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SEDIMENT FINGERPRINTING IN THE CRITICAL ZONE

Preface—evaluating the response of critical zone processes to human impacts with sediment source fingerprinting

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1 Background: critical zone processes in the Anthropocene

The Earth's critical zone encompasses a suite of interconnected processes in the near-surface lithosphere, pedosphere, biosphere, atmosphere, and hydrosphere (Brantley et al. 2007; Lin 2010) (Fig. 1). Processes and interactions both within and between these various critical zone components support life-sustaining ecosystem services and resources that establish the foundation for humanity (NRC 2001). This includes the production of fertile soils; flourishing vegetation; productive rivers, lakes, and oceans; and our life-sustaining atmosphere (Gaillardet 2014; Guo and Lin 2016).

Rapid population growth, land-use intensification, and global environmental change are disturbing many of these fundamental critical zone processes. More than half of the Earth's terrestrial surface is now impacted by anthropogenic activities (e.g., clearing, grazing, plowing, mining, and logging) (Richter and Mobley 2009; Hooke et al. 2012). These changes are so widespread and pervasive that the great acceleration of socioeconomic development that occurred around 1950 (Fig. 2) has been identified as the dawn of the Anthropocene (Waters et al. 2016). Although the utility of adopting and delineating the Anthropocene as the current epoch is subject to debate (Crutzen 2002; Smith and Zeder 2013; Ruddiman et al. 2015), the concept effectively highlights both the nature and the extent of our global impact on Earth's critical zone.

Soil-forming processes and ecosystem services provided by the pedosphere are central to the critical zone (Lin 2010; Banwart et al. 2011). Many of these processes have been disturbed by the agricultural intensification that coincided with the great acceleration resulting in unsustainable land-use practices now outpacing soil formation processes (Brantley et al. 2007). As agricultural landscapes now cover an area equivalent to what was scoured during the last glacial maximum (Amundson et al. 2007), the broad-scale intensification of anthropogenic activities has resulted in significant on- and off-site impacts. On-site, soil loss has resulted in decreases in soil fertility and agricultural yields (Ladha et al. 2009) threatening the ability to feed the world's growing population (Brantley et al. 2007). Off-site, the excess delivery of particulate matter downstream is degrading riverine, lacustrine, and estuarine ecosystems (Clark 1985; Owens et al. 2005; Bilotta and Brazier 2008; Smith et al. 2018).

The challenge, as noted by Brantley et al. (2007), is that despite our society having over 10,000 years of experience working with soils, our conceptual and quantitative models remain inadequate at predicting critical zone dynamics under current conditions. Notwithstanding growing pressure for improved environmental management, we still have a limited capacity to predict changes in the critical zone in response to anthropogenic activities owing to the multiple spatial and temporal scales at which these complex processes and feedbacks are manifested. As river basin systems are impacted by many of these processes, a deeper understanding of the dynamics of the soil-sediment continuum may provide a valuable framework for evaluating the disturbance response of critical zone processes. Understanding these processes may also provide

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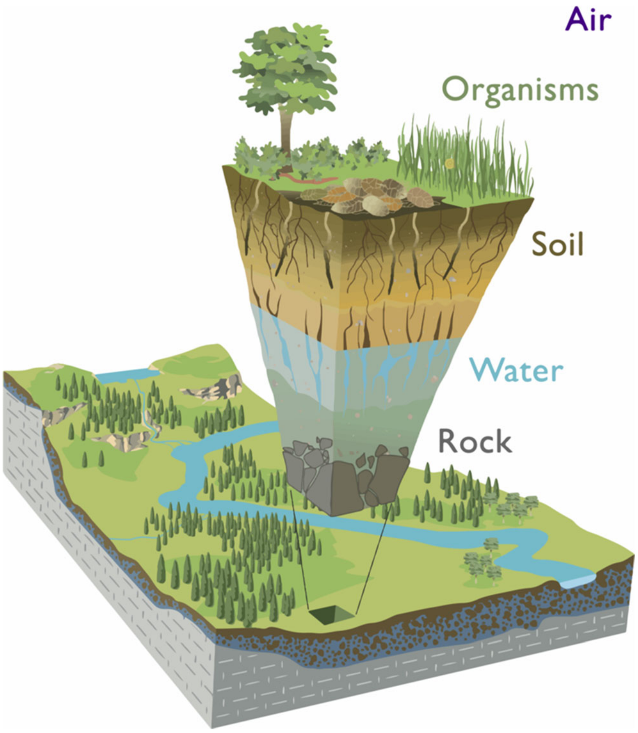

