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A global survey of radiogenic strontium isotopes in river sediments

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ABSTRACT

Radiogenic strontium isotopes are routinely used in provenance studies, but their application to sediments is often complicated by various grain size and weathering effects, which can influence measured $^{87}Sr/^{86}Sr$ ratios. Here, we report Sr isotopic data for a large number of sediment samples ($n = 61$) from the world's largest rivers and other river catchments draining particular geological and climatic settings; using both clay-rich (<4 μm) and silt-size (4-63 μm) detrital fractions to re-examine the factors controlling their distribution in sediments. In agreement with previous studies, the detrital material transported by world rivers defines general inverse Nd-Sr isotope relationships, which provide further empirical evidence for the utility of radiogenic Sr isotopes in sediment provenance studies. In a novel departure, however, we show that the $^{87}Sr/^{86}Sr$ difference between paired clay- and silt-size fractions ($\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$) relates to the degree of chemical alteration of river sediments, as inferred from various relationships with weathering indices, such as the CIA, WIP and Na_2O/Al_2O_3. The weathering dependence of $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ appears to be mainly controlled by temperature. In sub-Arctic and temperate regions, river sediments systematically display positive $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ indicative of preferential alteration of biotite in soils. In contrast, in sub-tropical watersheds characterized by mean annual temperatures >20 °C, intense feldspar weathering leads to the preferential incorporation of unradiogenic Sr into secondary clay minerals; a process which results in negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ values. In addition to climate forcing, the degree of size-dependent Sr isotope decoupling is also shown to be dependent on the type of weathering regime in watersheds, being more pronounced in low-elevation environments (< 2000 m), where transport-limited conditions and the presence of thick soil sequences can be associated with intense silicate weathering, than in high mountain regions (> 4000 m) dominated by kinetically-limited weathering regimes.

While further studies will be required to test the validity of these conclusions at the local scale of weathering profiles, these findings suggest that combined Sr isotopic analyses of separate size fractions could be used as a new weathering proxy in sediment records, ideally complementing the conventional use of radiogenic Sr isotopes as provenance tracers. Finally, our results are also used to re-assess the mean Sr flux and $^{87}Sr/^{86}Sr$ composition of the suspended sediment exported to the ocean yearly, yielding a global flux-weighted average of 0.7160, identical to that proposed earlier in the seminal study of Goldstein and Jacobsen (1988).

1. Introduction

On continents, rubidium and its ^{87}Rb isotope, which slowly decays to ^{87}Sr with a half-life of almost 50 billion years (Nebel et al., 2011), is preferentially enriched in potassium-rich crustal rocks compared to mantle-derived rocks. Over geological timescales, this process has resulted in continental rocks and soils having contrasted radiogenic $^{87}Sr/^{86}Sr$ signatures (*e.g.*, Gast, 1960; Faure and Hurley, 1963; Armstrong, 1968; Dymond et al., 1974; Faure and Powell, 2012). On this principle, strontium isotope geochemistry has been used for decades in sediment provenance studies (*e.g.*, Dasch, 1969; Biscaye and Dasch, 1971; Shaffer and Faure, 1976; Asahara et al., 1999; Walter et al., 2000; Grousset and Biscaye, 2005; Hemming et al., 2007; Franzese et al., 2009). At the mineral scale, Rb and Sr are also partitioned differently

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Table 1
Sr isotopic compositions of clay- and silt-size detrital fractions in river sediments.

River		Type	Area	Lat.	Long.	MAT	Max elev.	$^{87}Sr/^{86}Sr$			Sr	Rb	$^{87}Sr/^{86}Sr$			Sr	Rb	$\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$
								Clay			Clay	Clay	Silt			Silt	Silt	
			(10^3km^2)	(°N)	(°E)	(°C)	(m)	± 2 se			(ppm)	(ppm)	± 2 se			(ppm)	(ppm)	
Rivers draining crystalline basements																		
1	Congo	Margin	3800	−5.70	11.23	23.9	1760	0.745035	±	8	42	88	0.756287	±	10	52	102	−0.011
2	Kasai (*Congo)	SPM	890	−3.18	16.20	23.5	1550	0.744120	±	11	19	16	0.753739	±	11	34	21	−0.010
3	Lualaba (*Congo)	SPM	1035	0.60	24.80	22.2	4250	0.764651	±	49	38	96	0.754644	±	10	91	142	0.010
4	Niger	Margin	2200	3.20	6.68	29.4	1340	0.722114	±	8	67	163	0.716456	±	10	260	108	0.006
5	Benue (*Niger)	River	305	7.86	6.89	26.6	1340	0.720564	±	9	93	189	0.715904	±	8	269	106	0.005
6	Orinoco[a]	River	1100	7.65	−66.18	23.9	6000	0.740648			64		0.734340	±	11	57	53	0.006
7	Rio Caroni (*Orinoco)	River	95	8.33	−62.71	25.0	2660	0.786745	±	15	53	72	0.779570	±	9	69	90	0.007
8	Rio Caura (*Orinoco)	River	48	7.58	−64.94	25.0	2350	0.789728	±	12	58	77	0.785338	±	10	68	91	0.004
9	Rio Aro (*Orinoco)	River	30	7.39	−64.01	25.0	810	0.768099	±	11	24	57	0.760058	±	7	81	69	0.008
10	Churchill	Estuary	290	58.97	−94.10	−3.0	860	0.744773	±	8	227	161	0.730543	±	8	290	87	0.014
11	Murchinson	River	82	−27.83	114.69	22.0	710	0.759074	±	9	63	124	0.777882	±	10	36	54	−0.019
12	Gascoyne	River	79	−29.83	113.77	22.0	514	0.749595	±	9	61	122	0.766852	±	9	80	94	−0.017
13	Fortescue	River	50	−21.29	116.14	25.1	1150	0.800699	±	14	31	85	0.791301	±	9	40	72	0.009
14	Kymijoki	Estuary	37	60.46	26.91	3.0	500	0.765864	±	11	111	165	0.751964	±	12	144	143	0.014
15	Ume	Estuary	26	63.72	20.27	1.2	100	0.763772	±	9	85	153	0.735794	±	11	189	100	0.028
16	Lule	River	25	65.68	21.82	−2.5	1000	0.749513	±	10	99	133	0.729469	±	8	256	92	0.020
17	East Alligator	River	14	−12.43	132.97	28	480	0.724718	±	10	66	108	0.735708	±	#	33	27	−0.011
18	Betsiboka	Margin	12	−15.52	45.72	26.3	940	0.713964	±	9	68	52	0.717193	±	12	126	70	−0.003
19	Elorn	Estuary	0.3	48.40	−4.38	11	340	0.724731	±	9	104	184	0.727313	±	10	86	104	−0.003
Rivers draining volcanic provinces																		
20	Brantas	River	11.8	−7.44	112.46	25	3480	0.705372	±	12	34	12	0.705160	±	10	35	12	0.000
21	River Maine	River	0.3	54.75	−6.32	8.7	460	0.706737	±	14	46	7	0.704979	±	9	136	8	0.002
22	Six Mile River	River	0.3	54.70	−6.15	8.7	420	0.711056	±	9	44	20	0.707455	±	13	124	17	0.004
Rivers draining mixed/sedimentary basins																		
23	Amazon	Sub delta	6300	3.10	−49.50	26.7	5500	0.726631	±	10	92	163	0.722333	±	10	135	129	0.004
24	Mississipi	Sub delta	3300	28.93	−89.49	12.8	3700	0.726629	±	10	70	154	0.722969	±	10	110	99	0.004
25	Nile	Margin	2900	32.51	30.38	26.7	3800	0.713491	±	8	115	92	0.715455	±	8	141	72	−0.002
26	Parana	River	2600	−27.47	−58.86	21.1	4800	0.738986	±	9	79	188	0.728374	±	8	74	63	0.011
27	Yangtze	Estuary	1800	31.62	121.01	15.6	3200	0.729454	±	10	72	212	0.723176	±	8	116	142	0.006
28	Mackenzie	Sub delta	1800	69.26	−137.29	−3.3	3600	0.727033	±	7	133	191	0.728817	±	9	99	110	−0.002
29	Ganges-Brahmaputra	Delta	1650	23.17	90.47	18.3	7000	0.746839	±	17	80	213	0.735326	±	10	140	110	0.012
30	Brahmaputra (*G-B)	Delta	670	24.91	89.78	15.0	5500	0.729109	±	10	103	235	0.727080	±	8	163	161	0.002
31	Volga	Estuary	1400	45.71	47.92	3.8	600						0.715912	±	10	142	63	
32	Nelson	Estuary	1100	57.39	−91.80	−3.0	3400	0.742330	±	9	208	164	0.727383	±	8	241	71	0.015
33	Murray[b]	River	1100	−35.41	139.23	18.3	2200	0.724560			84		0.721599	±	10	61	124	0.003
34	Yukon	River	850	61.93	−162.88	−5.1	6000	0.716103	±	9	150	124	0.708781	±	9	221	32	0.007
35	Danube	River	820	45.06	29.62	10.0	4100	0.723907	±	9	65	177	0.718675	±	10	102	92	0.005
36	Mekong	Delta	800	10.96	105.06	21.1	5100	0.722977	±	9	70	175	0.718744	±	8	69	61	0.004
37	Yellow River	Delta	750	37.80	118.91	12.8	3100	0.727390	±	32	73	150	0.717307	±	8	163	84	0.010
38	Amu Darya	River	535	42.22	60.12	8.8	6990	0.718455	±	9	103	147	0.712903	±	13	185	62	0.006
39	Don	River	420	47.29	39.10	6.8	180	0.718970	±	10	77	130	0.718179	±	10	104	51	0.001
40	Northern Dvina	Estuary	357	65.09	39.00	0.6	200	0.733683	±	8	125	142	0.729567	±	8	137	109	0.004
41	Fraser	Margin	230	49.16	−123.37	4.4	4000	0.710757	±	17	101	75	0.709104	±	7	252	64	0.002
42	Rhine	Estuary	220	51.91	4.48	8.1	3500	0.723074	±	9	75	199	0.718736	±	10	83	85	0.004
43	Vistula	Gulf	200	54.65	19.28	7.6	2500	0.758240	±	20	56	181	0.750031	±	10	79	134	0.008
44	Red River	Delta	160	20.26	106.52	24.0	3100	0.733489	±	7	91	223	0.737989	±	10	73	136	−0.005
45	Chao Phraya	Delta	160	13.57	100.58	28	2500	0.714750	±	9	125	175	0.719568	±	9	68	109	−0.005
46	Loire (Port-Lavigne)	Estuary	120	47.20	−1.64	10.9	1900	0.723349	±	10	61	166	0.716021	±	9	164	139	0.007
47	Loire (Donges)	Estuary	‘	47.30	−2.07	‘	‘	0.723702	±	10	81	218	0.716528	±	12	161	151	0.007
48	Loire (Cordemais)	Estuary	‘	47.28	−1.89	‘	‘	0.723558	±	11	83	217	0.715955	±	8	172	137	0.008

(*continued on next page*)

Table 1 (*continued*)

