0.978**	*0.001*	7.1	-9.6	80.2	5.8	93.4	5.0	Dry	2	11 6 0	380	11 26 2	29 00
42	Northern Dvina	Mixed/sedimentary formations	Russia	65.09	39.00	2.5	2.4	**0.944**	*0.004*	**0.963**	*0.001*	17.7	-11.1	69.6	6.1	87.9	8.6	SAr	1	7 4 0	200	29 06	35 7
43	Orinoco	Mixed/sedimentary formations	Venezuela	7.65	-66.18	4.3	5.8	**1.036**	*0.016*	**0.972**	*0.001*	13.8	-13.2		7 0.4		8 8.0	Wa.H	2 4	14 0 0	600	45 87	11 00
44	Pamisos	Mixed/sedimentary formations	Greece	37.02	22.02	2.8	2.1	**0.849**	*0.001*	**0.942**	*0.004*							Dry			230 0		0.5
45	Red River	Mixed/sedimentary formations	Vietnam	20.26	106.52	4.4	3.2	**0.888**	*0.001*	**0.942**	*0.001*	-12.2	-12.8	80.7	7 5.2	96.9	9 3.5	Tr.W	2 4	17 5 0	310 0	11 26	16 0
46	Rhine	Mixed/sedimentary formations	Netherlands	51.91	4.48	3.7	3.2	**0.819**	*0.003*	**0.943**	*0.001*	-9.3	-9.1	75.5	5 5.9	93.0	7 6.1	Te.H	8	12 1 0 0	350 0	16 39	22 0
47	Rio Aro	Igneous/metamorphic terranes	Venezuela	7.39	-64.01	3.6	4.7	**1.162**	*0.005*	**1.014**	*0.001*	-25.2	-28.5		7 7.9		9 3.4	Tr.W	2 5	37 0 0	810	27 38	30
48	Rio Caroni	Igneous/metamorphic terranes	Venezuela	8.33	-62.71	6.5	7.3	**1.061**	*0.003*	**1.006**	*0.001*	-20.9	-21.1	95.7	8 6.2	99.5	9 7.6	Tr.W	2 5	28 0 0	266 0	20 82	95
49	Rio Caura	Igneous/metamorphic terranes	Venezuela	7.58	-64.94	6.4	6.3	**1.045**	*0.002*	**1.018**	*0.001*	-21.1	-22.0	95.6	8 6.3	99.5	9 7.7	Tr.W	2 5	37 0 0	235 0	20 86	48

50	Ropotamo	Mixed/sedimentary formations	Bulgaria	42.32	27.75	2.1	2.0	**1.007**	*0.004*	**0.967**	*0.006*										Dry			400			0.2
51	Sefid Rud	Mixed/sedimentary formations	Iran	37.47	49.94	2.6	2.2	**0.946**	*0.002*	**0.943**	*0.002*	-4.6	-4.5	81.2	62.3	95.1	80.0	Dry	14	520	423	100	2				
52	Sepik river	Mixed/sedimentary formations	PNG	-3.13	142.78	1.8	1.1	**0.957**	*0.002*	**0.984**	*0.003*		0.4					Tr. W	25		400	160	84				
53	Severn	Mixed/sedimentary formations	UK	51.49	-2.78	2.6	2.6	**0.849**	*0.001*	**0.947**	*0.002*							Dry			610			11.4			
54	Shannon	Mixed/sedimentary formations	Eire	52.69	-8.91	3.6	3.3	**0.873**	*0.001*	**0.935**	*0.001*	-11.2	-1.5	80.2	67.6	97.0	85.3	Te. H	9	1200	570	923	23				
55	Six Mile	Volcanic Rocks	Ireland	54.70	-6.15	1.2	0.8	**1.278**	*0.004*	**1.128**	*0.002*	-3.2	-2.8	91.1	56.4	96.1	71.0	Te. H	9	1000	420	813	0.3				
56	Sperchios	Mixed/sedimentary formations	Ireland	54.93	-7.81	1.0	1.7	**0.866**	*0.017*	**0.954**	*0.004*							Dry			230			1.8			
57	Swilly	Igneous/metamorphic terranes	Ireland	54.93	-7.81	6.5	2.7	**1.340**	*0.002*	**1.013**	*0.002*	-13.9	-1.3		54.4		72.1	Te. H	9	1110	15	905	0.1				
58	Thames	Mixed/sedimentary formations	Sweden	63.72	20.27	2.1	2.7	**0.820**	*0.002*	**0.922**	*0.002*		-1.4					Dry	10		330			12.9			
59	Umeå	Igneous/metamorphic terranes	Sweden	63.72	20.27	5.0	3.2	**1.180**	*0.002*	**1.011**	*0.002*	-18.7	-1.7		54.3		73.8	SAr	1	520	100	0	26				
60	Upper Rive	Mixed/sedimentary	Ireland	54.38	-6.33	7.0	4.1	**1.337**	*0.004*	**1.073**	*0.002*							Te. H			110			2			