Fig. 1 Conceptual illustration of the critical zone, courtesy: National Science Foundation (NSF-CZO, 2019)

land and resource managers with the information necessary to manage both the on-site and off-site effects of accelerated soil erosion.

2 Sediment source fingerprinting

The sediment source fingerprinting technique is well suited to investigate critical zone processes within river basins. The dynamics of material transported from source to sink through river networks reflect physical and biogeochemical processes occurring in the critical zone (Amundson et al. 2007). Accordingly, the sediment fingerprinting technique is uniquely situated to investigate these processes across a range of spatial and temporal scales, from small fields to regional river basins and from individual rainfall events to decadal timeframes.

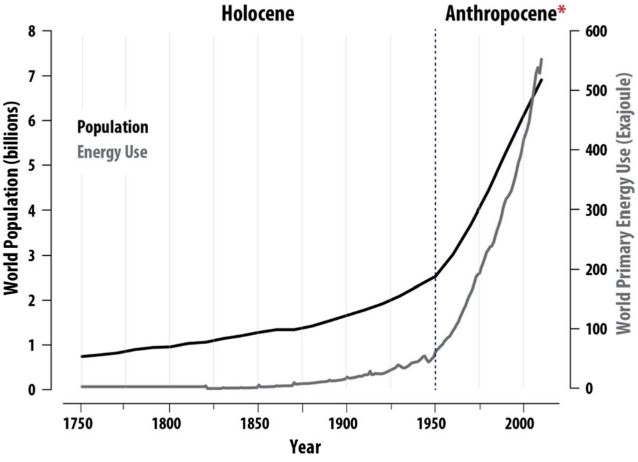

Fig. 2 The dawn of the Anthropocene, delineated by the great acceleration in 1950, which is highlighted in this figure by an exponential increase in the global population and primary energy use (after Steffen et al. 2015)

The sediment source fingerprinting technique uses a variety of physical and biogeochemical parameters, or fingerprints, to trace sediments back to their sources (Fig. 3). For parameters to be effective fingerprints, they need to discriminate between sediment sources while behaving conservatively (Walling et al. 1993; Collins et al. 1996). Conservative fingerprints remain constant during the erosion cycle (sediment detachment, entrainment, transportation, deposition, and delivery), or vary in a predictable way (Davis and Fox 2009; Koiter et al. 2013b; Belmont et al. 2014; Laceby et al. 2017). A variety of fingerprints have been used to investigate sediment dynamics within river basins, such as mineral magnetic properties, fallout radionuclides, color properties, major and trace element geochemistry, carbon and nitrogen isotopes, and compound-specific stable isotopes (Walling and Kane 1984; Caitcheon 1993; Murray et al. 1993; Martínez-Carreras et al. 2010; Evrard et al. 2011; Blake et al. 2012; Hancock and Revill 2013; Laceby et al. 2016). Fingerprints that discriminate between the sources of interest (e.g., land use, soil types, geology, surface versus subsoils) are used to estimate relative source contributions to target sediment with end-member mixing models that are generally solved stochastically in frequentist (Walling et al. 1993; Collins et al. 2012; Tiecher et al. 2019) or Bayesian frameworks (Small