	River	Type	Area	Lat.	Long.	MAT	Max elev.	$^{87}Sr/^{86}Sr$			Sr	Rb	$^{87}Sr/^{86}Sr$			Sr	Rb	$\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$
								Clay			Clay	Clay	Silt			Silt	Silt	
			(10^{3}km^2)	(°N)	(°E)	(°C)	(m)	± 2 se			(ppm)	(ppm)	± 2 se			(ppm)	(ppm)	
49	West Fitzroy	River	86	−17.73	123.64	19.0	490	0.791683	±	10	49	156	0.786724	±	10	70	122	0.005
50	Seine	River	79	49.47	0.42	13	900	0.725631	±	9	77	183	0.723558	±	9	78	60	0.002
51	Fly River	Estuary	76	−8.67	144.00	26.2	4000	0.712928	±	9	116	126	0.711739	±	8	139	86	0.001
52	Sepik	Margin	78	−3.13	142.78	25.0	4000	0.709698	±	8	105	86	0.706465	±	8	186	53	0.003
53	Narva	Estuary	56	59.54	27.58	5.5	320	0.775341	±	10	85	180	0.761562	±	10	100	134	0.014
54	Mae Klong	River	31	13.43	99.95	28.0	2200	0.746881	±	12	34	256	0.747546	±	10	33	191	−0.001
55	Shannon	Estuary	23	52.69	−8.91	9	570	0.738141	±	21	80	207	0.725672	±	8	61	51	0.012
56	Adour	River	16	43.49	−1.47	13.0	2800	0.717362	±	9	159	209	0.716208	±	10	107	66	0.001
57	Sefid Rud	River	13	−12.43	132.97	14.0	4230	0.712471	±	10	118	121	0.708193	±	11	225	76	0.004
58	Lower River Bann	River	5.8	37.47	49.94	8.7	640	0.726947	±	9	57	87	0.716825	±	9	95	48	0.010
59	Mayenne	River	4.4	54.86	−6.48	12	420	0.735049	±	9	70	171	0.731434	±	8	71	105	0.004
60	River Blackwater	River	1.1	47.50	−0.55	8.7	360	0.733693	±	10	46	123	0.719380	±	10	107	54	0.014
61	Moyola River	River	0.3	54.51	−6.58	8.7	540	0.734450	±	12	67	132	0.729770	±	10	99	87	0.005
				54.75	−6.52													

* Tributary of a larger river system. Mean annual temperatures (MAT) are from Pinet and Souriau (1988) or from the CLIMWAT climatic database. Maximum elevation are from Milliman and Farnsworth (2011) or calculated from GIS (Bayon et al., 2020a).

[a] Clay 87Sr/86Sr and Sr concentrations are from Parra et al. (1997).

[b] Clay 87Sr/86Sr and Sr concentrations are from Gingele and De Deckker (2005).

into rock-forming minerals, leading with time and radioactive decay of ^{87}Rb to markedly different Sr isotopic compositions (see Fig. 1 for an updated compilation of Rb and Sr partition coefficient ratios in common minerals). This decoupling between Rb-Sr at the mineral scale provides a means for investigating silicate weathering processes in soils (*e.g.*, Brooks, 1968; Blum and Erel, 1995; Blum and Erel, 1997; Aubert et al., 2001; Stewart et al., 2001; White et al., 2001; Oliva et al., 2004; Négrel, 2006; Pett-Ridge et al., 2009). Silicate weathering does not affect rocks uniformly, but only particular mineral phases, hence releasing a dissolved Sr fraction with a $^{87}Sr/^{86}Sr$ signature that differs from the bulk rock composition. Over the past decades, extensive work has been done on Sr isotopes in the dissolved loads of river waters (*e.g.*, Goldstein and Jacobsen, 1987; Palmer and Edmond, 1989; Palmer and Edmond, 1992; Krishnaswami et al., 1992; Négrel et al., 1993; Gaillardet et al., 1997; Viers et al., 2000; Dessert et al., 2001; Millot et al., 2002; Singh et al., 2006; Wu et al., 2009; Pearce et al., 2015). While providing information on silicate rock weathering, dissolved riverine $^{87}Sr/^{86}Sr$ compositions are also strongly influenced by weathering of carbonates and other marine-derived sedimentary rocks, generally characterized by relatively uniform $^{87}Sr/^{86}Sr$ compositions inherited from past oceans (*e.g.*, Veizer and Compston, 1974; Veizer, 1989; McArthur et al., 2012). Most of the world's largest rivers have now been characterized for radiogenic Sr isotopes (for a recent review, see Peucker-Ehrenbrink and Fiske, 2019). Comparatively, the river suspended loads and associated sediments have received less attention with regard to Sr isotopes. Following the seminal work of Goldstein and Jacobsen (1988), which reported $^{87}Sr/^{86}Sr$ data for suspended loads from rivers across North America and a few other locations worldwide, several case studies investigated the radiogenic Sr isotope geochemistry of suspended particulates, bedloads and other river-borne sediments from large river systems (*e.g.*, Douglas et al., 1995; Allègre et al., 1996; Derry and France-Lanord, 1996; Eisenhauer et al., 1999; Galy and France-Lanord, 2001; Viers et al., 2008; Singh et al., 2008; Bouchez et al., 2011; Mao et al., 2011; Garçon et al., 2014; Rousseau et al., 2019). However, its application to river suspended loads and other sediments is often complicated by the fact that measured $^{87}Sr/^{86}Sr$ ratios can reflect various grain-size and weathering effects in addition to tracing sediment provenance (*e.g.*, Biscaye and Dasch, 1971; Eisenhauer et al., 1999; Bouchez et al., 2011; Meyer et al., 2011; Garçon et al., 2014). One major difficulty is that rubidium-rich mineral phases are commonly preferentially enriched in the finest sediment fractions, hence typically resulting in clay-size sediments being more radiogenic than coarser silty and sandy sediments (*e.g.*, Biscaye and Dasch, 1971; Eisenhauer et al., 1999; Bouchez et al., 2011). To date, the potential of radiogenic Sr isotopes in river sediments for tracing continental weathering processes is yet to be fully explored.

In this study, we investigate the radiogenic Sr isotope composition of modern river sediments collected from various geological, tectonic and climatic settings worldwide. In contrast with soil studies, which allow detailed reconstruction of weathering processes along soil depth profiles, but are also strongly affected by local effects related to differences in bedrock composition and primary mineral assemblages, the geochemical composition of fine-grained sediments deposited near the mouth of rivers provides insights into weathering processes at the catchment scale. Such a global approach is ideally suited for investigating the mechanisms controlling the distribution of geochemical tracers in sediments and for 'calibrating' weathering proxies against modern basin parameters (*e.g.*, Bayon et al., 2016; Bayon et al., 2018; Bayon et al., 2020a; Bindeman et al., 2019). Two different grain-size fractions were investigated: 1) the finest clay-rich (<4 μm) detrital material, which includes secondary clays formed in soils during silicate weathering processes; and 2) the silt-size (4-63 μm) terrigenous component, corresponding to a mixture of both primary and weathered minerals transported in the suspended particulate load (Bindeman et al., 2019). The difference between $^{87}Sr/^{86}Sr$ ratios in clay- and silt-size detrital fractions is found to display general relationships with proxies for chemical weathering intensity, but also with various basin

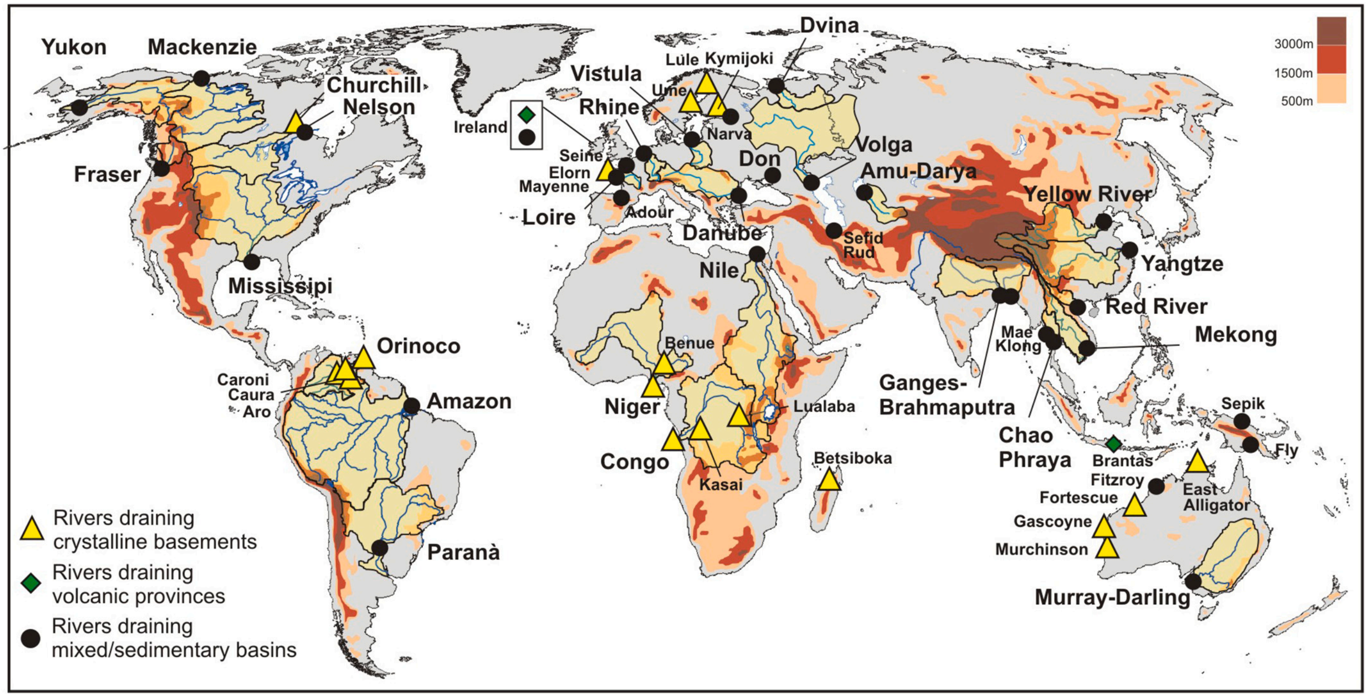

Fig. 2. The location of studied sediment samples and corresponding river basins. Yellow triangles and green diamonds correspond to rivers from ancient cratons and other areas dominated by igneous/metamorphic crystalline basement rocks, and volcanic watersheds, respectively. Black circles indicate rivers draining mixed and sedimentary rock formations.

parameters, such as temperature and relief, which altogether indicate that the size-dependent ^{87}Sr/^{86}Sr variability in river sediments is largely inherited from relatively recent weathering processes (*i.e.* at the timescale of soil formation in drainage basins).