	r Ban n	formations																									
61	Vanuatu	Volcanic Rocks	Vanuatu	-17.76	168.38	2.5	1.9	**1.020**	*0.00 3*	**1.007**	*0.00 2*								Tr.W			1900				
62	Vistula	Mixed/sedimentary formations	Poland	54.65	19.28	2.7	1.8	**1.300**	*0.02 1*	**1.101**	*0.00 8*	-14.5	-1 4.5	74.0	7 0.2	94.0	9 1.9	SAr	8	7 5 0	2500	22 35	20 0
63	Yangtze	Mixed/sedimentary formations	China	31.62	121.01	3.0	2.6	**0.896**	*0.00 1*	**0.941**	*0.00 1*	-10.5	-1 1.4	77.7	6 8.5	94.2	8 7.5	Te.H	1 6	12 7 0	3200	16 14	18 00
64	Yellow River	Mixed/sedimentary formations	China	37.80	118.91	2.5	2.2	**0.972**	*0.00 3*	**1.001**	*0.00 2*	-11.9	-1 0.9	68.3	5 7.6	86.6	7 6.5	Dry	1 3	7 6 0	5100	24 69	75 0
	Mean values					**3.3**	**3.0**	**1.001**	***0.00 3***	**0.987**	***0.00 2***	**-11.5**	**-1 2.7**														

**εNd are from Bayon et al. (2015).*
CIA values (refers to Chemical Index of Alteration) are from Bayon et al. (2015);
CIW calculated based on data from Bayon et al. (2015)
Climatic categories are from Bayon et al. (2018) with (1) 'SAr': cold and dry regions, with MAT < 8 °C and MAP < 800 mm; (2) 'Dry': Temperate and warm dry environments, with MAT > 10 °C and MAP < 800 mm; (3) 'Te.H': Temperate and humid regions, with 8 °C < MAT < 16 °C and MAP > 1000 mm; (4) 'Wa.H': Tropical regions with humid conditions, with MAT > 20 °C and MAP < 1500 mm; and (5) 'Tr.W': Tropical wet regions, with MAT > 20 °C and MAP > 1500 mm.
Mean annual temperature (MAT) and mean annual precipitation (MAP) data are from Bayon et al. (2016, 2018, 2019)
Maximum elevation and area of river basins was either derived from Milliman and Farnsworth (2011), or determined in the geographical information system (GIS) software ArcGis (ESRI 2001, ArcGis Desktop 10.3.1) using the hydrological data and maps based on shuttle elevation derivatives HydroBASINS (Lehner and Grill, 2013)
Depth to bedrock (mean absolute depth to bedrock) were extracted from the SoilGrids system (https://www.soilgrids.org).