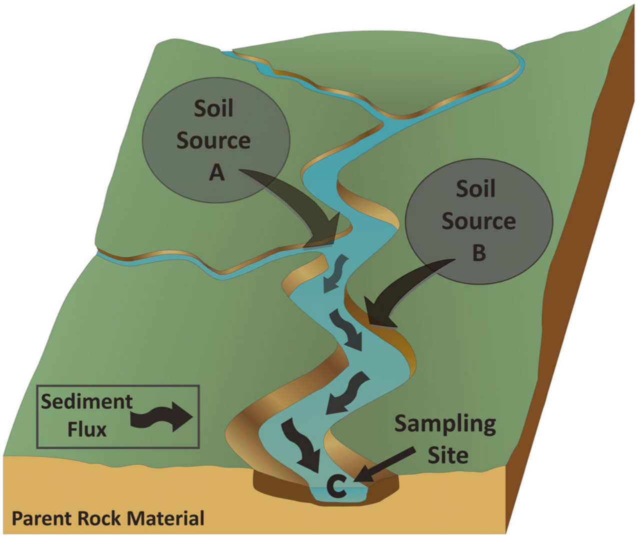

Fig. 3 A simplified two source conceptual model of the sediment source fingerprinting technique where end-member mixing models solve equations (i.e., $Ax + B\,(1 - x) = C$) to determine the relative contributions (x) of source A and source B to the target sediment (i.e., C in the equation) obtained from the sampling site

et al. 2002; Cooper and Krueger 2017; Davies et al. 2018). The sources discriminated in sediment fingerprinting are typically related to the scale of the study catchment, the complexity of land use, land cover, soil types, and geology, along with the fundamental objectives of the research and/or management program.

Sediment source fingerprinting research originally had a strong focus on understanding erosion dynamics (Wood 1978; Oldfield et al. 1979; Longmore et al. 1983; Stott, 1986; Wallbrink and Murray 1993; Wallbrink et al. 1998). Thereafter, this research started to increasingly focus on identifying sediment sources in the context of developing effective sediment management strategies (Wallbrink 2004; Walling 2005; Davis and Fox 2009; Porto et al. 2010; Gellis and Walling 2011; Mukundan et al. 2012). This emphasis on developing a sediment fingerprinting management tool coincided with a significant research focus on advancing end-member mixing modeling techniques and reducing mixing model uncertainty (Collins et al. 2012; Cooper et al. 2015; Laceby and Olley 2015; Pulley et al. 2015; Sherriff et al. 2015).

Over the last several decades, there has been significant progress in the application and development of the sediment source fingerprinting technique to contribute to the improved management of water bodies around the world. There have been multiple comprehensive review papers on fingerprinting techniques (Collins and Walling 2002; Davis and Fox 2009; D'Haen et al. 2012; Guzmán et al. 2013; Haddadchi et al. 2013; Koiter et al. 2013b; Smith et al. 2013; Owens et al. 2016; Davies et al. 2018) and several special issues dedicated to advancing the sediment source fingerprinting technique and facilitating targeted sediment management strategies (Gellis and Mukundan 2013; Walling et al. 2013; Smith et al. 2015; Collins et al. 2017). Although the sediment source fingerprinting technique has advanced significantly, it has simultaneously drifted away from one of its original foci, researching fundamental Earth system science processes.

Error analysis and uncertainty associated with sediment source fingerprinting has also significantly improved using methods such as Monte Carlo substitutions (Kraushaar et al. 2015; Gellis and Gorman Sanisaca 2018; Collins et al. 2019), virtual mixtures (Laceby and Olley 2015; Palazón et al. 2015; Collins et al. 2019), synthetic mixtures (Sherriff et al. 2015), and Bayesian uncertainty estimations (i.e., Markov chain Monte Carlo framework) (Small et al. 2002; Stewart et al. 2015).