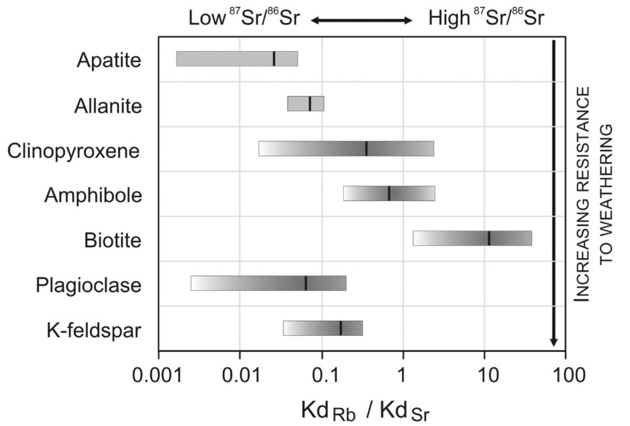

Fig. 1. The Rb-Sr mineral-liquid partition coefficient ratios (K$_D$) for most common granite-forming minerals and associated sequence of alteration. Partition coefficients were compiled from the GERM database (http://earthref.org/GERM) after removal of K$_D$ values for rare rock types. The thin black line corresponds to the mean Rb/Sr partition coefficient ratio for each mineral. Darker shaded areas indicate Minimum, maximum and mean K$_D$ values for the minerals reported in this plot are: apatite (0.002; 0.05; 0.026; n = 2); allanite (0.037; 0.106; 0.071; n = 2); clinopyroxene (0.018; 2.5; 0.35; n = 32); amphibole (0.19; 2.23; 0.66; n = 15); biotite (1.33; 38; 11.4; n = 12); plagioclase (0.002; 0.19; 0.040; n = 36); K-feldspar (0.03; 0.31; 0.17; n = 9). The alteration of accessory calcium and phosphate mineral phases (*e.g.*, apatite, sphene, allanite) typically occur at early stages of granitoid dissolution (*e.g.*, Harlavan and Erel, 2002; Erel et al., 2004; Oliva et al., 2004), followed by dissolution of ferromagnesian minerals (*e.g.*, clinopyroxene, amphibole, biotite). More resistant K-feldspars weather under intense weathering conditions, associated with the release of unradiogenic dissolved Sr fraction (with relatively low ^{87}Sr/^{86}Sr).

2. Materials and methods

2.1. World river sediments and catchment characteristics

A total of 61 modern sediment samples were analysed, corresponding to either river, estuarine, or marine fine-grained sediments deposited near the mouth of rivers. Information on sampling location and depositional settings is provided in Table 1 and Table S1, respectively. Most samples were collected either manually, from recent river bank deposits, or by gravity coring, using core-tops or near-surface sediment layers. Two additional samples correspond to river suspended particulates from the Congo Basin, collected in December 2013 at the end point of the Kasai and Lualaba (Upper Congo) rivers, with a respective suspended sediment concentration of 53 and 108 mg/l (Bayon et al., 2018). All studied samples correspond either to modern or relatively recent sediments deposited presumably during the last few centuries. Selected samples include some of the world's largest rivers (*e.g.*, Amazon, Congo, Mississippi, Nile, Yangtze, Mackenzie, Ganges-Brahmaputra), in addition to smaller river basins associated with particular geological settings,

Table 2
Sr isotopic compositions of the NIST 987 standard and rock reference materials.

Standard	Measured ^{87}Sr/^{86}Sr				Reference ^{87}Sr/^{86}Sr			
	± 2 sd			N	± 2 sd			Ref
NIST-987	0.710250	±	9	38	0.710250	±	12	Weis et al. (2005)
BHVO-2	0.703484	±	15	10	0.703487	±	19	Weis et al. (2006)
BIR-1	0.703113	±	19	8	0.703116	±	26	Jochum et al. (2016)
BCR-1	0.705033	±	15	10	0.705025	±	19	Weis et al. (2006)
BCR-2	0.705031	±	14	10	0.705019	±	16	Weis et al. (2006)

such as ancient cratons or volcanic provinces (Fig. 2). Studied samples were classified into three categories based on the lithological composition of corresponding watersheds (Table S1), following the approach described in Bayon et al. (2020a): 1) Rivers draining crystalline basements dominated by igneous, metamorphic and/or siliciclastic sedimentary rocks (n = 19). This category includes those rivers flowing through diverse Precambrian regions in Africa (Niger, Congo, and their tributaries, Betsiboka), northern South America (Orinoco, and its right-bank tributaries), Canada (Churchill), Australia (Murchinson, Gascoyne, Fortescue, East Alligator), Fennoscandia (Kymiijoki, Ume, Lule), and a small river from the Hercynian Armorican Massif in France (Elorn); 2) Rivers draining volcanic provinces (n = 3) in Northern Ireland (River Maine, Six Mile Water) and Indonesia (Brantas); 3) Rivers draining basins of mixed lithology dominated by sedimentary rocks, which include most of the major river systems investigated in this study (n = 39). Note that three sediment samples from the Loire River were analysed in this study, collected along a 50-km transect in the river estuary, in order assess the analytical uncertainty associated with sediment sampling and preparation. Information on mean annual air temperatures (MAT) in studied watersheds was derived from the literature (*e.g.*, Pinet and Souriau, 1988) or, when unavailable, from the CLIMWAT climatic database managed by the Food and Agriculture Organization of the United Nations (FAO; http://www.fao.org/land-water/databases-and-software/climwat-for-cropwat/en/). The maximum elevation in river basins was obtained from Milliman and Farnsworth (2011), or was determined using a geographical information system (see Bayon et al., 2020a for details).

2.2. *Chemical preparation*

Sieved (<63 μm) bulk sediment samples were treated using a sequential leaching procedure that successively removes carbonate minerals, reducible Fe-Mn oxide fractions, and organic matter, while ensuring no partial dissolution of the residual detrital fractions (Bayon et al., 2002). Clay-rich (<4 μm) and silt-size (4–63 μm) detrital fractions were then separated from the residual detritus by centrifugation (Bayon et al., 2015). The reproducibility of our centrifuge method and its efficiency at separating <4 μm fractions from coarser silt particles was thoroughly assessed through grain size measurements using a Malvern Mastersizer 3000 laser diffraction particle size analyser. For Sr isotopes and major/trace element analyses, about 25 mg of powder samples were digested in cleaned PTFE vials using ultrapure concentrated HF (1.5 ml) and HCl (0.5 ml) solutions, left on hotplate for 5–7 days (140 °C) after addition of a Tm spike. After evaporation, the dry residues were further digested in 6 M HCl (2 days at 140 °C), and finally taken (after drying) in sub-boiled 3 M HNO_3 (2 ml) for subsequent geochemical analyses.

2.3. *Sr isotopes*

Strontium isotopic measurements were performed at the Pôle Spectrométrie Océan (Brest) using a Thermo Scientific Neptune multi-collector ICPMS, after Sr purification onto chromatographic columns containing 1 ml Eichrom Sr spec resin (100–150 μm mesh). After sample loading (1 ml) and subsequent addition of 3 M HNO_3 (5 ml) onto the columns, the Sr fractions were collected in ultrapure (18.2 MΩ) water (4 ml). Strontium isotopic abundances were determined during one analytical session using a sample-standard bracketing technique, during which an in-house Sr standard solution (PlasmaCAL) was analysed every three samples. Potential interferences of Kr and Rb on masses ^{86}Sr and ^{87}Sr were monitored and corrected using ^{82}Kr, ^{83}Kr and ^{85}Rb, respectively. All standards and samples were prepared as 500 ppb solutions, which corresponded to a ^{88}Sr beam of ~40 V. Mass bias corrections on Sr were made with the exponential law, using $^{86}Sr/^{88}Sr = 0.1194$. Repeated analyses of a NIST-987 reference solution during the analytical session gave $^{87}Sr/^{86}Sr$ of 0.710250 ± 0.000009 (2 SD, n = 38), identical to the reference value of 0.710250 ± 0.000012 (Weis et al., 2005). The accuracy and repeatability of measured $^{87}Sr/^{86}Sr$ ratios were further assessed by analysing several replicates of a series of geological reference materials (BHVO-2; BIR-1; BCR-1; BCR-2), which all gave values and precisions in full agreement with literature data (Table 2).

2.4. *Major elements and Sr and Rb abundances*

The major element composition of most studied clay- and silt-size fractions, previously determined by wavelength dispersive X-ray fluorescence (WD-XRF), was already reported in Bayon et al. (2015). In this

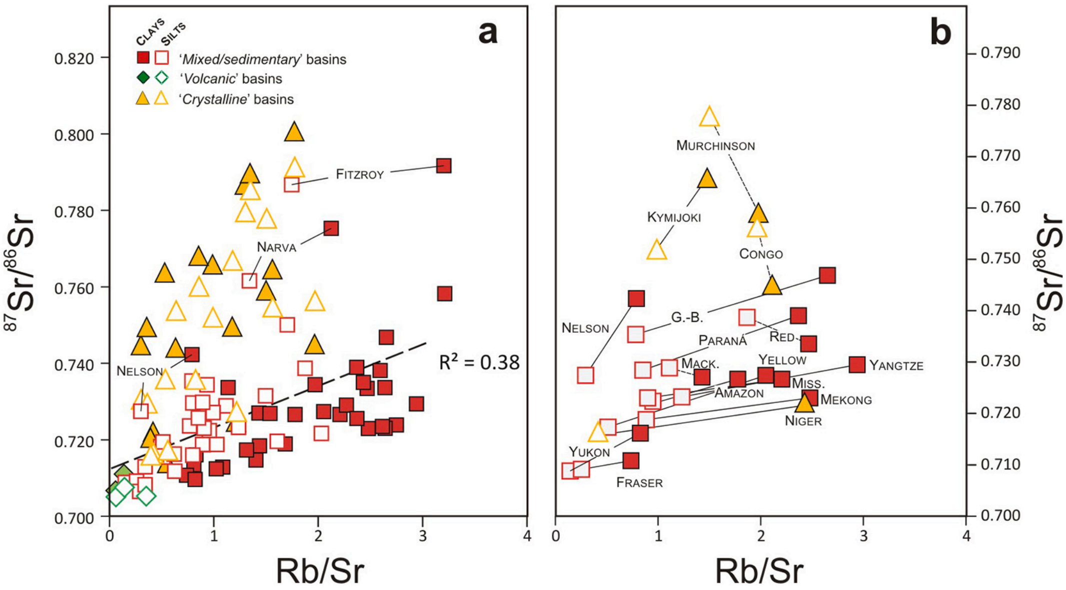

Fig. 3. Pseudo-isochron Rb-Sr diagrams in river sediments. (a) $^{87}Sr/^{86}Sr$ *versus* Rb/Sr in studied clay- and silt-size detrital fractions, indicating a positive relationship in mixed/sedimentary basins (with a best-fit regression line of $R^2 = 0.38$; excluding the samples from the Fitzroy, Narva and Nelson rivers), which reflects both the average age of the upper continental crust drained in river catchments and a grain-size effect. (b) $^{87}Sr/^{86}Sr$ *versus* Rb/Sr in selected paired clay- and silt-size fractions. While most paired size fractions display typical pseudo-isochrons, a few river sediment samples exhibit inverse grain-size relationships (dashed lines) with clay fractions being characterized by lower $^{87}Sr/^{86}Sr$ and higher Rb/Sr values than corresponding silts.