River (*n*= *64*)	Main drained lithology	Country	Latitude	Longitude	clay U (ppm)	silt U (ppm)	clay **($^{234}U/^{238}U$)**	clay **2s.e.**	silt **($^{234}U/^{238}U$)**	silt **2s.e.**	bulk **($^{234}U/^{238}U$)**	**% of silt**	**% of clay**
Adour	M+S	France	43.49	-1.47	2.7	3.7	**0.891**	*0.002*	**0.969**	*0.001*	**0.956**	83	17
Amazon	M+S	Brazil	3.10	-49.50	3.2	3.0	**0.917**	*0.001*	**0.956**	*0.001*			
Betsiboka	I	Madagascar	-15.52	45.72	1.6	2.0	**1.049**	*0.002*	**1.122**	*0.002*		51	49
Blackwater	M+S	Ireland	54.51	-6.58	3.2	2.5	**0.927**	*0.002*	**0.957**	*0.002*	**0.000**		
Brantas	V	Indonesia	-7.44	112.46	0.6	0.6	**0.980**	*0.003*	**0.988**	*0.003*			
Chao Phraya	M+S	Thailand	13.57	100.58	3.2	3.2	**0.972**	*0.004*	**0.991**	*0.002*	**0.985**	67	33
Churchill	I	Canada	58.97	-94.10	3.3	1.5	**0.866**	*0.002*	**0.933**	*0.002*			
Clarence	M+S	Australia	-29.43	153.25	3.2	5.1	**0.961**	*0.002*	**0.946**	*0.002*			
Danube	M+S	Romania	45.06	29.62	2.4	2.8	**0.872**	*0.002*	**0.954**	*0.001*		84	16
Don	M+S	Russia	47.29	39.10	1.8	1.8	**0.993**	*0.009*	**0.985**	*0.006*	**0.985**	92	8
Dordogne	M+S	France	45.03	-0.59	2.7	3.3	**0.888**	*0.002*	**0.952**	*0.001*			
Elbe	M+S	Germany	53.54	9.81	2.4	2.8	**0.897**	*0.003*	**0.965**	*0.003*			
Elorn	I	France	48.40	-4.38	3.0	3.5	**0.941**	*0.002*	**0.961**	*0.002*	**0.959**	87	13
Fitzroy river	I	Australia	-17.73	123.64	3.0	2.9	**1.098**	*0.002*	**0.973**	*0.002*			
Fly	M+S	PNG	-8.67	144.00	2.6	2.3	**0.894**	*0.001*	**0.948**	*0.001*	**0.933**	71	29
Fortescue river	I	Australia	-21.29	116.14	2.6	2.1	**1.072**	*0.004*	**1.029**	*0.002*			
Foyle	I	Ireland	54.76	-7.45	3.5	2.1	**1.208**	*0.003*	**0.989**	*0.002*	**0.000**		
Fraser	V	Canada	49.16	-123.37	1.7		**0.948**	*0.004*				86	14
Ganges	M+S	Bangladesh	23.17	90.47	4.8	2.8			**1.022**	*0.002*			
Gascogne	I	Australia	-29.83	113.77	3.3	2.2	**1.197**	*0.001*	**1.082**	*0.002*			
Glenariff	V	Ireland	55.02	-6.11	0.4	0.2	**1.119**	*0.007*	**1.070**	*0.006*	**1.071**	98	2
Jamata	I	Nigeria	6.13	6.76	4.1	8.1	**1.052**	*0.002*	**0.932**	*0.001*			
Kymijoki	I	Finland	60.46	26.91	3.7	3.6	**0.961**	*0.003*	**0.972**	*0.002*	**0.970**	76	24
Lee	M+S	Ireland	51.88	-8.27	2.9	4.6	**0.871**	*0.002*	**0.939**	*0.002*			
Loire	M+S	France	47.28	-1.90	3.0	4.4	**0.937**	*0.002*	**0.959**	*0.001*	**0.955**	80	20
Lough Erne	M+S	Ireland	54.30	-7.64	4.0	3.8	**0.908**	*0.002*	**0.976**	*0.003*			
Lower Bann	M+S	Ireland	54.86	-6.48	1.6	1.5	**0.935**	*0.003*	**0.966**	*0.002*	**0.965**	98	2
Lule	I	Norway	65.68	21.82	9.9	3.1	**1.317**	*0.002*	**1.076**	*0.002*	**1.093**	93	7
MacKenzie	M+S	Canada	69.26	-137.29	3.9	3.2	**0.848**	*0.002*	**0.897**	*0.002*	**0.885**	75	25
Mae Klong	M+S	Thailand	13.43	99.95	4.1	3.6	**1.004**	*0.001*	**1.008**	*0.001*	**1.007**	71	29
Maine	V	Ireland	54.75	-6.32	0.6	0.3	**1.298**	*0.002*	**1.138**	*0.006*	**1.150**	93	7
Mayenne	M+S	France	47.50	-0.55	3.5	2.6	**1.003**	*0.001*	**0.945**	*0.002*	**0.957**	80	20
Mekong	M+S	Cambodia	10.96	105.06	3.9	3.3	**1.008**	*0.002*	**1.051**	*0.002*	**1.034**	61	39
Mississippi	M+S	USA	28.93	-89.49	2.8	2.8	**0.858**	*0.001*	**0.918**	*0.001*	**0.898**	67	33
Moyola	M+S	Ireland	54.75	-6.52	2.3	1.9	**1.104**	*0.002*	**0.989**	*0.001*	**0.992**	97	3
Murchison	I	Australia	-27.83	114.69	5.9	2.5	**1.239**	*0.002*	**1.152**	*0.002*			
Nalon	M+S	Spain	43.56	-6.07	3.6	4.8	**0.892**	*0.002*	**0.947**	*0.001*			