Limitations still exist within the sediment source fingerprinting framework including spatial and temporal challenges that may affect the interpretation of individual studies. Temporal limitations include the time period of source assessment where longer time periods may be required to characterize seasonal sources (i.e., during cultivation), the general hydrology of the study area, and sample large storm events. The main challenge is that longer study periods involve significant additional costs. Sediment sources may also change over the storm hydrograph and integration of sediment for an entire storm based on individual samples may not reflect the true source contributions (Carter et al. 2003; Nosrati et al. 2018). Separating target samples into rising and falling stages may allow for the interpretation of sediment sources over the hydrograph (Carter et al. 2003; Belmont et al. 2014) but require proper instrumentation and collection of enough sediment mass for analysis. Spatial limitations in sediment source fingerprinting include the catchment area, whereas size increases the number of samples collected to characterize sources, along with additional costs (Nosrati and Collins 2019). Parts of the watershed that are difficult to access because of landowner permission, or are remote, may also present a challenge for acquiring representative datasets (Nosrati and Collins 2019). Finally, while sediment fingerprinting quantifies the general sources of delivered sediment to the point of interest, it does not highlight specific locations or "hot spots" of erosion within a watershed, which often need to be identified with sediment budgets or other approaches (Gellis and Walling 2011).

3 Sediment source fingerprinting in the critical zone

This special issue presents a series of research articles demonstrating how sediment source fingerprinting research can be used to investigate a variety of critical zone processes. Understanding critical zone processes and their response to human impact is imperative for adapting to global change and meeting United Nations' sustainable development goals (Griggs et al. 2013). Therefore, we hope to demonstrate how the sediment source fingerprinting technique offers potential to further our understanding of critical zone processes in river basin systems around the world. Accordingly, in this section, we highlight the key contributions from the research papers in this special issue from multiple researchers applying the sediment source fingerprinting technique in Asia, Europe, North America, and South America.

Uber et al. (2019) investigate the spatial origin of suspended sediment in two nested catchments (i.e., Claduègne and Gazel Basins, France) from a French critical zone observatory network: the Cevennes-Vivarais Mediterranean Hydrometeorological Observatory. Critical zone observatories are important collaborative platforms for research that often operate at the watershed scale and focus on the interconnected chemical, physical, and biological processes shaping Earth's surface. At this particular hydro-meteorological observatory, these authors incorporate multiple suites of fingerprints (i.e., color, X-ray fluorescence, and magnetic susceptibility) to investigate whether sediment is derived from erosion processes on badlands, sedimentary topsoils, or basaltic topsoils. Uber et al. (2019)

found that erosion processes on the badlands contributed between 74 and 84% of the suspended sediment, followed by erosion processes on sedimentary (12–29%) and basaltic (1–8%) surface soils. Importantly, these authors demonstrated that the choice of the fingerprints included in the mixing model had a larger impact on the model results than the actual model used to apportion sediment sources. One of Uber et al.'s (2019) key findings is the importance of using multi-fingerprint and multi-model techniques to detect and quantify potential biases (e.g., source variability, particle-size selectivity) in order to obtain reliable and robust estimates of source contributions to target sediment.

Batista et al. (2019) examine how pedogenetic processes in soils help influence the development of the geochemical signals that are used in sediment source fingerprinting research. Their research in the Ingaí River Basin (Brazil) incorporated a tributary fingerprinting technique, multiple particle-size fractions, and artificial mixtures to understand erosion dynamics in three areas of this basin: the upper, middle, and lower regions. In particular, Batista et al. (2019) found that erosion processes on Ustorthrent soils from the lower catchment were dominating the supply of sediment at the basin outlet. In particular, these authors reported that using different techniques to select elements for inclusion in mixing models (e.g., knowledge and statistics based), along with artificial mixtures, helped provide multiple lines of evidence necessary to produce robust estimates of source contributions to target sediments. Batista et al. (2019) illustrate the importance of understanding how pedogenetic processes drive source signal (i.e., fingerprint) development. In closing, Batista et al. (2019) argue that the use of knowledge-based techniques to select fingerprints for modeling will encourage researchers to further develop their understanding of processes that drive erosion and sediment geochemistry across multiple spatial and temporal scales.