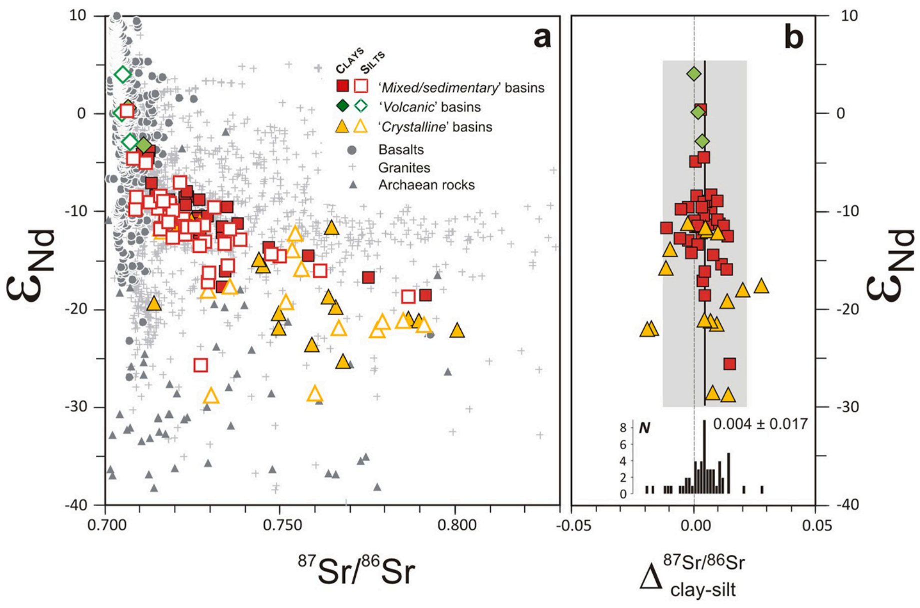

Fig. 4. Neodymium and strontium isotopes in river sediments. (a) ε_{Nd} *versus* $^{87}Sr/^{86}Sr$ in studied clay- and silt-size detrital fractions, displaying typical inverse relationships. For comparison is shown a set of Nd-Sr isotopic data for various granites, basalts and Archaean rocks complied from the GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc/). Note that Nd isotopic data are from previous studies (Bayon et al., 2015, 2016, 2019, 2020a). (b) ε_{Nd} *versus* $\Delta^{87}Sr/^{86}Sr$ $_{\text{Clay-Silt}}$, which represents the $^{87}Sr/^{86}Sr$ difference between paired clay- and silt-size fractions in river sediments. The observed range of $\Delta^{87}Sr/^{86}Sr$ $_{\text{Clay-Silt}}$ is larger in river catchments dominated by old crystalline rocks characterized by unradiogenic Nd isotopic signatures (between −0.019 and +0.028), than in rivers draining mixed/sedimentary basins (from −0.011 to +0.015) and volcanic provinces (< +0.004). This reflects the age dependency of the radiogenic Sr isotope decoupling between different grain sizes.

study, new major element data for an additional set of sediment samples ($N = 36$) were determined with a Thermo Scientific Element XR sector field ICP-MS, using the following peaks (^{23}Na, ^{24}Mg, ^{27}Al, ^{31}P, ^{39}K, ^{44}Ca, ^{49}Ti, ^{55}Mn, ^{57}Fe) acquired in medium mass resolution. The Sr and Rb concentrations were also measured for all studied samples in low resolution mode, using ^{88}Sr and ^{85}Rb, respectively. Elemental abundances were calculated using the Tm addition method, following procedures described in Barrat et al. (1996) and Barrat et al. (2012). The in-run uncertainties on all measurements were better than 2%. The precision and accuracy of our data were assessed by analysing a series of geological certified reference materials having various chemical compositions (AN-G, AGV-1, BCR-1, DNC-1, DR-N, G-2, WS-E). The results obtained for these six reference materials were in full agreement with reference values from the literature (typically <8%), with precisions generally better than 10% (RSD; N = 3).

3. Results

Major element concentrations of studied samples are listed in the supplementary Table S2. Measured Sr concentrations in studied clay- and silt-size detrital fractions range from 19 (Kasai) to 227 μg/g (Churchill) and from 33 (Mae Klong, East Alligator) to 290 μg/g (Churchill), respectively, with mean values of 83 and 122 μg/g (Table 1). As expected, river clays generally display higher Rb abundances (mean 142 μg/g) than silts (mean 88 μg/g), with concentrations ranging from 7 and 8 μg/g (River Maine) to 256 and 191 μg/g (Mae Klong), respectively. Both clayey and silty fractions encompass a large range of $^{87}Sr/^{86}Sr$ values: from ~0.705, for the volcanogenic sediments of Northern Ireland (River Maine, Six Mile Water) and Indonesia (Brantas), to ~0.780–0.800, for the sediments derived from the ancient cratons of Western Australia (Fortescue, Fitzroy) and Guiana (Rio Caroni and Rio Caura). The variation of the Sr isotopic composition in studied river-borne sediments can be illustrated using a (pseudo) isochron diagram, where $^{87}Sr/^{86}Sr$ ratios are plotted against Rb/Sr (Fig. 3). While $^{87}Sr/^{86}Sr$ *vs.* $^{87}Rb/^{86}Sr$ diagrams are routinely used in geochronological studies, typically forming linear arrays with a slope proportional to the age, Rb-Sr isochron correlations in sediments have generally no direct age significance due to the strong mobility of both Rb and Sr during weathering (*e.g.*, Douglas et al., 1995). In this study, except for a few samples derived from rivers draining Precambrian terranes (Fitzroy, Narva, Nelson; Fig. 3a), the clay-rich and silt-size detrital fractions derived from multi-lithological catchments display a weak mixing relationship ($R^2 = 0.38$) between a mantle-derived endmember, corresponding to those samples collected from volcanic provinces and characterized by low $^{87}Sr/^{86}Sr$ and Rb/Sr ratios, and crustal materials having higher Rb/Sr and more radiogenic Sr isotopic compositions. Compared to those mixed/sedimentary basins, which typically integrate the lithological and chemical diversity of the upper continental crust, the sediments derived from ancient crystalline basements generally display higher $^{87}Sr/^{86}Sr$ ratios for any given Rb/Sr, reflecting the presence of older bedrocks in corresponding watersheds (Fig. 3a). As shown previously (*e.g.*, Eisenhauer et al., 1999), the observed relationship defined by multi-lithological catchments in Fig. 3a is mostly controlled by the average age of the upper continental crust drained in river catchments, but also by a grain-size effect, by which clay-size fractions generally display higher $^{87}Sr/^{86}Sr$ and Rb/Sr ratios than corresponding silt fractions (Fig. 3b). Note however that a few samples display an opposite Rb-Sr isochron relationship (*e.g.*, Congo, Mackenzie, Murchinson, Red River), with silt-size fractions being characterized by high Rb/Sr ratios and more radiogenic Sr isotopic compositions compared to corresponding clay fractions (Fig. 3b).

When plotted against corresponding neodymium (Nd) isotopic compositions (expressed here as ε_{Nd}), a well-established tracer of provenance in sediments (*e.g.*, Grousset et al., 1988), our $^{87}Sr/^{86}Sr$ data for river sediments display typical curved (hyperbolic) relationships (Fig. 4a). To a large extent, the inverse correlation between $^{87}Sr/^{86}Sr$ and ε_{Nd} in the detrital material transported by world rivers reflects variations in the Sm/Nd and Rb/Sr ratios of source rocks having different geological ages on continents (*e.g.*, Goldstein, 1988; Goldstein and Jacobsen, 1988; Fig. 3a). In this study, this general relationship brings additional evidence for the utility of Sr isotopes as provenance tracers in sediment records. Note that it is mostly apparent for sediments from river catchments draining mixed/sedimentary rock formations, with correlation coefficients (R^2) of 0.71 and 0.64 (excluding the sample from the Nelson River) for clay-rich and silt-size fractions, respectively. Considered separately, the detrital fractions derived from ancient cratons and other crystalline catchments exhibit weaker correlations between $^{87}Sr/^{86}Sr$ and ε_{Nd} (with R^2 of 0.23 and 0.40 for clay- and silt-fractions, respectively; Fig. 4a), hence suggesting that the original Sr isotopic provenance signal in these sediments may be overprinted by weathering processes.

As commonly reported in previous studies (*e.g.*, Biscaye and Dasch, 1971; Eisenhauer et al., 1999), the clay-size fractions investigated here are commonly more radiogenic (*i.e.* having higher $^{87}Sr/^{86}Sr$ ratios) than corresponding coarser sediment fractions. Nevertheless, amongst our series of sediments ($n = 61$), a total of 11 samples, including the Congo, Kasai, Nile, Mackenzie, Red River and Chao Phraya rivers, are characterized by clays being less radiogenic than corresponding silts (Table 1; Fig. 4b). The observed $^{87}Sr/^{86}Sr$ difference between paired clay- and silt-size fractions, defined from hereafter as $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$, appears to be larger in sediments derived from ancient crystalline basements characterized by unradiogenic Nd isotopic signatures ($\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ ranging from −0.019 to +0.028; with a mean value of 0.003 ± 0.013; 1 SD; $n = 19$), compared to those from mixed/sedimentary basins (from −0.011 to +0.015; mean 0.005 ± 0.005; $n = 39$) and volcanic provinces (mean 0.002 ± 0.002; n = 3) associated with more radiogenic Nd isotopic compositions (Fig. 4b). Note that the analyses of the Loire River samples ($N = 3$; Table 1) give a mean $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ of 0.0074 ± 0.0004 (2 SD), which can be taken, to a first approximation, as an estimated uncertainty on given $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ values in this study.

4. Discussion

4.1. Significance of measured $^{87}Sr/^{86}Sr$ ratios in clay- and silt-size fractions

As will be discussed in the following sections, our main working hypothesis is that the observed $^{87}Sr/^{86}Sr$ differences between paired clay- and silt-size fractions ($\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$) mostly reflect the type and intensity of silicate weathering in river catchments. One alternative hypothesis would be that the degree of Sr isotope grain-size decoupling in river sediments is mainly controlled by the average bedrock age of corresponding catchments. As suggested previously (Eisenhauer et al., 1999), this would imply that the slope of the pseudo-isochrons defined by paired clay- and silt-size fractions in Fig. 3 is directly dependent on the mean bedrock age. This hypothesis can be directly tested using Nd isotopes, because the Nd isotopic composition of suspended particulates and river sediments strongly correlates with the average bedrock age of corresponding catchments (Goldstein and Jacobsen, 1988; Peucker-Ehrenbrink and Fiske, 2019). Here, the evidence that no particular relationship can be drawn when plotting the slope of these pseudo-isochrons with corresponding ε_{Nd} ratios for both clay- and silt-size fractions ($R^2 \sim 0.02$; graph not shown here) suggests that this hypothesis is unlikely to account for all of the observed size-dependent decoupling of Sr isotopes in studied river sediments. In addition to the mean bedrock age of river catchments, several other parameters could also partly influence the distribution of radiogenic Sr isotopes in studied size fractions. These factors are briefly reviewed below.