Narva	M+S	Estonia	59.54	27.58	4.9	3.1	**0.887**	*0.001*	**0.920**	*0.001*	**0.917**	92	8
Nelson river	I	Canada	57.39	-91.80	2.8	1.4	**0.823**	*0.002*	**0.932**	*0.002*			
Niger	M+S	Nigeria	3.20	6.68	4.0	8.7	**1.048**	*0.001*	**0.976**	*0.001*	**0.985**	87	13
Nile	V	Egypt	32.51	30.38	2.2	3.0	**0.947**	*0.001*	**0.978**	*0.001*	**0.968**	69	31
Northern Dvina	M+S	Russia	65.09	39.00	2.5	2.4	**0.944**	*0.004*	**0.963**	*0.001*	**0.959**	77	23
Orinoco	M+S	Venezuela	7.65	-66.18	4.3	5.8	**1.036**	*0.016*	**0.972**	*0.001*	**0.974**	96	4
Pamisos	M+S	Greece	37.02	22.02	2.8	2.1	**0.849**	*0.001*	**0.942**	*0.004*			
Red River	M+S	Vietnam	20.26	106.52	4.4	3.2	**0.888**	*0.001*	**0.942**	*0.001*	**0.928**	74	26
Rhine	M+S	Netherlands	51.91	4.48	3.7	3.2	**0.819**	*0.003*	**0.943**	*0.001*	**0.926**	86	14
Rio Aro	I	Venezuela	7.39	-64.01	3.6	4.7	**1.162**	*0.005*	**1.014**	*0.001*	**1.042**	81	19
Rio Caroni	I	Venezuela	8.33	-62.71	6.5	7.3	**1.061**	*0.003*	**1.006**	*0.001*	**1.012**	88	12
Rio Caura	I	Venezuela	7.58	-64.94	6.4	6.3	**1.045**	*0.002*	**1.018**	*0.001*	**1.024**	79	21
Ropotamo	M+S	Bulgaria	42.32	27.75	2.1	2.0	**1.007**	*0.004*	**0.967**	*0.006*			
Sefid Rud	M+S	Iran	37.47	49.94	2.6	2.2	**0.946**	*0.002*	**0.943**	*0.002*	**0.943**	90	10
Sepik river	M+S	PNG	-3.13	142.78	1.8	1.1	**0.957**	*0.002*	**0.984**	*0.003*			
Severn	M+S	UK	51.49	-2.78	2.6	2.6	**0.849**	*0.001*	**0.947**	*0.002*			
Shannon	M+S	Eire	52.69	-8.91	3.6	3.3	**0.873**	*0.001*	**0.935**	*0.001*	**0.925**	83	17
Six Mile	V	Ireland	54.70	-6.15	1.2	0.8	**1.278**	*0.004*	**1.128**	*0.002*	**1.146**	88	12
Spercheios	M+S	Ireland	54.93	-7.81	1.0	1.7	**0.866**	*0.011*	**0.954**	*0.004*			
Swilly	I	Ireland	54.93	-7.81	6.5	2.7	**1.340**	*0.002*	**1.013**	*0.002*	**1.015**	99	1
Thames	M+S	Sweden	63.72	20.27	2.1	2.7	**0.820**	*0.002*	**0.922**	*0.002*			
Ume	I	Sweden	63.72	20.27	5.0	3.2	**1.180**	*0.002*	**1.011**	*0.002*	**1.021**	94	6
Upper River Bann	M+S	Ireland	54.38	-6.33	7.0	4.1	**1.337**	*0.004*	**1.073**	*0.002*			
Vanuatu	V	Vanuatu	-17.76	168.38	2.5	1.9	**1.020**	*0.003*	**1.007**	*0.002*			
Vistula	M+S	Poland	54.65	19.28	2.7	1.8	**1.300**	*0.021*	**1.101**	*0.008*	**1.135**	83	17
Yangtze	M+S	China	31.62	121.01	3.0	2.6	**0.896**	*0.001*	**0.941**	*0.001*	**0.924**	61	39
Yellow River	M+S	China	37.80	118.91	2.5	2.2	**0.972**	*0.003*	**1.001**	*0.002*	**0.999**	93	7
Mean values					**3.3**	**3.0**	**1.001**	***0.003***	**0.987**	***0.002***	**0.936**		