Evrard et al. (2019a) use colorimetric fingerprints to investigate the contribution of different erosion processes to material transiting the Mano and Niida catchments in the Fukushima Prefecture, Japan. The authors demonstrated that erosion processes on cultivated landscapes supplied the majority of sediment (56%) to the river networks followed by subsoil sources (including decontaminated materials—26%) and forest sources (21%). The relative contribution of these sources changed over time owing to the implementation of decontamination activities in the region and also to the occurrence of major rainfall events, including typhoons. Importantly, the authors concluded that the relatively inexpensive, rapid, and non-destructive colorimetric measurements have significant potential to provide comprehensive information on erosion processes occurring in the critical zone.

Boudreault et al. (2019) combine the use of colorimetric fingerprints with fallout radionuclides to compare different sampling designs for sediment source fingerprinting in an agricultural catchment in Atlantic Canada. Specifically, the authors investigated whether suspended sediments were derived from streambanks, agricultural topsoil, or forested areas. Boudreault et al. (2019) used a novel nested approach in their sample design, including five sites with drainage basins ranging from 3.0 to 13.4 km^2. These authors determined that sediment sampled in the headwaters was predominantly derived from erosion processes in forested areas. Progressing downstream, the authors illustrated that erosion processes on agricultural landscapes started to dominate the supply of sediment in the Black Brook watershed. Boudreault et al. (2019) reported that sediment was mainly derived from local sources rather than upstream sediment entering an individual subcatchment, highlighting the importance of assessing sediment sources over a range of spatial scales to understand geomorphic connectivity.

Kitch et al. (2019) use elemental geochemistry to investigate the sediment source contributions to both suspended sediment and channel bed sediment in the Merriott Stream catchment in rural Somerset, UK. In particular, the authors investigate how upstream agricultural land-management practices impact overland flow generation and affect downstream fluvial processes. Kitch et al. (2019) found that while cultivated landscapes were the dominant source of suspended sediment, channel bank erosion was the main source of channel bed sediment. The authors attributed differences in the suspended versus channel bed sources to in-channel incision and bank failure. Importantly, Kitch et al. (2019) highlight how there are likely different processes driving the source dynamics for suspended sediment and sediment deposited on the channel bed. These differences provide not only useful comparisons to help understand sediment source and storage dynamics, but they also provide fundamental information for targeted management strategies focusing on upstream processes that may be responsible for deleterious particulate material migrating downstream.

Mahoney et al. (2019) use a sediment fingerprinting technique to investigate the equilibrium sediment exchange processes in the Upper South Elkhorn Basin in the USA. In particular, these authors use carbon stable isotopes ($\delta^{13}C$) to help understand the instantaneous deposition and erosion of suspended sediment on, and from, the streambed. Mahoney et al. (2019) found that streambed sediments were an important source of suspended sediment and dominated the supply of sediment in the fluvial load for low and moderate flow events. In contrast, during high and extreme flow events, upland sources became increasingly important. These authors demonstrated that the equilibrium sediment exchange is a potentially important critical zone process in many riverine systems. Researchers should therefore be cognizant of streambeds that may behave as a potential source. This may be particularly important when using stable isotope signatures or other fingerprints that may potentially undergo biotransformation processes (e.g., diagenesis) when deposited in the riverbed. Furthermore, Mahoney et al. (2019) highlight the importance of coupling sediment source fingerprinting techniques with watershed-modeling research to help develop a potentially new class of sediment transport studies with combined fingerprinting/watershed modeling research designs. Importantly, Mahoney et al. (2019) provide six important conclusions for helping develop these new coupled models that will help drive future research programs.