First, while our centrifuge method aims at separating weathered clays from primary detrital minerals in river sediments, this approach is not quantitative, resulting in partial admixtures of weathered and primary mineral phases in both clay-rich and silt-size fractions, with possible influence on measured $^{87}Sr/^{86}Sr$ ratios. As a consequence, an important pre-requisite to interpreting Sr isotopic data in separate size fractions is to estimate the contribution of weathered *versus* primary detrital signatures in each studied sample. This can be done using the total water contents previously determined by thermal conversion elemental analysis on the same suite of samples (Bindeman et al., 2019; Table S3). While the primary detrital silicate component of river sediments is mostly anhydrous (mean 1.8 wt% H_20), phyllosilicate clay minerals display water contents ranging from ~14% (kaolinite), ~11% (smectite, chlorite) to ~6% (illite) (Bindeman et al., 2019). Using clay mineral data previously determined on the same <4 μm fractions (Bayon et al., 2015), a theoretical water content can be computed for clay mineral assemblages in each sample, which, when compared to corresponding (measured) H_2O wt% values, can provide an estimate for the proportion of primary detrital minerals (see details in Table S3). These calculations indicate that primary minerals generally account for less than 20 wt% in studied <4 μm fractions (average ~ 9%), except for three samples from the Nelson (~45%), Amu Darya (~42%) and Fraser (34%) rivers (Table S3). Apart from these latter samples, this means that the Sr isotopic composition of studied clay-size fractions is largely dominated by weathered minerals, even if considering that primary minerals are preferentially enriched in Sr relative to weathered products (by a factor of ~1.6 ± 0.7, as inferred from the average Sr concentrations for studied clay-rich and silt-size fractions; Table 1). Similarly, the proportion of weathered mineral phases in river silts can be estimated using measured total water contents, indicating that weathered minerals can account for up to 30 wt% in these fractions (Table S3). Amongst studied river silts, a few samples appear to host substantially higher amounts of weathered minerals (Table S3). This is the case for the Mackenzie (34%), Amazon (35%), Caura (65%), Murray-Darling (68%), Congo (71%), and Niger (74%) rivers. In these particular samples, the presence of clays could reflect either incomplete particle size separation during our centrifuge procedure and/or, for the case of marine sediment samples (*i.e.* Amazon, Mackenzie, Congo, Niger, Murray-Darling), the presence of non-disintegrated (hence >4 μm) faecal pellets containing fine clay material. Using a simple mass balance model, these estimates can be used to compute a theoretical $^{87}Sr/^{86}Sr$ silt ratio, corrected from the presence of weathered minerals (Table S3). Except for the Murray-Darling silt fraction, for which the corrected Sr isotopic composition returns an unrealistically low value (0.6742) that departs significantly from the measured ratio (0.7216), the observed $^{87}Sr/^{86}Sr$ difference between corrected and measured values in all other samples is systematically lower than ~0.0015. Importantly, this difference is small (~ 3%) compared to the observed range of $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ in this study (~0.047), meaning that the potential presence of weathered material in studied silt fractions should not affect any of the interpretations that will

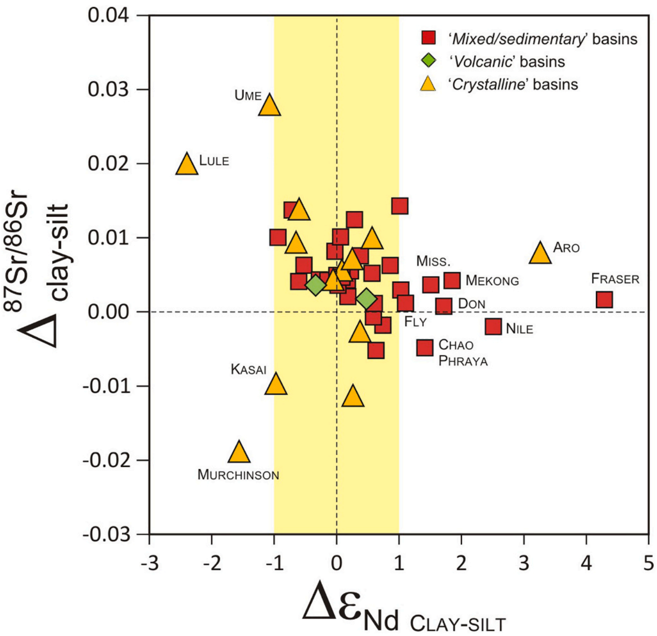

Fig. 5. The relationship between $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ and $\Delta\varepsilon_{\text{Nd Clay-Silt}}$ in river sediments. In mixed/sedimentary basins, the samples characterized by $\Delta\varepsilon_{\text{Nd Clay-Silt}} > 1$ are generally interpreted as reflecting the preferential presence of fine volcanogenic particles in clay-size fractions relative to corresponding silts (Bayon et al., 2015). In crystalline basements, the size-dependent Nd isotope decoupling in those samples with $\Delta\varepsilon_{\text{Nd Clay-Silt}} < 1$ is most likely caused by incongruent silicate weathering (Öhlander et al., 2000; Dausmann et al., 2019). The absence of any significant correlation between size-dependent Nd and Sr isotope decoupling suggests that the observed $\Delta^{87}Sr/^{86}Sr$ variability in river sediments is not controlled by a source effect. The vertical yellow band indicates the range of $\Delta\varepsilon_{\text{Nd Clay-Silt}}$ values < |1| for which both clay- and silt-size fractions are expected to derive from the same sediment sources.

be drawn in the sections below, when discussing about the factors controlling the variability of $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$. Note however that the silt fraction from the Murray-Darling sample will be discarded in the discussion below.

Furthermore, the degree of size-dependent decoupling of Sr isotopes in river sediments could also reflect a source effect, in the case where clay- and silt-size detrital particles are derived from distinct source rocks. For instance, such source effect could possibly relate to the presence of fine atmospheric particles derived from distant dust source regions (*e.g.*, Probst et al., 2000; Pett-Ridge et al., 2009). Additionally, recent studies based on Nd isotopes have documented the preferential presence of radiogenic volcanogenic particles (*i.e.* characterized by relatively high ε_{Nd} signatures) in the finest size fractions of river suspended loads (*e.g.*, Garçon and Chauvel, 2014; Bayon et al., 2020b). To investigate whether such effect could partially account for the observed size-dependent Sr isotope decoupling in the rivers draining mixed lithologies, $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ is plotted against $\Delta\varepsilon_{\text{Nd clay-silt}}$, *i.e.* the difference of Nd isotopic compositions between the same clay-rich and silt-size fractions (Bayon et al., 2015; Fig. 5). While a few samples from multi-lithological catchments indicate $\Delta\varepsilon_{\text{Nd clay-silt}}$ values >1 (*i.e.* Fraser, Nile, Mekong, Don, Mississippi, Chao Phraya and Fly) that most likely reflect the preferential contribution of radiogenic volcanogenic particles to the finest size fractions of the sediment, no particular correlation exists between $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ and $\Delta\varepsilon_{\text{Nd clay-silt}}$ in studied river sediments ($R^2 = 0.02$; $p \ll 0.001$), hence suggesting the absence of any significant source effect. Previous work showed that the addition of as little as 5–10% of young volcanogenic material could shift the Sr isotopic composition of any sediment towards significantly lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (from *e.g.* > 0.730 to <0.720), without affecting much its ε_{Nd} value (Goldstein, 1988). As a consequence, the potential control of fine volcanogenic particles on the observed size-dependent Sr isotope decoupling was also assessed by plotting $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ *versus* other geochemical indices for basic source rock signatures (*e.g.*, $\text{TiO}_2/\text{SiO}_2$; shale-normalized La/Gd ratios) and the percentage of volcanic rocks in studied river catchments (Table S1). In case of any significant contribution of volcanogenic particles in the clay-rich fractions, one would expect $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ from multi-lithological catchments to display an inverse relationship with both $\text{TiO}_2/\text{SiO}_2$ and the proportion of volcanic rocks, and a positive correlation with $(\text{La/Gd})_{\text{N}}$ ratios. The absence of any particular correlations between the above-mentioned parameters

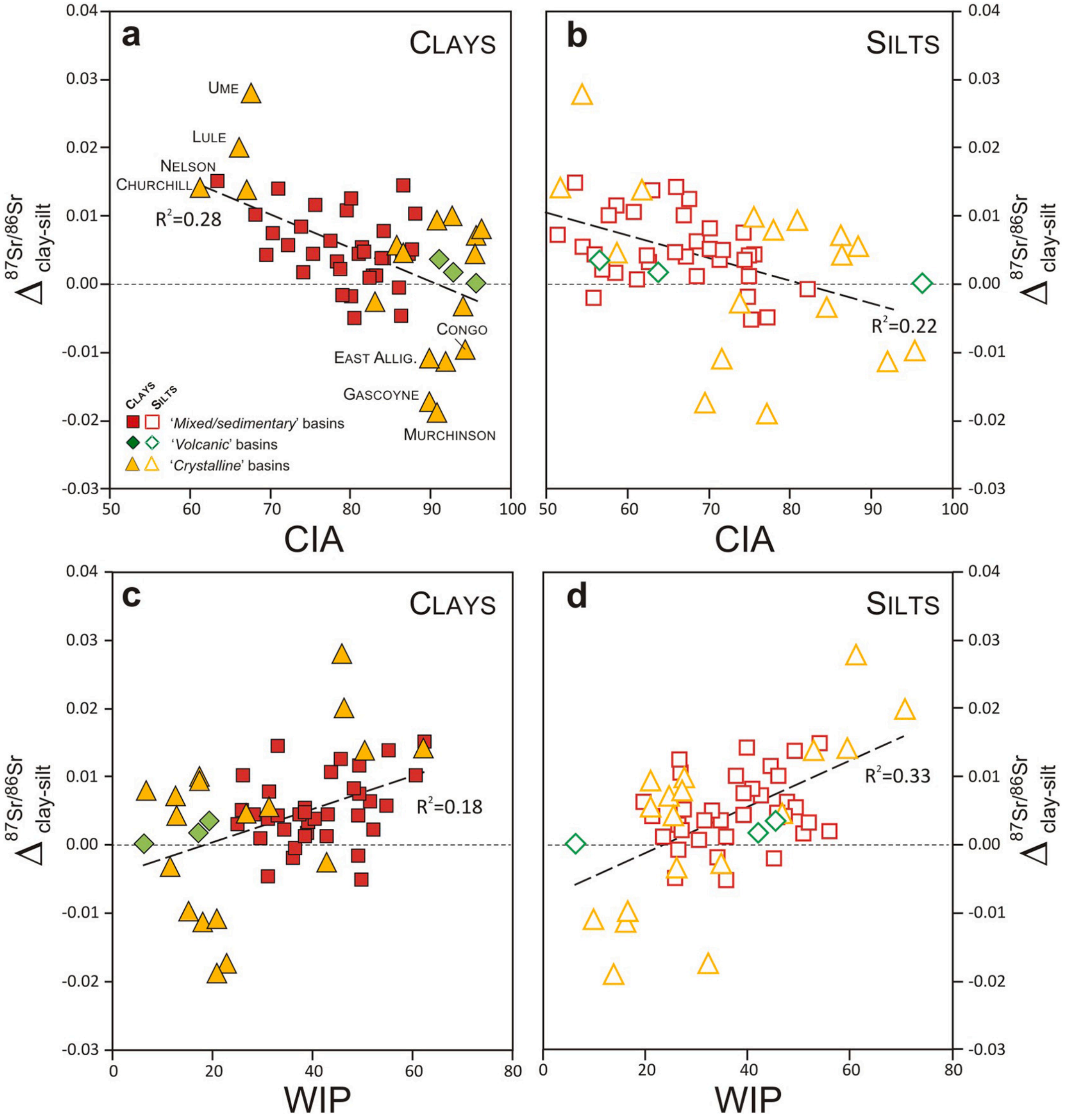

Fig. 6. The relationships between $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ and chemical weathering indices in river clays and silts. (a,b) $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ *versus* the chemical index of alteration (CIA). (c,d) $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ *versus* the weathering index of Parker (WIP). Both river clays and silts define diffuse trends indicating more negative $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ values as their degree of alteration increases. Poorly to moderately weathered sediments (with $\text{CIA}_{\text{clay}} < 75$) are characterized by positive $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ values, while more intensively weathered sediments ($\text{CIA}_{\text{clay}} > 75$) commonly exhibit negative $\Delta^{87}\text{Sr}/^{86}\text{Sr}_{\text{Clay-Silt}}$ values.

(plots not shown here) further suggests that the presence of fine-grained volcanic detritus is unlikely to represent a dominant factor accounting for the observed $^{87}Sr/^{86}Sr$ differences between clay-rich and silt-size fractions. Interestingly, in Fig. 5, a few samples from ancient cratonic areas (*i.e.*, Ume, Lule, Murchinson, Kasai) appear to display significant size-dependent decoupling for both Sr and Nd isotopes. While a source effect cannot be discarded in these particular samples, the occurrence of $\Delta\varepsilon_{Nd\ clay\text{-}silt}$ variability in old crystalline catchments is likely to be caused by incongruent silicate weathering (*e.g.*, Öhlander et al., 2000; Bayon et al., 2015; Dausmann et al., 2019). The effect of incongruent dissolution of silicate rocks on radiogenic Sr isotopes and $\Delta^{87}Sr/^{86}Sr_{\ Clay\text{-}Silt}$ will be discussed below in Sections 4.2 and 4.3.

Additional source effects could also potentially relate to the dissolution of carbonaceous sedimentary rocks and/or rainwater Sr inputs in river catchments; both known to locally influence the Sr budget in soils (*e.g.*, Borg and Banner, 1996; Clow et al., 1997; Capo et al., 1998). In contrast to igneous rocks, the $^{87}Sr/^{86}Sr$ composition of carbonates and other marine-derived precipitates on continents is typically unradiogenic and relatively homogenous, reflecting the fact that seawater chemistry has fluctuated within a limited range of low $^{87}Sr/^{86}Sr$ values over geologic times (between ~0.706 to 0.710; *e.g.*, Veizer, 1989; McArthur et al., 2001). Similarly, the Sr isotopic composition of rainwater is influenced by marine sources and typically exhibit relatively low $^{87}Sr/^{86}Sr$ values (~ 0.706–0.719; see Hajj et al., 2017 for a recent review). In soils, the Sr isotopic composition of the exchangeable cation pool represents a mixture between local dissolved Sr inputs derived from bedrock weathering and atmospheric contributions associated with both dry (dust) and wet (rainfall) deposition (*e.g.*, Miller et al., 1993; Clow et al., 1997; Probst et al., 2000; Pett-Ridge et al., 2009). Presumably, the combination of both carbonate dissolution and atmospheric inputs in soils could thus be accompanied by the incorporation of unradiogenic (seawater-like) Sr upon clay mineral formation, which, in turn, would also affect measured $\Delta^{87}Sr/^{86}Sr_{\ Clay\text{-}Silt}$ values in corresponding river sediments. In this study, we did not observe any particular differences for $\Delta^{87}Sr/^{86}Sr_{\ Clay\text{-}Silt}$ between catchments influenced by intense carbonate weathering (*e.g.*, Seine, Rhine, Northern Dvina; Gaillardet et al., 1999a) and those dominated by silicate weathering (*e.g.*, Orinoco, Parana, Niger, Amazon; Gaillardet et al., 1999a), suggesting that the effect of carbonate weathering on the Sr isotopic composition of clays is probably minor. In addition, while one cannot exclude that atmospheric deposition can influence locally the $^{87}Sr/^{86}Sr$ composition of weathered mineral phases (*e.g.*, Pett-Ridge et al., 2009), it is also likely that any adsorbed Sr fraction associated with clay-size minerals would have been leached away during our sequential leaching procedure. To summarize, all the above consideration suggests that the combined influence of carbonate dissolution, rainwater and any other source effect in river catchments only plays a minor role in controlling the observed distribution of radiogenic Sr isotopes in studied fine-grained detrital fractions.

Finally, the presence of fine-grained sediments recycled from former sedimentary cycles also raises important concern about the utility of Sr isotopes in river sediments for tracing weathering processes. The geochemistry of suspended sediment loads in large river basins is indeed strongly influenced by the recycling of old sedimentary rocks (*e.g.*, Goldstein, 1988; Gaillardet et al., 1999b; Dellinger et al., 2014). Despite this evidence, recent investigations of the same suite of river sediment samples have demonstrated clear correlations between proxies for silicate weathering and modern climatic parameters (Bayon et al., 2016; Bayon et al., 2018; Bayon et al., 2020a). For instance, in the Congo Basin, the Si isotopic composition ($\delta^{30}Si$) of river clays transported in catchments dominated by sedimentary rocks exhibits strong relationships with precipitation levels in corresponding watersheds (Bayon et al., 2018). Similarly, in the Appalachians, the degree of weathering of the same Paleozoic shale formation along a latitudinal climate gradient also exhibits remarkable correlation with modern climatic parameters (Dere et al., 2013). Collectively, these multiple lines of evidence suggest that the weathered material eroded from catchments draining extensive areas of sedimentary rocks can still carry useful information on modern weathering conditions. In other words, this provides additional support to our working hypothesis that measured Sr isotopic ratios in paired clay- and silt-size fractions (and by inference $\Delta^{87}Sr/^{86}Sr_{\ Clay\text{-}Silt}$) can be used to discuss about silicate weathering processes.

4.2. $\Delta^{87}Sr/^{86}Sr_{\ Clay\text{-}Silt}$ as a proxy for silicate weathering intensity

As mentioned in the Results section, the observation that the degree

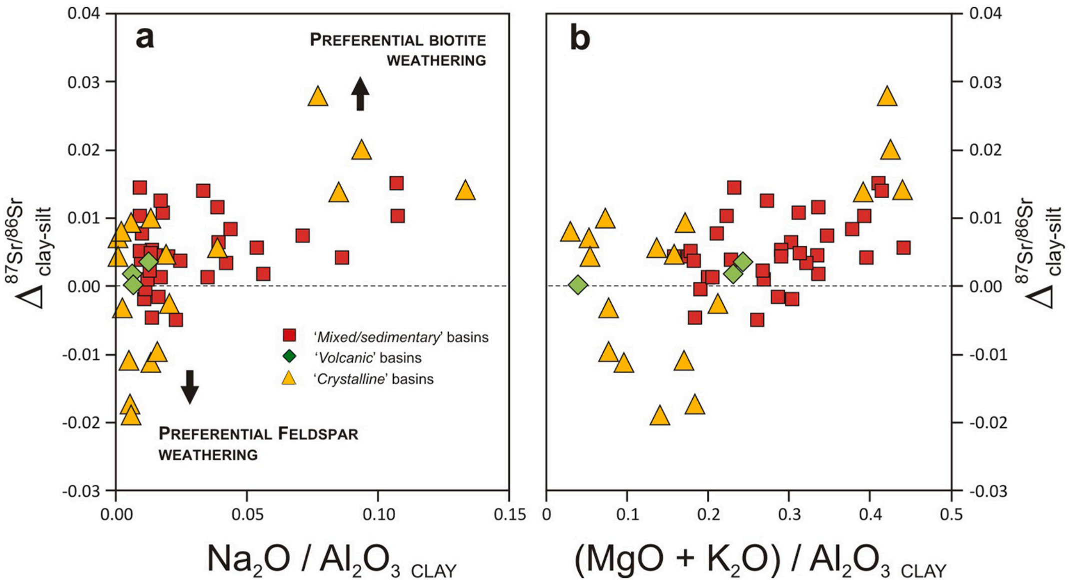

Fig. 7. $\Delta^{87}Sr/^{86}Sr_{\ Clay\text{-}Silt}$ *versus* major element ratios in clays: (a) Na_2O/Al_2O_3 and (b) $(MgO + K_2O)/Al_2O_3$. The observed positive relationships suggest the link between the size-dependent Sr isotopic decoupling in river sediments and the intensity of silicate weathering, which results in the progressive depletion of mobile elements (Na, Mg, K) relative to immobile elements such as Al. In soils, the early alteration of amphibole and biotite (enriched in K and Mg) leads to formation of clay minerals that incorporate a radiogenic Sr isotopic signature compared to the corresponding residual silicate material. As chemical weathering intensifies, the degree of feldspar weathering increases, resulting in kaolinite-bearing mineral assemblages characterized by low Na_2O_2/Al_2O_3 ratios and unradiogenic Sr signatures.

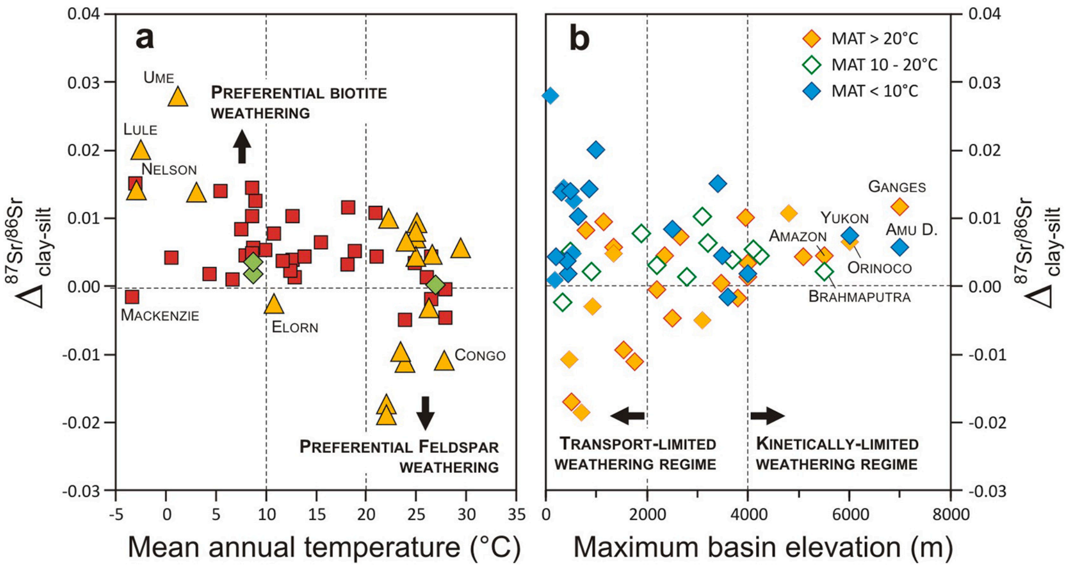

Fig. 8. Relationships between $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ and selected basin characteristics for (a) climate (mean annual temperature, MAT) and (b) topography (maximum basin elevation). In cold regions (MAT <10 °C), positive $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ indicate preferential alteration of biotite in soils, while in warm environments (MAT >20 °C), intense feldspar weathering results in negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$. The degree of size-dependent Sr isotope decoupling is also more pronounced in low-elevation environments (<2000 m), where transport-limited conditions are typically associated with intense silicate weathering processes, than in high mountain regions (>4000 m) dominated by kinetically-limited weathering regimes.

of the size-dependent $^{87}Sr/^{86}Sr$ difference in studied sediments ($\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$) is more pronounced in river catchments draining ancient crystalline rocks can be explained by incongruent weathering processes. Upon rock formation, the radioactive decay of ^{87}Rb to ^{87}Sr results in a range of radiogenic Sr signatures amongst rock-forming minerals that progressively increases through geologic time. As a consequence, the alteration of 'old' silicate rocks can generate weathering products that have markedly different Sr isotopic compositions, while weathering of more recent rocks (such as basalts from recent volcanic provinces) results in secondary clays and residual detrital fractions with $^{87}Sr/^{86}Sr$ signatures that remain closer to the bulk rock composition. On top of this age-dependency of radiogenic Sr isotope decoupling during weathering processes, the degree of silicate weathering intensity also plays an important role in controlling the distribution of $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ in river sediments. This is suggested when plotting $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ against weathering indices (Fig. 6; Fig. 7), such as the chemical index of alteration (CIA), the weathering index of Parker (WIP), and various elemental ratios between mobile and immobile elements (Na_2O/Al_2O_3; $(MgO + K_2O)/Al_2O_3$). The CIA, calculated in molar proportions as $[Al_2O_3/(Al_2O_3 + CaO + Na_2O + K_2O)] \times 100$ (Nesbitt and Young, 1982), provides a quantitative measure of the depletion of mobile (Ca, Na, K) *versus* immobile (Al) elements during chemical weathering, which can be used as an estimate for the degree of feldspar alteration in weathered rocks and sediments. Sediments and sedimentary rocks exhibit a large range of CIA values, which depend on both the lithology of corresponding source rocks and chemical weathering intensity (Nesbitt and Young, 1982). The use of CIA and other weathering indices such as WIP in fine-grained fluvial sediments generally reflects the integrated weathering history of drainage basins at the timescale of soil formation (Li and Yang, 2010; Shao et al., 2012). In Fig. 6a,b, studied river sediments define diffuse trends indicating more negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ values as their CIA increases. The observed correlation between CIA and the degree of size-dependent Sr isotopic decoupling in river sediments is slightly improved when considering the CIA values of clays ($R^2 = 0.28$; $p \ll 0.001$) compared to silts ($R^2 = 0.22$; $p \ll 0.001$). Presumably, this could reflect the fact that silt-size residual fractions contain a greater proportion of primary (unweathered) minerals, meaning that their CIA is likely to be more strongly influenced by lithological effects rather than by their degree of chemical alteration. While poorly to moderately weathered sediments (with $\text{CIA}_{\text{clay}} < 75$) are systematically characterized by positive $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ values (*e.g.*, Lule and Ume rivers; Fig. 6a), more intensively weathered sediments ($\text{CIA}_{\text{clay}} > 75$) commonly exhibit negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ values (*e.g.*, Murchinson, Gascoyne, Kasai; Fig. 6a). Measured $\Delta^{87}Sr/^{86}Sr$ values also appear to yield similar relationships with the plagioclase index of alteration (PIA; calculated in molar proportions as $[(Al_2O_3 - K_2O)/(Al_2O_3 + CaO + Na_2O - K_2O)] \times 100$; Fedo et al., 1995), with coefficient correlations (R^2) of 0.28 and 0.22 for clay- and silt-size fractions, respectively (plots not shown here). Amongst other commonly used weathering indices, WIP ($[(2Na_2O/0.35) + (MgO/0.9) + (2K_2O/0.25) + (CaO/0.7)] \times 100$, in molar proportions; Parker, 1970) is particularly useful when applied to heterogeneous parent rocks (Price and Velbel, 2003), providing a measure of the depletion in the most mobile major cations (Na, K, Mg, Ca) that takes into account their individual mobility in soils, based on their bond strengths with oxygen (Parker, 1970). In this study, $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ and WIP display diffuse relationships, with correlation coefficients (R^2) of 0.18 ($p < 0.005$) and 0.33 ($p \ll 0.001$) in clay- and silt-size fractions, respectively (Fig. 6c,d). The slightly improved degree of correlation observed in river silts probably reflects the fact that WIP is most applicable to mildly leached primary detrital material (Price and Velbel, 2003), rather than to detrital fractions dominated by secondary clay mineral assemblages. In Fig. 6d, the silt-size fractions exhibiting the lowest WIP values (<20; *e.g.*, Murchinson, Kasai), hence showing the most pronounced degrees of mobile cation depletion, are typically associated with negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ values.

To a large extent, the above mentioned general relationships between $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ and weathering indices can be explained by considering both the relative partition coefficients (Kd) of Rb and Sr in rock-forming minerals and the respective susceptibility of different mineral phases to weathering (Fig. 1). In soils, secondary clay minerals are mostly derived from the alteration of ferromagnesian silicate minerals (clinopyroxene, amphibole, biotite) and feldspars (plagioclase, K-feldspar), characterized by relatively high and low Kd_{Rb}/Kd_{Sr} values, respectively (Fig. 1). Because biotite and other ferromagnesian minerals weather more rapidly than feldspars, clay minerals formed in poorly to moderately weathered environments (*e.g.*, chlorite, illite, vermiculite) are expected to incorporate a radiogenic Sr isotopic signature compared

Table 3
Regional and global solid fluxes of riverine Sr and $^{87}Sr/^{86}Sr$ to the ocean.

Regions	Representative rivers used for regional estimates	TSS	CLAYS			SILTS		
			Sr flux		$^{87}Sr/^{86}Sr$	Sr flux		$^{87}Sr/^{86}Sr$
		(Mt/yr)	(t/yr)	(μg/g)			(μg/g)	
America								
American Arctic	Mackenzie, Nelson, Churchill	270	41,732	155	0.7329	37,896	140	0.7285
E. North America	Mississippi	630	44,063	70	0.7266	69,391	110	0.7230
Western Americas	Yukon, Fraser	1530	1,36,822	137	0.7150	2,29,128	229	0.7089
N.E. South America	Amazon, Orinoco	1600	2,09,338	93	0.7289	2,68,198	124	0.7232
S. South America	Parana	120	9529	79	0.7390	8846	74	0.7284
Africa								
W. Africa	Congo, Niger	550	29,657	54	0.7358	83,585	152	0.7235
E. Africa	Nile	400	45,808	115	0.7135	56,265	141	0.7155
Eurasian Arctic / Russia	Northern Dvina, Volga	150	17,125	114	0.7222	21,100	141	0.7202
Europe								
Scandinavia	Narva, Kymijoki, Ume, Lule	20	1753	88	0.7720	1752	88	0.7538
W. Europe	Rhine, Loire, Vistula, Seine	25	1781	71	0.7333	2168	87	0.7296
Iceland	Maine	48	2197	46	0.7067	6519	136	0.7050
Black Sea	Danube, Don	140	9120	65	0.7237	14,353	103	0.7187
Western Med. Sea	Adour	620	98,590	159	0.7174	66,557	107	0.7162
S.E. Asia								
E. Asia	Yangtze, Mekong, Red, Yellow, Chao Phraya	2500	1,85,997	74	0.7277	3,41,001	136	0.7193
E. Himalaya	Amu-Darya	500	51,338	103	0.7185	92,678	185	0.7129
Bay of Bengal	Ganges, Brahmaputra	2100	1,93,558	92	0.7367	3,18,828	152	0.7308
Indonesia & W. Pacific	Fly, Sepik, Brantas	7230	7,54,228	104	0.7112	1,097,486	152	0.7087
Australia	Murray, Fitzroy, East Alligator	170	12,810	75	0.7290	9765	57	0.7352
World Rivers		18,600	18,45,000	100	0.7197	2,725,000	145	0.7156
Global riverine fluxes of Sr to the ocean			(Mt/yr)	(μg/g)	$^{87}Sr/^{86}Sr$			
Detrital load (This study; 85% silts - 25% clays)			2.52	138	0.7160			
Suspended load (Goldstein and Jacobsen, 1988)			2.98	221	0.7160			
Dissolved load (Peucker-Ehrenbrink and Fiske, 2019)			4.18	0.10	0.7111			
Dissolved load (Goldstein and Jacobsen, 1987)			2.52	0.06	0.7101			

to the corresponding residual silicate material. As chemical weathering intensifies, ferromagnesian minerals become depleted in soil profiles and the degree of feldspar weathering increases (*e.g.*, Nesbitt and Young, 1989), resulting in kaolinite-bearing mineral assemblages depleted in labile cations (Ca, Na, Mg, K) and presumably characterized by unradiogenic Sr signatures. This presumed relationship between $^{87}Sr/^{86}Sr$ and chemical weathering intensity in soils is generally well supported by field observations. For example, an investigation of glacial soil chronosequences in Wyoming documented a $^{87}Sr/^{86}Sr$ shift in both exchangeable and weathered mineral fractions towards less radiogenic signatures with increasing soil age, indicative of progressively intensifying weathering of unradiogenic plagioclase over biotite with time (Blum and Erel, 1997). In more intensively weathered soil sequences, the predominance of feldspar weathering is commonly associated with the presence of kaolinite characterized by low Sr isotopic compositions (*e.g.*, White et al., 2001; Pett-Ridge et al., 2009). While the degree of feldspar weathering probably largely accounts for the observed relationships between $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ and weathering indices (Fig. 6; Fig. 7), the cause for the negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ values observed in highly weathered environments remains unclear. Understanding the occurrence of such negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ signatures requires identification of the mineral phases that are likely to drive residual silt fractions towards more radiogenic compositions than corresponding clays. We propose that this effect could be possibly explained by the particular behaviour of biotite in soils, which, unlike feldspars, weathers by a mechanism in which the primary phyllosilicate lattice is generally conserved (*e.g.*, Velbel, 1985; Kretzschmar et al., 1997). Altered grains of biotite are typically composed of a core of biotite, intertwined and surrounded by variable amounts of kaolinite, which may possibly act as a protective layer under intense weathering conditions (*e.g.*, Kretzschmar et al., 1997; Murphy et al., 1998). This would be in agreement with previous evidence for the persistence of altered grains of biotite in highly weathered soil horizons, after all other primary minerals had completely weathered out (Pett-Ridge et al., 2009). Presumably, such kaolinite pseudomorphs of biotite could represent a substantial source of radiogenic Sr in the residual coarse fraction of tropical soils otherwise dominated by feldspar weathering, thereby possibly accounting for the observed negative $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ in our study.

To some extent, the observed dispersion of $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ in river sediments could also be partly controlled by the alteration of calcium and phosphate trace minerals in soils, such as sphene, apatite and allanite, which all display low Kd_{Rb}/Kd_{Sr} values (Fig. 1). In granitic environments, these accessory phases are dissolved during the early stages of chemical weathering (*e.g.*, Harlavan and Erel, 2002; Erel et al., 2004; Oliva et al., 2004), releasing substantial amounts of unradiogenic Sr in soil and river systems (*e.g.*, Aubert et al., 2001). In weathered soil sequences, apatite dissolution is typically followed by precipitation of more resistant secondary phosphate minerals, such as rhabdophane and florencite, which integrate elements previously released during early weathering stages, until more intense weathering conditions ultimately lead to their dissolution (*e.g.*, Banfield and Eggleton, 1989; Taunton et al., 2000; Oliva et al., 2004). Therefore, in addition to the relative contribution of biotite *versus* feldspar weathering, the successive phases of dissolution/precipitation of accessory phosphate minerals in soils could also possibly account for some of the observed size-dependent decoupling of Sr isotopes in river sediments.

4.3. Climatic and tectonic controls on $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ in modern river sediments

Many earlier laboratory and field studies have provided evidence for the dependence of silicate mineral dissolution rates on climate and tectonics (*e.g.*, Velbel, 1993; Chen and Brantley, 1997; Murphy et al., 1998; Brantley et al., 1998; White et al., 1999; West et al., 2005; West, 2012; Dere et al., 2013; Li et al., 2016). The climatic and geomorphic characteristics of studied catchments can hence be used to further investigate the mechanisms controlling the size-dependent $^{87}Sr/^{86}Sr$ decoupling in modern river sediments worldwide. Amongst the various parameters affecting silicate weathering on continents (*e.g.*, temperature, precipitation, denudation rates, elevation, slope, soil thickness, curvature), we examine in Fig. 8 the relationships between $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ and the mean annual temperature (MAT; Fig. 8a) and maximum basin elevation in studied catchments (Fig. 8b), which can be used, to a first approximation, as proxies for the climate and tectonic controls. Note that no particular relationships were found between $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ and the catchment size of studied river basins (graph not shown here), meaning that the observed $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ variability does not reflect any levelling effect that would occur on the geochemical signatures of suspended particulates in the largest river catchments. Of course, such a global approach suffers from inherent limitations due to the fact that major river basins commonly display large temperature gradients across their watershed between warm floodplains and cold high-elevation regions, such as for instance in the Amazon and the Himalayan river basins. Nevertheless, many studies have previously shown that the variability of weathering indices in river systems indicates significant relationships with MAT, providing useful information on the climatic control of chemical weathering in catchments (*e.g.*, Gaillardet et al., 1999a; Gaillardet et al., 2019; Li and Yang, 2010; Bayon et al., 2016, 2018, 2020a). In Fig. 8, a striking feature is that, except for two sediments from the Mackenzie River ($\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}} = -0.002$) and Elorn ($-0.003$), all the samples yielding negative $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ correspond to tropical or sub-tropical river basins with MAT >20 °C. As discussed above, this finding is best explained by enhanced alteration of feldspars under intense weathering conditions, resulting in the formation of kaolinite-bearing mineral assemblages characterized by unradiogenic Sr signatures. In contrast, the highest $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ values ($> +0.012$) are encountered in sediments from sub-Arctic or temperate river basins with MAT <10 °C (Fig. 6). In cold and glacial environments, the alteration of biotite strongly dominates over feldspar weathering (*e.g.*, Blum et al., 1993; Anderson et al., 1997). As discussed above, this process results in the preferential release of dissolved radiogenic Sr signatures and the formation of weathered products characterized by high $^{87}Sr/^{86}Sr$ ratios (Blum et al., 1993; Blum and Erel, 1997). In this study, the same mechanism most likely accounts for the high $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ values observed in most sub-Arctic rivers (Churchill, Nelson, Narva, Ume, Lule, Kymijoki). These observations are in full agreement with previous experimental works showing that at lower temperature, the initial cation release from biotite is significantly faster than cation release from plagioclase, and *vice versa* (White et al., 1999).

In addition to climate, tectonics also plays a major role in controlling silicate weathering rates in watersheds (*e.g.*, Raymo and Ruddiman, 1992; Riebe et al., 2001; Jacobson et al., 2003; West et al., 2005; West, 2012; Hartmann et al., 2014). High mountains are characterized by high physical erosion rates and thin soils, where kinetically-limited conditions generally reduce silicate weathering intensity. In high elevation environments, the rate of sediment supply is generally more rapid than silicate mineral dissolution, so that limited $^{87}Sr/^{86}Sr$ decoupling is expected to occur during weathering. In Fig. 8b, this is illustrated by the fact that all rivers draining areas of high topography (with maximum elevation >4000 m; *e.g.* Ganges-Brahmaputra, Amu-Darya, Orinoco, Yukon, Amazon) display a very limited range of $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ values (between $+0.002$ and $+ 0.012$). As elevation decreases, physical erosion rates decrease and soils thicken, leading to an increase in the proportion of silicate mineral dissolution relative to sediment supply. In this context, enhanced silicate weathering can result in more pronounced decoupling of Sr isotopes between secondary clays and residual weathered products, as exemplified by the comparatively larger range of $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ (from -0.005 to $+0.015$) observed in the elevation range of 2000 to 4000 m (Fig. 8b). In Fig. 8b, the sediments exhibiting the most pronounced size-dependent $^{87}Sr/^{86}Sr$ decoupling (with $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ between -0.019 to $+0.028$) correspond to rivers draining areas of low topography (< 2000 m), typically characterized by thick soil sequences and low denudation rates. In such transport-limited weathering regimes, the longer sediment residence time in soils leaves sufficient time for silicate mineral weathering to proceed. This implies that significant $^{87}Sr/^{86}Sr$ decoupling can occur between weathering products, leading either to mostly negative $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ in tropical regions dominated by feldspar weathering, or to positive $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ in sub-Arctic environments dominated by biotite alteration (Fig. 8b). Importantly, the proposed mechanism linking the degree of size-dependent $^{87}Sr/^{86}Sr$ decoupling in river sediments to the type of weathering regime in watersheds will be also dependent on the fact that ancient Precambrian shields are generally encountered in areas of low topography, while high elevation regions correspond to geologically young orogenic belts. This means that the observed trends between $\Delta^{87}Sr/^{86}Sr_{\text{ Clay-Silt}}$ and the maximum elevation in river basins are also crucially dependent on the age of the source rocks, hence representing a time-integrated relationship.

4.4. *Regional and global solid fluxes of riverine Sr and $^{87}Sr/^{86}Sr$ to the ocean*

Taken together, the cumulative area of the river basins investigated in this study accounts for a total of 37.5×10^6 km^2, hence representing about 35% of the entire continental area that drains into the global ocean (105×10^6 km^2; Milliman and Farnsworth, 2011). For each continental region (*e.g.*, Scandinavia, Western Europe, West Africa, East Asia), the largest and/or most representative rivers were used to extrapolate regional solid fluxes of riverine Sr ($F_{Reg.}Sr$) and associated Sr isotopic composition ($(^{87}Sr/^{86}Sr)_{Reg.}$) following the equations listed below:

$$F_{Reg}Sr = \frac{\sum TSS_i.C_iSr}{\sum TSS_i} \times TSS_{Reg}$$

$$\left(^{87}Sr/^{86}Sr\right)_{Reg} = \frac{\sum TSS_i.C_iSr\left(^{87}Sr/^{86}Sr\right)_i}{\sum TSS_i.C_iSr}$$

where TSS_i, C_iSr and $(^{87}Sr/^{86}Sr)_i$ refer to the total suspended load (Mt/yr), Sr concentration and associated Sr isotopic compositions in either clay- or silt-size detrital fractions (µg/g) of individual river systems; and $TSS_{Reg.}$ represents the regional TSS value (Milliman and Farnsworth, 2011). An inherent uncertainty in this approach is the fact such regional Sr fluxes are strongly dependent on the $^{87}Sr/^{86}Sr$ and [Sr] values used for the largest river basins, where suspended particulate loads have been shown to display significant seasonal variability over the hydrological year (*e.g.*, Viers et al., 2008; Mao et al., 2011; Rousseau et al., 2019). As a consequence, the Sr regional fluxes calculated here should be only regarded as informative. Nevertheless, the significant $^{87}Sr/^{86}Sr$ seasonal variability previously identified in suspended particles from large river catchments can be generally ascribed, to a large extent, to a grain-size effect (*e.g.*, Bouchez et al., 2011). In this study, this effect is minimized because we analysed separate fractions of both clay-rich and silt-size detrital sediments. In addition, while the $^{87}Sr/^{86}Sr$ composition of suspended loads represents an instantaneous snapshot of the chemical composition of river particulates, fine-grained sediments deposited near the mouth of rivers integrate spatially-integrated source signatures over a much longer period of time. Therefore, we are confident that the approach described above can still provide indicative estimates of regional Sr fluxes worldwide. The obtained Sr fluxes are directly proportional to regional sediment yields (Table 3), indicating that about 40% of the solid Sr riverine flux to the world ocean is derived from Indonesia and the Western Pacific area, followed by contributions from South East Asia and the large Himalayan rivers (~25%) and northern South America (~10%, corresponding mainly to the Amazon River). In Europe, the total sediment flux to the ocean is dominated by inputs from Western Mediterranean regions, which account for more than ~70% of associated solid Sr fluxes. During weathered Collectively, our new global estimates for the total solid Sr fluxes from rivers to the ocean are about 1.8 and 2.7 Mt./yr for river clays and silts, respectively (Table 3). The corresponding average Sr concentration and $^{87}Sr/^{86}Sr$ of the clay- and silt-size detrital fractions, calculated using the equations listed below, are 100 and 145 μg/g, and 0.7197 and 0.7156, respectively (Table 3), which confirms that the river clays exported to the ocean are, at the global scale, slightly more radiogenic than corresponding silts.

$$[Sr]Global = \frac{\sum_{Reg} TSS \times [Sr]Reg}{\sum_{Reg} TSS}$$

$$\left(\frac{87Sr}{86Sr}\right)Global = \frac{\sum_{Reg} TSS \times [Sr]Reg \times \left(\frac{87Sr}{86Sr}\right)Reg}{\sum_{Reg} TSS \times [Sr]Reg}$$

To the best of our knowledge, there are no estimates for the proportion of clays and silts associated with the suspended particulate loads delivered to the ocean. Considering the average proportion of each grain-size fraction in the studied river sediment samples (*i.e.* 85% silts and 15% clays; Bayon et al., 2016), we can calculate a global $^{87}Sr/^{86}Sr$ composition of 0.7160 for the average river sediment exported to the ocean, being coincidently identical to the value proposed more than 30 years ago by Goldstein and Jacobsen (1988).

5. Conclusions

This study presents new information about the factors controlling the distribution of radiogenic strontium isotopes in river sediments. We show that the Sr isotopic difference between clay- and silt-size fractions, termed here $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$, is related to the intensity of silicate weathering in river basins. The observed range of $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ in river sediments worldwide largely reflects the temperature dependence of silicate weathering and its influence on the preferential alteration of radiogenic biotite *versus* unradiogenic feldspar in cold and warm environments, respectively. Our global survey for Sr isotopes in river sediments also suggests a link between $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ and geomorphic parameters such as the maximum basin elevation, reflecting the fact that silicate weathering is most intense in areas of low topography, as opposed to high-mountain environments characterized by kinetically-limited weathering regimes. Future studies should now aim at assessing whether these findings hold true at the local scale, when investigating weathering profiles. Ultimately, the application of this new $\Delta^{87}Sr/^{86}Sr_{\text{Clay-Silt}}$ weathering proxy to sedimentary records could ideally complement the conventional use of radiogenic Sr isotopes as provenance tracers, thereby providing a unique set of geochemical tracers for reconstructing the evolution of past landscapes and silicate weathering over geologic times.

Declaration of Competing Interest

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemgeo.2020.119958.

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