Table 4
Characteristics of the studied samples and their sedimentary system

	River (*n= 64*)	Type of rocks inside the basin (%) S	I	V	εNd* Clays	εNd* Silts	CIA Clays	CIA Silts	CIW Clays	CIW Silts	Climatic zone	MAT (°C)	MAP (mm)	Maximum elevation (m)	DTB (cm)	Area (10^3 km^2)
1	Adour	45.3	15.8	0.3	-11.0	-11.6	83.4	68.5	96.8	86.7	Te.H	13	1260	2800	1717	16
2	Amazon	43.8	22.9	4.2	-10.5	-10.7	88.0	75.6	99.0	93.2	Tr.W	27	2030	5500	2342	6300
3	Betsiboka										Tr.W					
4	Blackwater	88.6	0.0	3.3	-11.6	-12.6	86.7	66.0	97.9	83.6	Te.H	9	1000	360	976	1.1
5	Brantas	9.0	0.0	65.7		4.1					Tr.W	25	2982	3480	3235	11
6	Chao Phraya	38.2	18.4	5.1	-8.4	-9.8	86.5	77.3	97.7	93.3	Tr.W	28	1500	2500	6378	160
7	Churchill	10.5	57.9	1.4		-28.7					SAr	-3		800	1138	290
8	Clarence										Wa.H		1500	250		0.132
9	Danube	66.9	12.6	3.0	-8.5	-9.1		70.1		88.0	Dry	10	760	4100	2106	820
10	Don				-9.3	-11.0	82.5	61.1	96.3	81.3	SAr	7	580	180		420
11	Dordogne										Dry		800	1700		24
12	Elbe					-10.8					Dry	7		800		148.3
13	Elorn	50.3	49.7	0.0	-10.9	-11.2	83.1	73.8	96.7	91.4	Te.H	11	1120	340	1289	0.3
14	Fitzroy river					-18.6					Dry	19	760	490	1305	86
15	Fly	32.3	0.3	3.9	-3.8	-4.9	82.9	75.0	95.5	89.5	Tr.W	26	2850	4000	2456	76
16	Fortescue river	10.9	19.4	8.1		-22.1					Wa.H	25		1150	727	50
17	Foyle	2.5	52.7	6.2	-15.2	-16.0	78.9	64.7	96.6	84.2	Te.H	9	1110	640	1063	2.9
18	Fraser	33.1	26.9	39.5	-4.2	-8.5	74.2	58.5	89.4	75.4	SAr	4	760	4000	2162	230
19	Ganges	29.6	27.1	6.4		-15.4					Te.H	18		7000	3061	1080

20	Gascogne										Dry	22	235	500		79
21	Glenariff	12.4	0.0	64.4	3.7	3.7	94.5	58.2	96.9	71.8	Te.H	9	100 0	410	1129	0.1
22	Jamata										Wa.H	29		820		
23	Kymijoki				-19.8	-19.2	67.1	61.6	87.8	83.2	SAr	3	600	100		37
24	Lee										Te.H					1.2
25	Loire	56.2	28.1	4.4	-7.9	-8.3	84.3	74.3	97.0	91.5	Dry	11	750	1900	1650	120
26	Lough Erne					-14.8					Te.H	9		650	948	4.4
27	Lower Bann				-8.9	-8.9	88.2	66.9	97.9	82.7	Te.H	9	100 0	640	943	5.8
28	Lule				-20.4	-18.0		47.7		67.2	SAr	-3	700	1000		25
29	MacKenzie	77.2	19.9	1.6	-12.2	-13.0	79.2	74.9	96.5	93.0	SAr	-4	380	3600	1099	1800
30	Mae Klong	62.0	15.2	3.7	-13.7	-14.3	86.2	82.1	97.8	97.6	Tr.W	28	120 0	2200	6785	31
31	Maine	0.0	0.0	98.0	0.6	0.1	92.8	63.7	96.3	76.4	Te.H	9	100 0	460	1032	0.29
32	Mayenne				-9.5	-9.6	84.1	74.4	96.7	91.6	Dry	12	630	420	1587	4.4
33	Mekong				-8.6	-10.5	86.6	75.0	97.5	90.8	Wa.H	21	127 0	5100	5292	800
34	Mississippi	68.5	4.5	1.8	-10.8	-12.3	84.3	71.3	97.2	90.1	Dry	13	760	3700	3262	3300
35	Moyola	47.6	19.2	32.4	-16.1	-16.2	81.8	65.8	97.1	85.4	Te.H	9	111 0	540	1087	0.3
36	Murchison	2.0	18.1	1.5		-23.6					Dry	22		520	1442	82
37	Nalon										Dry			1400		3.7
38	Narva				-16.7	-16.0	71.1	63.0	93.4	88.5	SAr	6	640	320	1941	56
39	Nelson river	66.5	24.0	3.1		-25.6					Te.H	-3		3400	2829	1100
40	Niger	5.6	40.3	1.2	-11.9	-11.9	85.8	88.3	94.5	97.0	Wa.H	29	114 0	820	1247 4	2200
41	Nile	17.8	34.5	7.3	-7.1	-9.6	80.2	55.8	93.4	75.0	Dry	27	610	3800	1126 2	2900
42	Northern Dvina	56.4	0.0	0.0	-17.7	-17.1	69.6	67.1	87.9	86.0	SAr	1	740	200	2906	357
43	Orinoco	9.8	20.7	2.8	-13.8	-13.2		70.4		88.0	Wa.H	24	140 0	6000	4587	1100
44	Pamisos										Dry			2300		0.5
45	Red River	80.4	14.6	1.7	-12.2	-12.8	80.7	75.2	96.9	93.5	Tr.W	24	175 0	3100	1126	160
46	Rhine	63.0	12.1	3.2	-9.3	-9.1	75.5	55.9	93.0	76.1	Te.H	8	121 0	3500	1639	220