Gateuille et al. (2019) combine fallout radionuclides (^{137}Cs and ^{210}Pb$_{xs}$) with elemental geochemistry to investigate the main sediment sources (i.e., stream bank, agricultural, and forest) in the Nechako River Basin in British Columbia, Canada. Not only did these authors investigate how these erosion sources vary spatially across this catchment, but they also investigated how these sources changed over time based on the analyses of a sediment core from an island on the main stem of the Nechako River. For the main-stem sites, channel bank erosion processes dominated the supply of sediment, particularly downstream of active cut banks or areas where the floodplains are actively eroding. Progressing downstream, there was an increase in sediment derived from erosion processes on agriculture and forested landscapes consistent with the changing land use in the catchment. Gateuille et al. (2019) also found that the construction of a dam in the 1950s resulted in a significant alteration of the sediment transport capacity in the Nechako River Basin, resulting in a change in sediment source dynamics. Overall, Gateuille et al. (2019) demonstrated that the sediment source fingerprinting technique can be utilized to investigate how the cumulative effects of anthropogenic and natural disturbances affect sediment source dynamics in a large river basin over short (i.e., annual) and longer (i.e., decadal) temporal scales.

Gellis et al. (2019) apportion sediment using elemental analysis and the sediment fingerprinting approach and age-date sediment with fallout radionuclides in the agricultural Walnut Creek Basin (Iowa, USA). In particular, the age-dating of sediment provides an important temporal context for understanding sediment source dynamics. In this study, the authors determine that erosion processes on agricultural cropland supply the majority of suspended sediment (62%) followed by streambank erosion processes (36%). Thereafter, the authors applied an age-date model with ^{210}Pb$_{xs}$ and ^{7}Be to illustrate that sediments typically reside in three different storage age boxes: a rapid box (< 1 year), a decadal box (10–100 years), and a geologic box (100–1000 years). This research highlights the potential of combining multi-fingerprint suites to simultaneously examine temporal and spatial erosion processes occurring in the critical zone.

Pawlowski and Karwan (2019) examine Pb and Be sorption dynamics to understand the potential limitations surrounding the use of these fingerprints. The authors use batch experiments with in-stream sediment deposits from two systems and varying solutions to replicate both background and elevated levels of iron oxide along with different dissolved organic carbon and sediment solution ratios. Pawlowski and Karwan (2019) found that the sorption of Pb and Be increased over time for all substrates and treatments. These authors demonstrated that sediment mineralogy, organic matter, and biogeochemical cycling processes may all affect the mobilization or retention of Pb and Be, potentially impacting their conservative behavior and thus their utility in sediment source fingerprinting research. Pawlowski and Karwan (2019) highlight that there may be a significant export of both ^{210}Pb$_{xs}$ and ^{7}Be in the solution phase along with other cations that may be exposed to the redox chemistry of a variety of oxides and hydroxides. These authors clearly illustrate how a variety of processes influence the development of fingerprint signals, and that it is important to strive to understand how these processes may affect sediment source fingerprints and source apportionment modeling.

Reiffarth et al. (2019) investigate the potential of compound-specific stable isotopes (CSSIs) to trace soils derived from different cultivated fields. In particular, the authors examine the spatial variability of carbon isotope ratios (δ^{13}C) from very-long-chain fatty acids at the point, transect, and field scales in an agricultural watershed in Manitoba, Canada. Reiffarth et al. (2019) found that very-long-chain fatty acids do have the potential to trace particulate matter derived from fields cultivated with different cropping species. Although this novel approach to targeting different species or fields could provide significant sediment source information, the authors demonstrate that more research is required into the weighting of subsamples of the source fingerprint, sample design (i.e., targeting flow paths and number of subsamples per field), tracer selection (i.e., which fatty acids to include in mixing models), and intra- and inter-annual tracer isotope variation (i.e., tillage effects and seasonality). The micro-targeting of individual fields with CSSIs and other targeted tracing techniques may help directly identify field and plot scale erosion processes that are disproportionately contributing sediment and sediment-bound contaminants to downstream river networks.

4 Perspectives for sediment source fingerprinting in the critical zone

Sediment source fingerprinting research directly or indirectly investigates multiple processes occurring in the critical zone. One sediment source fingerprinting technique uses different fingerprints to determine whether sediments are derived from surface soil erosion (e.g., agricultural topsoil) or subsoil erosion processes (e.g., channel banks, landslides, or gully erosion processes) (Olley et al. 2013; Ben Slimane et al. 2016; Jalowska et al. 2017). A second technique examines how erosion processes on different land uses, soil types, or geologies result in varying source contributions to sediment transiting river systems (Fox 2009; Le Gall et al. 2017; Tiecher et al. 2017). A third technique uses chronological fingerprints to investigate the temporal dynamics of erosion processes, providing information on whether sediment may have been eroded in the last year, the last several decades, or even potentially the last hundreds or thousands of years (Taylor et al. 2013; Matisoff 2014; Smith et al. 2014; Evrard et al. 2016). As material being transported through river networks reflect physical and biogeochemical processes occurring in the critical zone (Amundson et al. 2007), the sediment source fingerprinting technique is uniquely situated to investigate and provide further understanding regarding these critical and life-sustaining processes.

To further advance our understanding of the critical zone, we believe it is important for sediment source fingerprinting research to capitalize on combining multiple sediment source fingerprints to explicitly investigate Earth system science processes. In particular, the multi-fingerprint research in this special issue that simultaneously incorporates temporal fingerprints (e.g., ^{7}Be, ^{210}Pb$_{xs}$) with erosion process and spatial fingerprints (Gateuille et al. 2019; Gellis et al. 2019) outlines a potentially effective technique to investigate the response of critical zone processes to anthropogenic activities over multiple temporal scales. For example, short-term fingerprints such as ^{7}Be may provide information on erosion processes occurring on intra-annual temporal scales or even the individual rainfall event scale. Medium-term fingerprints, such as ^{210}Pb and ^{137}Cs provide erosion process information on decadal time scales (Douglas et al. 2009; Gartner et al. 2012). Long-term fingerprints, such as ^{14}C, ^{10}Be, and other properties (e.g., optically stimulated luminescence) may provide erosion process information over longer temporal scales (Fig. 4) (Wittmann et al. 2011; Belmont et al. 2014; Bartley et al. 2018).

Coupling temporal tracers with other fingerprints analyzed on sediment cores in riverine, estuarine, or lacustrine environments holds significant potential to provide additional information on processes occurring in the critical zone (e.g., Olley and Caitcheon 2000; Foster et al. 2007; Douglas et al. 2010). The analysis of fingerprints in lacustrine or riverine sediment cores and their comparison to source sample parameters allows for the investigation of particulate matter source dynamics through time; such as changes in geological sources (e.g., increased alluvial/sedimentary source contributions), soil type sources (e.g., Ustorthrent soils), and erosion processes (e.g., increased surface source contributions). Combining temporal tracers and other fingerprints may provide fundamental information on dynamics of multiple processes occurring in the critical zone before and after the great acceleration that has been recommended to mark the dawn of the Anthropocene.

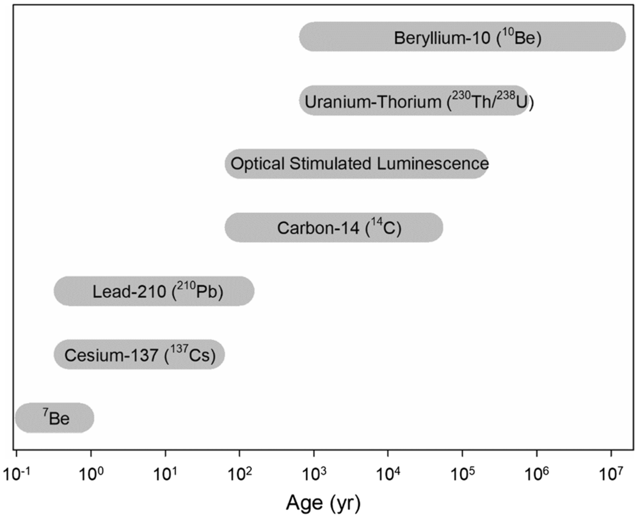

Fig. 4 Approximate dating ranges for a selection of chronological tracers

Combining spatial and temporal fingerprints also holds potential to increase our understanding of how the cumulative effects of anthropogenic and natural disturbances affect erosion process dynamics (Gateuille et al. 2019). In particular, it may be possible to investigate how natural disturbances (e.g., a major flooding event) or anthropogenic activities (e.g., agriculture, forestry, and mining) have affected erosion processes over the last century. Coupling sediment source fingerprinting and other watershed sediment modeling research (Mahoney et al. 2019) holds significant potential to improve our understanding of the cumulative effects of multiple disturbances on our increasingly degraded landscapes. Indeed, it will be beneficial to develop fingerprinting research designs that are fully integrated with watershed sediment modeling (Boudreault et al. 2019; Mahoney et al. 2019).

Advances with novel fingerprints such as CSSIs (Blake et al. 2012; Reiffarth et al. 2016) and environmental DNA (Evrard et al. 2019b) may provide direct information regarding changes in cropping or forestry species in a river basin. These next-generation fingerprints may help the sediment source fingerprinting technique move beyond focusing on estimating source contributions to developing a deeper understanding of how multiple processes in the critical zone have been affected by anthropogenic and natural disturbances during the great acceleration.

There are indeed other ways to capitalize on the sediment source fingerprinting framework to understand critical zone processes. For example, Mahoney et al. (2019) utilize the sediment source fingerprinting technique to investigate equilibrium sediment exchange processes. Other techniques may be able to investigate the sources and dynamics of nutrients (e.g., nitrogen and phosphorus) (Garzon-Garcia et al. 2017; Tiecher et al. 2019). Furthermore, sediment source fingerprinting may help investigate sediment connectivity (Koiter et al. 2013a; Chartin et al. 2017) and help validate watershed sediment models. Even striving to understand the behaviour of sediment source fingerprints and whether or not they are conservative (Pawlowski and Karwan 2019) and how pedogenetic and/or geologic processes drive source signal (i.e., fingerprint) development (Batista et al. 2019) will provide more information on a variety of different processes occurring in the critical zone. Indeed, more research is required to investigate the multitude of processes that establish the fingerprint source signals and drive their behavior during sediment generation, transportation, and deposition processes. Understanding fingerprint signal development and its behavior during these processes will go a long way to improving the reliability and robustness of sediment source fingerprinting research in addition to furthering our understanding of critical zone processes.

5 Conclusions

There has been a considerable advancement in the sediment source fingerprinting technique over the last several decades. In particular, the research focus has somewhat shifted away from understanding geomorphic processes towards highlighting the main sources (e.g., land use) contributing deleterious sediment loads in order to help guide management interventions. Along with a management focus, there has been a considerable drive in the literature to advance modeling techniques and reduce model uncertainty.

As the sediment source fingerprinting technique has advanced considerably, we believe it is time to return to one of the early foci of the technique: researching erosion and sediment delivery processes. As Uber et al. (2019) demonstrated, the model applied was not as important as the different fingerprints used in the model. As such, we believe it is time for sediment source fingerprinting research to move away from model-centric research programs to focus more on understanding the key processes driving the source contributions to sediment. Indeed, this may create a unique and yet a difficult balance for researchers to strive for. A balance that contributes to advancing some of the modeling nuances, sampling techniques, tracer selection approaches, and source apportionment strategies, while also including a direct objective to advance our understanding of dynamic processes in the critical zone.

Accordingly, we believe it would be beneficial for researchers to continue combining multiple sets of fingerprints (e.g., geochemistry and fallout radionuclides) together to help move sediment fingerprinting research forward. In particular, it will be important for researchers to use multiple sets of fingerprints in research projects and publications to capitalize on the power of simultaneously examining temporal, spatial, and process dynamics responsible for the relative source contributions of sediment transiting river networks. Furthermore, research with multiple sets of fingerprints may also help researchers investigate the cumulative effects of anthropogenic or natural disturbances with next-generation fingerprints. The more we research and understand the unique processes that establish the source fingerprint signals and influence the conservative behavior of fingerprints, the more we may begin to understand the key processes driving the mobilization, generation, and deposition of sediment, particulate matter, and even their bound contaminants, in the critical zone.

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