47	Rio Aro	0.0	54.6	25.6	-25.2	-28.5		77.9		93.4	Tr.W	25	370 0	810	2738	30
48	Rio Caroni	1.8	18.0	29.3	-20.9	-21.1	95.7	86.2	99.5	97.6	Tr.W	25	280 0	2660	2082	95
49	Rio Caura	1.8	60.6	23.9	-21.1	-21.0	95.6	86.3	99.5	97.7	Tr.W	25	370 0	2350	2086	48
50	Ropotamo										Dry			400		0.2
51	Sefid Rud	45.4	7.2	29.1	-4.6	-4.5	81.2	62.3	95.1	80.0	Dry	14	520	4230	1002	13
52	Sepik river	36.3	12.8	1.9		0.4					Tr.W	25		4000	1684	78
53	Severn										Dry			610		11.4
54	Shannon	96.0	0.0	0.7	-11.2	-11.5	80.2	67.6	97.0	85.3	Te.H	9	120 0	570	923	23
55	Six Mile	0.1	0.0	96.6	-3.2	-2.8	91.1	56.4	96.1	71.0	Te.H	9	100 0	420	813	0.3
56	Spercheios										Dry			2300		1.8
57	Swilly				-13.9	-13.3		54.4		72.1	Te.H	9	111 0	15	905	0.1
58	Thames					-12.4					Dry	10		330		12.9
59	Ume				-18.7	-17.6		54.3		73.8	SAr	1	520	1000		26
60	Upper River Bann										Te.H			110		2
61	Vanuatu										Tr.W			1900		
62	Vistula	23.5	0.1	0.0	-14.5	-14.5	74.0	70.2	94.0	91.9	SAr	8	750	2500	2235	200
63	Yangtze	69.5	10.6	3.8	-10.5	-11.4	77.7	68.5	94.2	87.5	Te.H	16	127 0	3200	1614	1800
64	Yellow River	40.3	9.2	2.0	-11.9	-10.9	68.3	57.6	86.6	76.5	Dry	13	760	5100	2469	750

Percentage of type of rocks inside the basin are from Bayon et al. (2020), S is for "sedimentary", I for "igneous", and V for "volcanic"
CIA values (refers to Chemical Index of Alteration) are from Bayon et al. (2015); CIW calculated based on data from Bayon et al. (2015)
Climatic categories are from Bayon et al. (2018) with (1) 'SAr': cold and dry regions, (2) 'Dry': Temperate and warm dry environments;
(3) 'Te.H': Temperate and humid regions; (4) 'Wa.H': Tropical regions with humid conditions; (5) 'Tr.W': Tropical wet regions.
Mean annual temperature (MAT) and mean annual precipitation (MAP) data are from Bayon et al. (2016, 2018, 2019)
Maximum elevation and area of catchments was either from Milliman and Farnsworth (2011), or determined in the geographical information system software ArcGis (ESRI 2001, ArcGis Desktop 10.3.1) using the hydrological data and maps based on shuttle elevation derivatives HydroBASINS (Lehner and Grill, 2013)
Depth to bedrock (mean absolute depth to bedrock) were extracted from the SoilGrids system (https://www.soilgrids.org).

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: