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Review

Heavy Minerals for Junior Woodchucks

Eduardo Garzanti and Sergio Andò *

Laboratory for Provenance Studies, Department of Earth and Environmental Sciences, Università di Milano-Bicocca, 20126 Milano, Italy; eduardo.garzanti@unimib.it

* Correspondence: sergio.ando@unimib.it; Tel.: +39-64482088

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Abstract: In the last two centuries, since the dawn of modern geology, heavy minerals have been used to investigate sediment provenance and for many other scientific or practical applications. Not always, however, with the correct approach. Difficulties are diverse, not just technical and related to the identification of tiny grains, but also procedural and conceptual. Even the definition of "heavy minerals" is elusive, and possibly impossible. Sampling is critical. In many environments (e.g., beaches), both absolute and relative heavy mineral abundances invariably increase or decrease locally to different degrees owing to hydraulic-sorting processes, so that samples close to "neutral composition" are hard to obtain. Several widely shared opinions are misleading. Choosing a narrow size-window for analysis leads to increased bias, not to increased accuracy or precision. Only point-counting provides real volume percentages, whereas grain-counting distorts results in favor of smaller minerals. This paper also briefly reviews the heavy mineral associations typically found in diverse plate-tectonic settings. A mineralogical assemblage, however, only reproduces the mineralogy of source rocks, which does not correlate univocally with the geodynamic setting in which those source rocks were formed and assembled. Moreover, it is affected by environmental bias, and by diagenetic bias on top in the case of ancient sandstones. One fruitful way to extract information on both provenance and sedimentological processes is to look for anomalies in mineralogical–textural relationships (e.g., denser minerals bigger than lower-density minerals; harder minerals better rounded than softer minerals; less durable minerals increasing with stratal age and stratigraphic depth). To minimize mistakes, it is necessary to invariably combine heavy mineral investigations with the petrographic analysis of bulk sand. Analysis of thin sections allows us to see also those source rocks that do not shed significant amounts of heavy minerals, such as limestone or granite, and helps us to assess heavy mineral concentration, the "outer" message carrying the key to decipher the "inner message" contained in the heavy mineral suite. The task becomes thorny indeed when dealing with samples with strong diagenetic overprint, which is, unfortunately, the case of most ancient sandstones. Diagenesis is the Moloch that devours all grains that are not chemically resistant, leaving a meager residue difficult or even impossible to interpret when diagenetic effects accumulate through multiple sedimentary cycles. We have conceived this friendly little handbook to help the student facing these problems, hoping that it may serve the purpose.

Keywords: relative and absolute abundances; sampling strategy; size-window for analysis; heavy mineral point-counting; provenance and plate-tectonic setting; chemical weathering; hydraulic sorting; recycling; diagenesis

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Well, according to the Junior Woodchuck's Guide Book, to get there we've have to take about 537 million steps straight up, till we reach the moon (Huey *in* Duck Tales).

1. Introduction

The aim of this script is to suggest a few "rules of thumb", challenge some widely held beliefs, pose some unsolvable problems, tell stories, and highlight some key aspects concerning heavy minerals, especially when they are used to infer provenance and reconstruct modern or ancient "source-to-sink" sediment-dispersal systems. Its content largely reflects the experience obtained by the writers mostly on modern sediments, with no ambition to reach any final truth. The reader can complete the panorama of heavy mineral research by consulting the monumental book by Mange and Wright [1] and the reviews by Morton [2] and von Eynatten and Dunkl [3] that focus on single-mineral techniques.

This paper deals first with only apparently trivial operational choices, and summarizes next the information gathered from modern sedimentary systems where everything is knowable in principle. Finally, it discusses the complex problems posed by ancient sedimentary systems, where little is known and most is lost by erosion from the sources and by chemical dissolution from the sink.

What are Heavy Minerals?

The simplest questions are often the hardest. A theoretically simple operational definition of heavy minerals includes all detrital components with density exceeding a given threshold, traditionally that of the once universally used bromoform (i.e., 2.89 g/cm^3). Because bromoform was discovered to be carcinogenic, other dense liquids came into use (generally polytungstates) and the standard density value is now generally rounded off to 2.90 g/cm^3.

In most continental or marine sediments, however, the 2.90 g/cm^3 fraction contains particles of diverse origin, including micas (e.g., biotite, chlorite), composite grains and rock fragments, chunks of ferruginous soils, altered grains of uncertain origin, iron oxides, titanium oxides, sulfides, sulfates (e.g., barite, celestite), ferriferous carbonates (e.g., ankerite, siderite), bioclasts with pyrite-filled chambers, glaucony, or phosphates (e.g., vertebrate teeth and bones). In some cases, even most of the dense fraction is formed by chemical or biochemical precipitation in the sedimentary basin rather than derived from erosion of source rocks. In ancient sandstones, dense grains may be of diagenetic or anchimetamorphic origin (e.g., anatase, brookite, barite, fluorite, epidote, iron oxides, sulfides, or even titanite and tourmaline) [4]. Anthropogenic grains such as moissanite or corundum may be present in modern sediments [5], and core samples and cuttings may contain barite or other heavy mineral contaminants associated with drilling muds [6]. Moreover, separation in the laboratory may not be perfect, and the recovered dense fraction occasionally includes tectosilicate grains or precipitated polytungstate crystals. Finally, among those that we should consider as heavy minerals proper, many are opaque to transmitted light and thus not readily identified under the microscope (e.g., Fe–Ti–Cr oxides), others are too altered to be safely identified or may belong to rare unknown species. These issues are rarely handled systematically or even exposed in scientific articles, with the result that researchers tend to use different and generally unexplicit criteria to decide what is included and what excluded from the heavy mineral string. Raman spectroscopy is a handy tool to solve many problems [7], but some remain elusive nonetheless.

2. Heavy Mineral Concentration

The definition of heavy minerals used in this article includes only minerals of certain extrabasinal terrigenous origin (i.e., ultimately eroded from bedrock exposed in source areas), denser than 2.90 g/cm^3, and occurring either as single detrital grains or in rock fragments. Grains of suspect intrabasinal (e.g., carbonates, bioclasts, glaucony), pedogenic or diagenetic (e.g., aggregates of iron or titanium oxides), and anthropogenic origin (e.g., barite in core samples) are thus neglected. Phyllosilicates are neglected as well. Transparent heavy minerals (tHM) identified under the microscope are considered separately from opaque and altered heavy minerals. Two indices are thus calculated, in either weight or volume, expressing the concentration of transparent heavy minerals (tHMC) and of total heavy minerals (HMC) relative to the bulk sediment. To avoid uncertainties involved in the identification of opaque grains and make datasets consistent and comparable, we choose here to refer to transparent heavy minerals only. Transparent-heavy mineral suites are considered conventionally as extremely poor (tHMC < 0.1), very poor (0.1 ≤ tHMC < 0.5), poor (0.5 ≤ tHMC < 1), moderately poor (1 ≤ tHMC < 2), moderately rich

($2 \leq$ tHMC < 5), rich ($5 \leq$ tHMC < 10), very-rich ($10 \leq$ tHMC < 20), or extremely rich (tHMC > 20). Heavy mineral-dominated sediments are called "placers" (HMC $\geq$ 50).

The Inner and Outer Messages

Heavy mineral studies take the relative percentages of heavy mineral species in the sample as the starting point for provenance interpretation. Relative percentages do contain the information we are looking for, but to read such an "inner" message correctly we need an "outer" message, a key that helps us to get it right [8] (p. 162ff). That key is heavy mineral concentration (Figure 1A). Otherwise, we would be tempted to call amphibole-rich a sample with 90% tHM amphibole and tHMC 0.1, and instead amphibole-poor another sample with 9% tHM amphibole but tHMC 10, which is wrong by a full order of magnitude. A particularly instructive case is presented by chromian spinel (Figure 1B), a mineral widely valued as a robust indicator of provenance from ultramafic rocks in ophiolitic complexes. Cr-spinel does occur in ultramafic rocks, where it is however rarer than olivine and pyroxenes by almost two orders of magnitude (Figure 3 in Reference [9]) [9]. Unless olivine and pyroxenes are massively destroyed by either serpentinization in the parent rock or by diagenesis during burial of the daughter sediment, Cr-spinel thus can represent only a low percentage of the very rich or extremely rich transparent-heavy mineral suite contained in ophioliticlastic sediments [10,11]. Conversely, because Cr-spinel resists diagenesis well [12], a high percentage of Cr-spinel in a very poor transparent-heavy mineral suite points to recycling of older sandstones ultimately derived in part from mafic or ultramafic rocks [5,13–15] (Figure 12 in Reference [14]; Figure 9 in Reference [15]).

Therefore, to avoid gross mistakes and misleading generalizations we need to estimate not only the relative abundance of heavy mineral species but also and always their absolute abundance (i.e., their concentration in the bulk sample). This is done simply by weighing the separated dense fraction and by making due corrections for spurious dense grains [9] (pp. 521–523). The concentration (and not just the spectrum) of heavy minerals in a sedimentary deposit depends on the composition of parent rocks, and increases by one order of magnitude or more during progressive unroofing of denser rocks found at deeper-seated crustal levels [16]. Drastic modifications of their concentration (as well as of their spectrum) may also occur by selective entrainment of low-density grains in the depositional environment [17], or by selective leaching of unstable species during diagenesis [18,19]. The concentration of heavy minerals in the sample (as well as their spectrum) is therefore *per se* crucial in provenance diagnoses and in the correct assessment of recycling and hydraulic or diagenetic processes.

The distortive fertility effect related to the different potential of different rock types to generate heavy minerals must always be taken into full account in the interpretation of heavy mineral suites, which tend to document aberrantly a limited number of sources (e.g., mafic igneous and medium/high-grade metamorphic rocks) whereas several others are barely recorded (limestone, chert, shale, granite). In the absence of significant chemical weathering and diagenetic dissolution, igneous and metamorphic rocks may impose their mark on the heavy mineral spectrum even where their outcrops are sparse (Figure 1A). As an extreme case, the heavy mineral assemblage of granite sand may be dominated by heavy minerals from medium/high-grade metamorphic country rocks contained as xenoliths within the granite body [16] (p. 541).

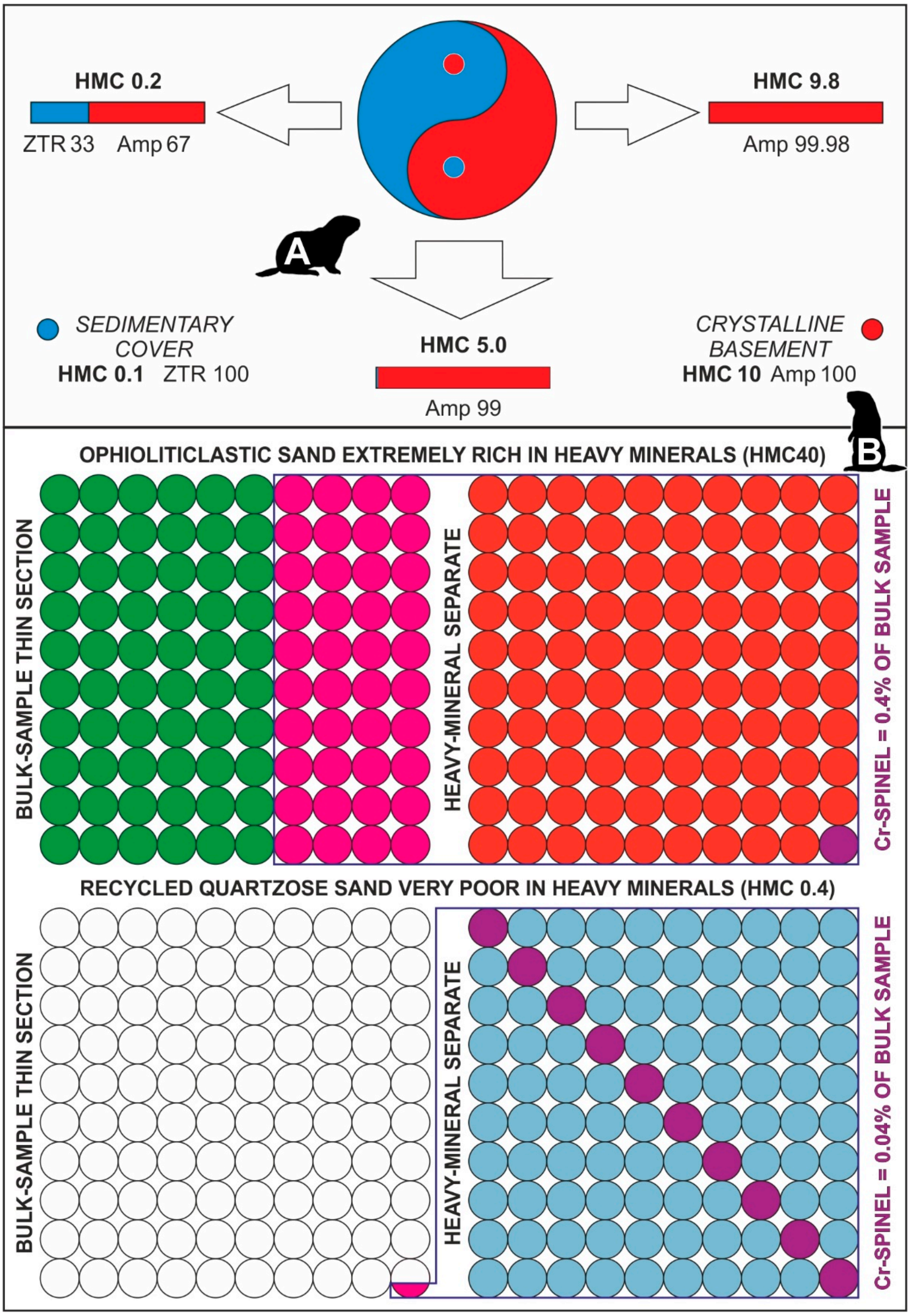

Figure 1. Heavy mineral concentration is the key to decipher provenance of heavy mineral suites. (**A**) The fertility effect (granitoid basement is supposed to supply 100% amphibole and cover strata 100% ZTR minerals; redrawn from Figure 1 in [9]). Amphibole shed from sparse basement outcrops dominates the heavy mineral assemblage in sand derived 98% from sedimentary covers (Amp 67% HM). Amphibole is ≥99% HM no matter whether crystalline basement represents 50% or 100% of total outcrops, although HMC is half in the former case. (**B**) Relative and absolute concentrations (stylistic representation

of the real case illustrated in Figure 9 in [15]). The sediment sample depicted schematically above is entirely derived first-cycle from an ophiolite complex: it contains 60% framework grains (plagioclase, mafic and ultramafic rock fragments; green circles) and 40% heavy minerals (magenta) including mostly pyroxene and olivine (red) and only 1% Cr-spinel (purple). The sample below is entirely recycled from older sandstones: it contains mostly quartz (white) with only 0.4% heavy minerals, all durable (greyish blue) and 10% of which is Cr-spinel. In the sample below, the absolute abundance of Cr-spinel is lower by one order of magnitude, but its relative abundance is higher by one order of magnitude.

3. Passing through Scylla and Charybdis

The results of heavy mineral analysis are strongly dependent on methodological choices, the very first of which is made in the field. Sampling and analytical biases are present in every heavy mineral study, although generally undeclared, uninvestigated, and consequently unquantified.

3.1. What are We Sampling?

What is apparently easier than collecting a sand sample in a dune field or along a sandy beach? Sand being everywhere, however, does not make things easier. Grain size may change from place to place, and colour may change from place to place indicating local enrichment or depletion in heavy minerals (Figure 2A). A commonly held prejudice is that, while sampling for heavy minerals, we should go for heavy minerals. Consequently, we may be attracted by beautifully coloured sand patches where heavy minerals are found in abundance (Figure 2B). Researchers may even choose to concentrate heavy minerals further by panning in the field, to reduce sample size and make the tedious job of separating them in the lab simpler and quicker. Although apparently reasonable and thus commonly adopted when aiming at zircon, apatite, or monazite for thermochronological or geochronological analysis, such an approach ends up to maximise bias [20]. Not only our artificially concentrated samples will be enriched anomalously in heavy minerals, hence leading to a gross overestimate of the natural heavy mineral concentration, but also all proportions among heavy minerals will be markedly altered. Denser and denser grains will be enriched more and more, hence leading to overestimate higher-density minerals such as zircon and to underestimate lower-density minerals such as amphibole.

The best sample is the one that most closely approximates "neutral" composition, i.e., the composition that sand would have everywhere in the absence of local hydraulic-sorting effects. Both coloured to black heavy mineral-enriched lags and lighter sand patches conversely enriched in slow-settling low-density tectosilicates or platy phyllosilicates should be carefully avoided. As grain size is concerned, it is advisable to collect samples as close as possible to the dominant grain size in the site for representativeness, and in the lower-fine to lower-medium sand range for practical reasons. Sediment finer than ~3ϕ is less suitable for petrographic analysis, and sand coarser than ~1.5ϕ is less suitable for heavy mineral analysis. Sample volume does not need to exceed 50–100 g and, provided the collected grain-size window is appropriate and separation procedures designed to maximise recovery, a few hundred grams are generally more than enough for the geochemical or geochronological study of any accessory mineral [21]. If automated-phase-mapping procedures by Scanning Electron Microscopy (SEM/EDS), Qemscan or Raman spectroscopy are used [22], then even a few grams may be enough for detrital-zircon geochronology. Single-mineral techniques can thus be routinely employed also on very small core samples.

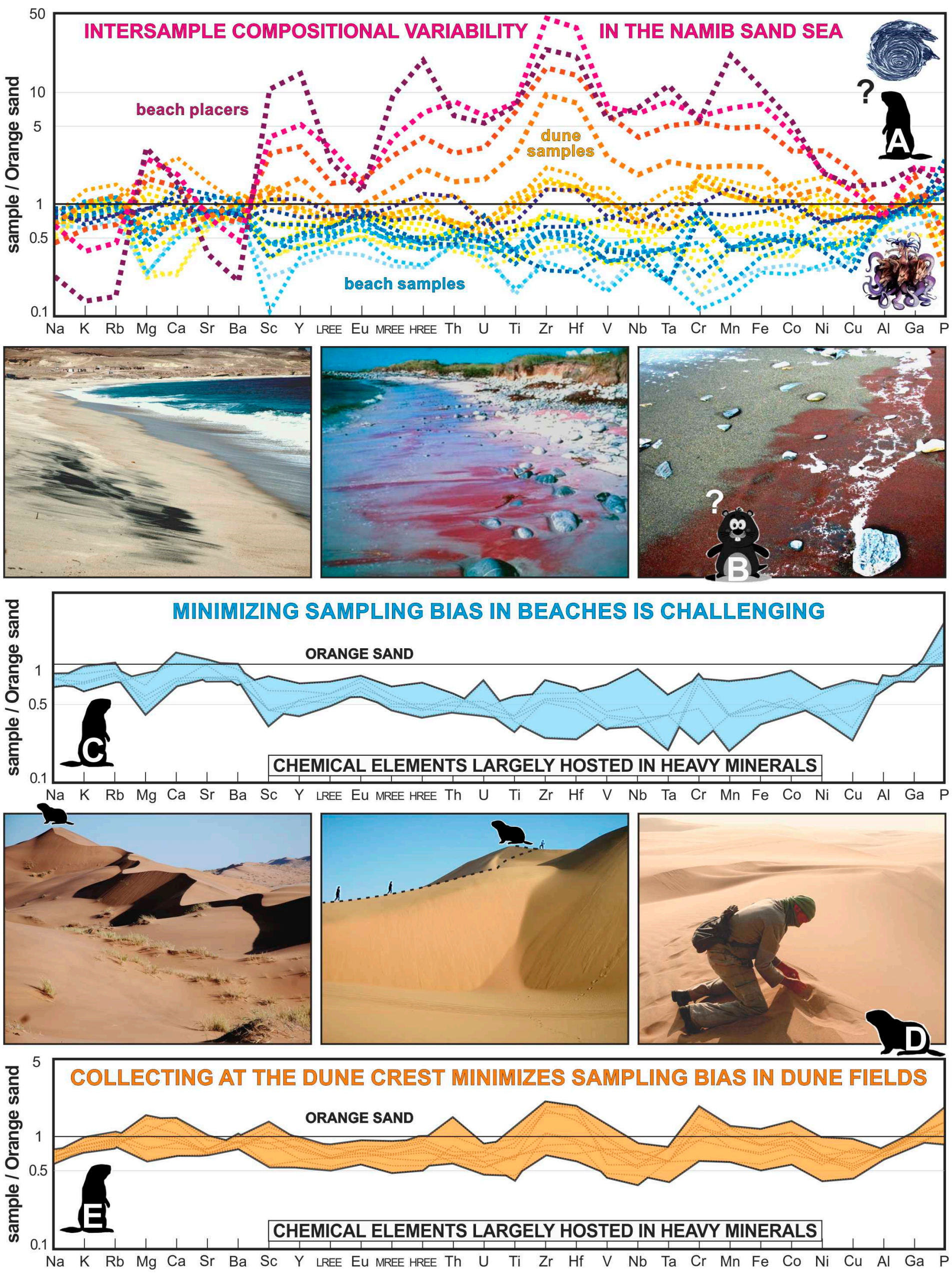

Figure 2. Sampling bias. Sampling, the most critical first step in any provenance study, should fulfill two basic requirements: (1) representativeness (sample composition as close as possible to "neutral" composition); (2) consistency (same sampling criterion applied for all samples). Geochemical data, particularly sensitive to anomalous concentrations of heavy minerals caused by selective entrainment at the local scale and made visible by marked colour differences, are most useful to check for sampling bias, as done for the Namib Sand Sea (**A**). In (**A**,**C**,**E**), elements are arranged following the periodic table group by group, and concentrations are normalized to average composition of Orange River sand. Colours: dunes in yellow-orange, beaches in blue, beach placers in purple. Data after [23]. For beaches (**B**), we could not identify a criterion that guarantees for both consistency and representativeness, and the geochemical test indicates that we ended up sampling heavy mineral depleted sites (Scylla's antiplacers) to avoid heavy mineral-enriched sites (Charybdis' placers) (**C**). For dune fields (**D**), the geochemical test indicates that collecting systematically at the crest of the highest dune is reproducible and reduces sampling bias (**E**). Unless we have specific aims (e.g., investigating grain-size dependent intersample variability or hydraulic-sorting effects), we routinely collect one sample in each locality.

Besides representativeness, the sampling plan must follow criteria that maximise consistency and comparability among samples. The criterion we currently use in dune fields is to collect sand systematically from the crest at the top of the largest dune in the area (Figure 2D). Geochemical analysis proves that such samples contain an amount of heavy minerals acceptably close to the "neutral" average heavy mineral concentration in the dune field (Figure 2E). On the contrary, beach sampling proved to be far less consistent. The geochemical test failed, indicating that in order to avoid being engulfed into the whirlpool of Charybdis (samples anomalously enriched in heavy minerals called "semiplacers"), we fell instead into the mouth of Scylla (samples anomalously depleted in heavy minerals called "antiplacers"; Figure 2C). The attention paid to design an appropriate sampling plan is never enough.

3.2. The Size-Window Problem

Contrary to what recommended by several authors, heavy mineral analysis of a narrow size class (e.g., 125–250 μm [24]; 63–125 μm [25]; 90–125 μm [26]) leads to biased results. The densest grains, markedly concentrated in the fine sediment tail [27,28], may be either notably overestimated or even completely cut off. The narrower is the class chosen for analysis, the larger the bias (Figure 3). A faithful characterization of detrital assemblages is obtained only if samples are analysed in bulk, which is feasible and fully advisable for well-sorted beach and eolian-dune sands.

In poorly sorted sediments, however, the presence of detrital grains with great size differences within a single concentrate makes mounting and identification difficult [29], and practical reasons force us to set lower and upper size limits for heavy mineral analysis in many cases. Heavy minerals in the finest silt fraction are difficult to identify with confidence under the petrographic microscope, whereas in coarse sand they are rare and commonly contained in rock fragments. For these reasons, we routinely consider a size window including 4ϕ (32–500 μm) or 5ϕ classes (15–500 μm) for heavy mineral analysis of moderately to poorly sorted sands. Not losing the finest tail of the size distribution is crucial, because the densest minerals such as zircon or monazite concentrate there. For a correct presentation of heavy mineral data, it is necessary to weigh the excluded finest and coarsest fractions, and to indicate what percentage of the total sediment the analysed size window represents and how much of the tails were cut off.

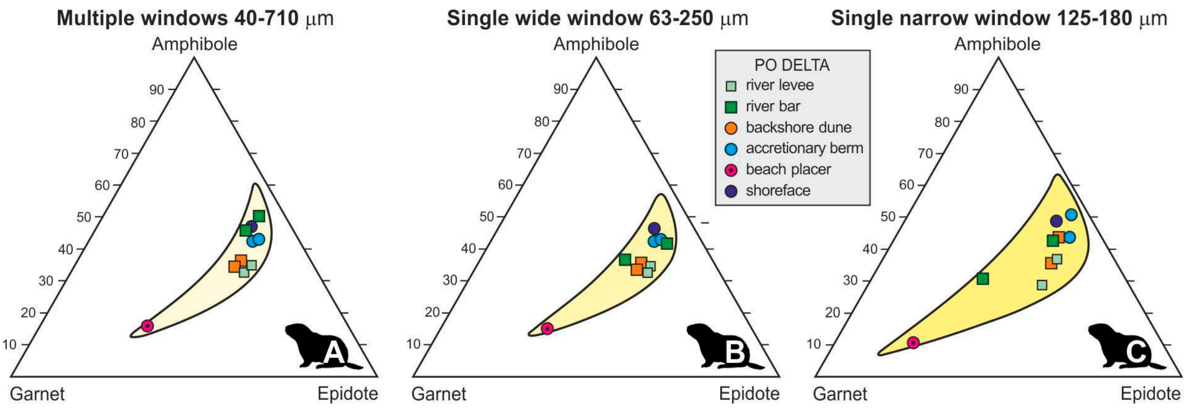

Figure 3. Analytical bias. Multiple-window analyses at 0.25ϕ or 0.5ϕ sieve intervals faithfully assess natural intersample mineralogical variability in Po Delta sand (**A**). Instead, single-window analyses introduce bias which decreases with the width of the analysed size class (**B**,**C**). All pale yellow fields are 90% predictive regions for data points. Data after [30].

3.3. Beyond Grain Counting

None of the three different methods generally used in grain counting (i.e., "ribbon/area", "line", and "Fleet"; [29,31] provides data which can be converted to correct volume or weight percentages [32]. Accurate determination of volume percentages by grain counting requires either the analysis of numerous closely-spaced sieve fractions for each sample, so that mineral grains on each slide can be considered to be of the same size [33] ("multiple-window" strategy of Garzanti et al. [34]), or measuring the size of all counted grains in an unsieved fraction [35]. Being these procedures very time-consuming, researchers seldom attempt to determine volume or weight percentages, and number percentages or number frequencies are used instead [36]. In such a way, however, abundances in volume or weight are systematically overestimated for higher-density heavy minerals and underestimated for lower-density heavy minerals, because in sorted sediments deposited by traction currents denser grains are smaller than settling-equivalent less dense grains. Errors may exceed 100% for high-density zircon, ilmenite, monazite and magnetite, and thus lead to significant imprecision in quantitative provenance analysis, comparison of mineralogical and geochemical data, calculation of sediment budgets, and assessment of mineral resources in placer deposits.

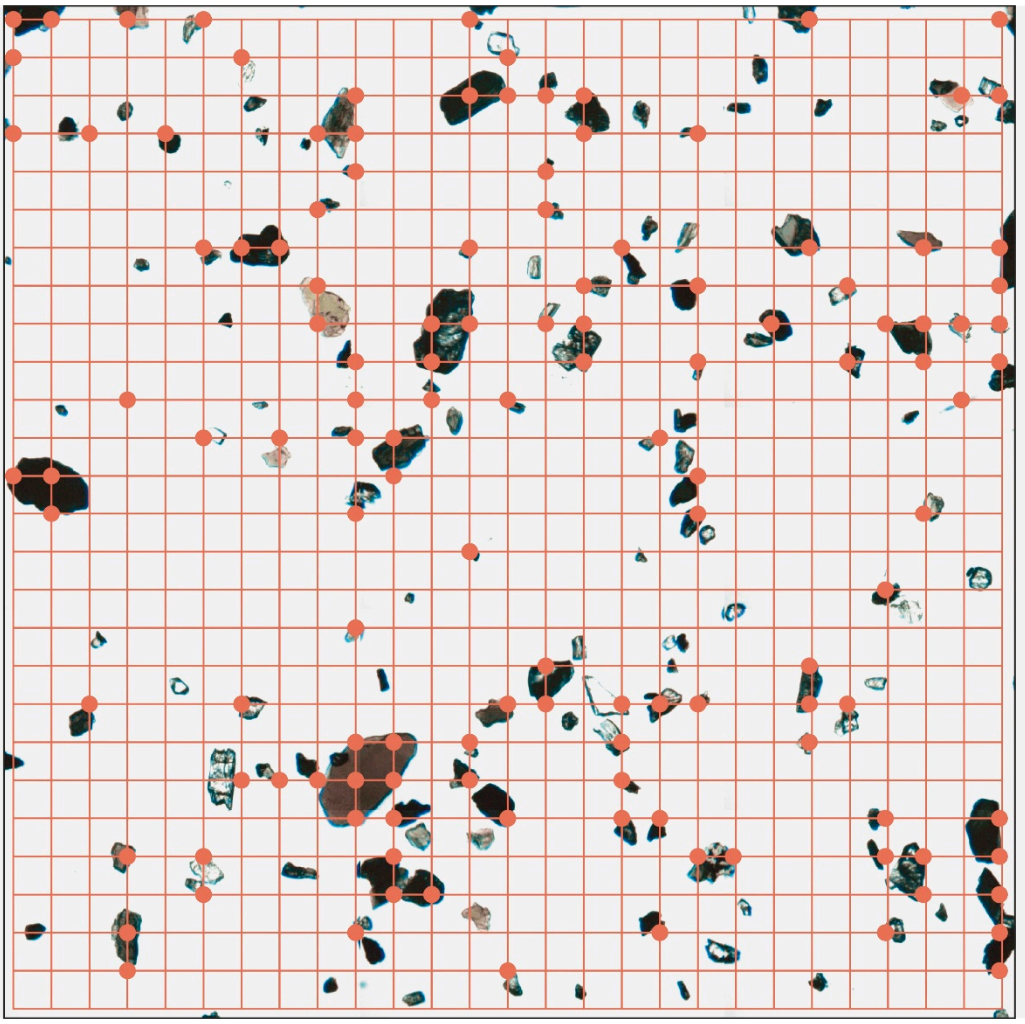

Figure 4. Point-counting allows obtaining real areal and therefore volumetric percentages of heavy minerals in grain mounts [32,37]. Choosing an appropriate grid is critical. In the case represented here—heavy mineral mount from the size window 15–500 m of a turbiditic silty sand from the Indus Fan (IODP1456A), square grid 125 µm—several grains are counted more than once, but with a larger spacing more than a single slide would be needed to count a representative number of transparent heavy minerals (usually $\geq$ 200).

In petrographic analysis, this problem was long solved by point-counting of impregnated sand or lithified sandstone in thin section following a grid of equally-spaced points along equally-spaced linear traverses (Figure 4; Glagolev-Chayes method of quantitative mineralogical analysis) [32]. Because the probability of a grain being hit by the cross-hair along this grid is proportional to its visible area, the areal percentages of various detrital components are obtained, which are equivalent to volume percentages [37]. Surprisingly, the Glagolev-Chayes method was hardly ever used in heavy mineral analysis. Nonetheless, it can be done, it is not significantly more time-consuming than grain-counting, and should be used routinely. As for petrographic analysis of thin sections, grid spacing needs to be large enough to ensure that grains are not counted twice. Heavy mineral mounts must be properly prepared to avoid that grains are either too numerous, otherwise they overlap and cannot be identified properly, or too sparse, otherwise a large number of voids will be counted before a sufficient number of heavy minerals is encountered and identified.

4. Heavy Minerals as Provenance Tracers

The investigation of multi-mineral suites provides crucial provenance information for paleotectonic reconstructions [38], especially if coupled with petrographic observations under the microscope. Four decades have passed since Dickinson and Suczek [39] have attempted to establish a link between sediment composition and plate-tectonic setting. Detrital modes, however, can only reflect—in many cases unfaithfully—the mineralogy of source rocks, which is not necessarily a good proxy for geodynamic environment [40,41]. Volcanic and plutonic rocks with very similar mineralogy are found in magmatic arcs, orogenic belts, rift shoulders, and continental interiors. The relationship between detrital modes and geodynamic setting is even looser for heavy mineral suites for a range of reasons, first of all because of the fertility bias: some sources lack heavy minerals (carbonates, chert), whereas others are extremely prolific (mafic and ultramafic igneous rocks). Hydraulic effects may alter both absolute and relative abundances of heavy minerals by an order of magnitude or more. In ancient sandstones, most heavy minerals may be leached out by diagenetic dissolution. And how can we assess the extent of sedimentary recycling? These are the main reasons why heavy mineral and petrographic analyses should always be coupled. Petrographic investigations prevent us from losing precious information conveyed especially by rock fragments and enable us to assess heavy mineral concentration more accurately. Only with this knowledge provenance interpretations can be based on firm ground. A brief summary of typical transparent-heavy mineral assemblages found in different anorogenic and orogenic geodynamic settings is provided in the following paragraphs as an update of Garzanti and Andò [42]. The expected unroofing trends produced by the progressive erosion of deeper-seated rocks are also envisaged.

4.1. Anorogenic Provenances

Geodynamic settings unrelated to oceanic or continental subduction include continental rifts, passive margins, and continental interiors. Three main sources of detritus may be distinguished in these settings: (a) siliciclastic to carbonate sedimentary covers; (b) generally polymetamorphic old basement rocks; (c) intraplate volcanic rocks including continental flood basalts. Three are the corresponding provenances: (A) undissected continental block and/or recycled clastic; (B) dissected continental block; (C) anorogenic volcanic [40].

Siliciclastic cover strata are commonly distinguished by notably poor heavy mineral suites containing zircon, tourmaline, and rutile (ZTR). Carbonate rocks and chert do not supply heavy minerals. Crystalline basement including granitoids and gneisses typically sheds rich assemblages including amphibole and epidote derived from mafic and intermediate rocks of respectively medium and low metamorphic grade, with subordinate metamorphic minerals including garnet and generally minor staurolite, kyanite, andalusite, or sillimanite. Anorogenic volcanic sources are revealed by very rich transparent-heavy mineral suites dominated by clinopyroxene, locally associated with olivine, apatite, zircon, pigeonite, Cr-spinel, or hypersthene. The expected unroofing trend in non-volcanic settings will see a progressive increase in heavy mineral concentration, with a progressive increase in amphibole relative to ZTR minerals as wider areas of basement rocks are erosionally exhumed through time. In volcanic settings, a decrease in pyroxene will be compensated by an increase in amphibole and metamorphic minerals derived from the underlying crystalline rocks [43–45].

4.2. Magmatic Arcs

Magmatic arcs formed above an oceanic subduction zone include two main sources of detritus: (a) volcanic rocks of the "calc-alkaline" series; (b) granitoid batholiths including granodiorite and tonalite. The two corresponding provenances are: (A) undissected magmatic arc; (B) dissected magmatic arc [46–48].

Volcaniclastic heavy mineral assemblages are commonly rich to extremely rich and dominated by clinopyroxene. Arc basalts also commonly shed olivine, whereas hypersthene and kaersutitic hornblende or oxy-hornblende occur in andesites and more felsic rocks. The orthopyroxene/clinopyroxene ratio may thus be used as an indicator of increasing silica contents [49]. Pigeonite may occur in tholeiitic andesites lacking olivine, and clinoenstatite in boninites. Relatively Ti-poor titanomagnetite is the dominant opaque mineral. Garnet (e.g., melanite) and Cr-spinel are rare. Such arc-related suites are unfortunately not readily discriminated from those derived from anorogenic lavas.

Granitoid batholiths supply rich, hornblende-dominated suites. Epidote and actinolite shed from metavolcanic complexes, kaersutitic hornblende and hypersthene shed from gabbro-norites, as well as titanite, allanite, zircon, and monazite may occur. The predicted unroofing trend would see a progressive increase of the hornblende/pyroxene ratio as granitoid batholiths are progressively exhumed through time.

4.3. Axial Belts and Obducted Ophiolites

Orogenic belts consist of stacked and juxtaposed tectonic domains that may include any kind of rock assemblages and thus cover all previously described provenances. Two additional domains typical of orogenic settings can, however, be identified: (a) fossil continental subduction zones, characterized by high-pressure neometamorphic rocks; (b) obducted ophiolitic complexes, characterized by mafic and ultramafic rocks. The two corresponding provenances are: (A) axial belt; (B) ophiolite [50].

The neometamorphic axial backbone of orogenic belts associated with continental subduction consists of exhumed high-pressure to ultra-high-pressure crustal and mantle rocks shedding rich to extremely rich transparent-heavy mineral suites dominated by garnet, amphibole or epidote depending on protoliths (continental vs. oceanic), depth reached during subduction, and pressure/temperature paths followed during exhumation [51]. Diverse metamorphic minerals, including glaucophane, chloritoid, staurolite, kyanite, sillimanite, and diopsidic clinopyroxene may also occur. Ultramafic mantle rocks formerly belonging to the lower plate and transformed into antigorite schists during subduction at eclogitic depths shed mainly magnetite clusters formed during release of iron from the olivine lattice during serpentinization.

Tectonic deformation is much less extensive in obducted ophiolites belonging to the upper plate, which supply rich to extremely rich suites typically dominated by enstatite derived from mantle harzburgites and including olivine from mantle rocks and cumulates, diopside and diallage from gabbros, hypersthene from gabbro-norites, hornblende from high-level gabbros, epidote and actinolite from the sheeted-dyke complex, and clinopyroxene from pillow basalts. Cr-spinel is derived from ultramafic rocks, zircon from plagiogranites, orthopyroxene and clinoenstatite from boninites [10,11].

4.4. Mixed Orogenic Provenances

Orogenic provenance is a complex issue, difficult to summarize in brief. At least five radically different types of orogenic belts can be identified on our rotating planet, depending on subduction polarity and on the continental or oceanic nature of the lower and upper plates involved in orogeny. Himalayan-type collision orogens are generated by continent-beneath-continent eastward subduction, Andean-type cordilleras by oceanic-beneath-continent eastward subduction, Oman-type obduction orogens by continent-beneath-ocean eastward subduction, Burman-Andaman and Barbados-type subduction complexes by ocean-beneath-ocean subduction, and finally Apennine-type orogens by retreating westward subduction [50,52]. Each of these archetypal orogenic belts consists of peculiar rock assemblages, and thus sheds different quantities and types of heavy minerals. Alpine-Himalayan collision orogens are characterized by neometamorphic axial-belt detrital signatures of trunk-river sands [17,51,53], Andean cordilleras by magmatic-arc signatures on the pro-side and by largely recycled detrital suites on the retro-side [54], Oman-type ophiolitic allochthons by enstatite and other pyroxenes, emerged subduction complexes and Apennine-type belts by largely recycled heavy mineral suites [5,11,15,55,56].

5. Environmental Bias: Turning Problems into Opportunities

Provenance signals may become progressively distorted by physical and chemical processes during transfer from source to sink along the sediment-routing system, producing what we are used to considering as noise that limits our capacity to understand. Particles with different size, density, and shape are segregated in different sediment fractions according to physical laws in all three stages of the sedimentary cycle: erosion, transport, and deposition. Most environmental "noise" is therefore coherent, and closer inspection allows us to discover that it can be converted profitably into both environmental and clearer provenance information.

5.1. Heavy Minerals as Tracers of Hydraulic Processes

There are two distinct ways to consider mineralogical variability, either among the different size classes of the same sediment sample (intrasample variability) or among different samples (intersample variability). Each is controlled by a different hydraulic process of size-density sorting: intrasample variability is fundamentally a consequence of settling equivalence (Figure 5A), intersample variability of selective entrainment (Figure 5B).

The settling-equivalence principle states that grains found together in a sediment layer deposited by a traction current under a single set of hydraulic conditions must have the same settling velocity [27]. A temporary deposit, such as a fluvial bar or a beach, therefore, consists of coarser low-density grains such as quartz or feldspar associated with a range of heavy mineral species the size of which decreases progressively as their density increases. Low-density tectosilicates will thus protrude above the sediment layer higher than heavy minerals. When the deposit is impacted by a high-energy current, during a storm or a flood, the larger grains that have smaller pivoting angles and experience greater flow velocities and drag forces will be selectively removed (Figure 5B) [57]. As a consequence of such a selective-entrainment process, lag deposits are progressively enriched in heavy minerals in proportion to their density, until placers are formed [58–60].

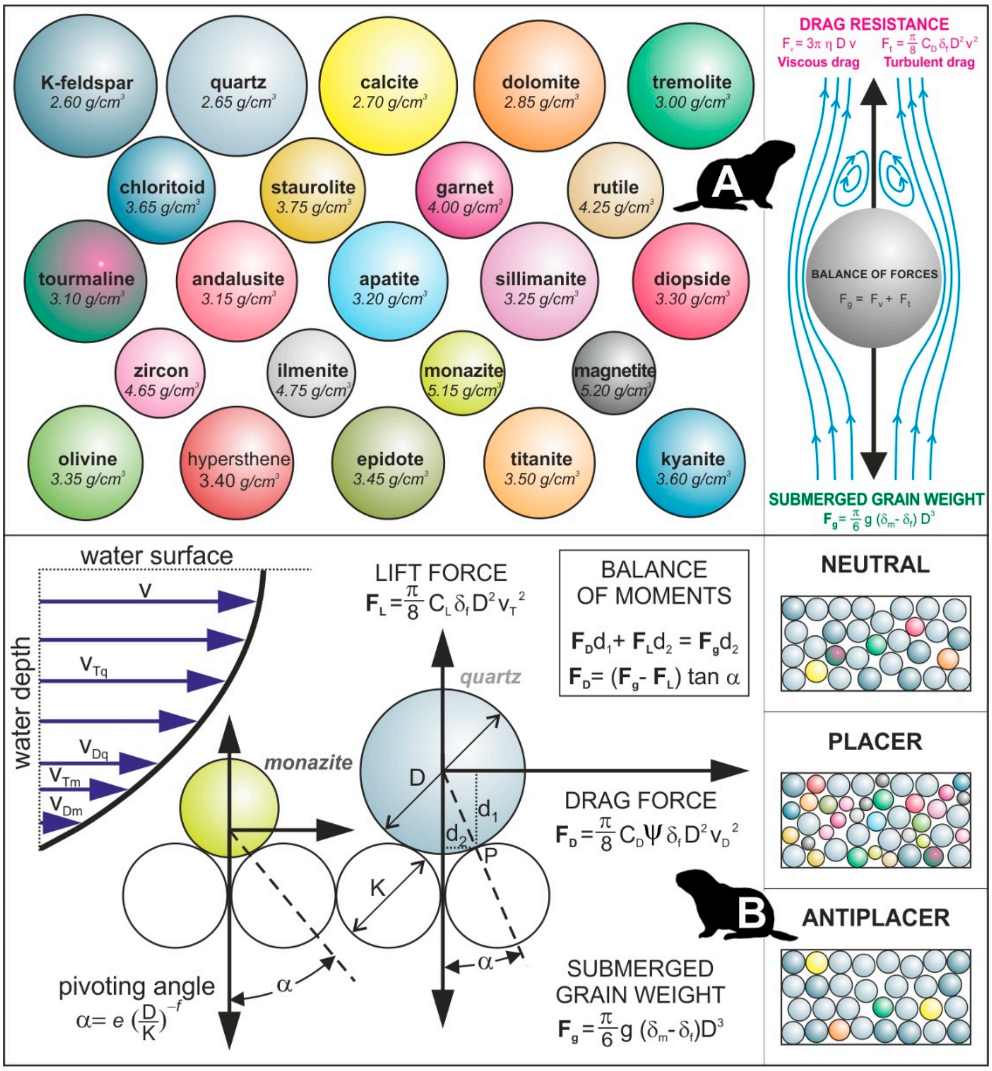

Figure 5. Hydraulic sorting. Grains are sorted by their size, density, and shape during erosion, transport, and deposition by traction currents. Complex shape effects are not taken into account here. (**A**) The settling-equivalence principle [27] controls size-density relationships among deposited grains and is responsible for intrasample compositional variability [34]. Spherical minerals in the picture have all the same settling velocity of 2.67 cm/s (calculated for a quartz sphere 250 μm in diameter according to the empirical formula of Cheng [61]). (**B**) Selective entrainment of coarser low-density grains while smaller settling-equivalent heavy minerals are left behind [62] is responsible for intersample compositional variability [30]. Sediments of theoretical "neutral" composition are thus partitioned into "placer" and "antiplacer" deposits readily distinguished by colour contrast even at the very local scale (Figure 2B).

5.2. Extracting Environmental and Provenance Information

Understanding how detrital minerals of different size, density, and shape behave under the action of traction currents allows us to add details to sedimentological interpretations and to choose correctly among provenance options. By means of settling-equivalence analysis, we can reconstruct hydraulic conditions at the instant of deposition, and calculate the settling velocity of specific laminae and laminasets. We can discriminate between sorting processes occurring in water or air, and thus distinguish between eolian backshore dunes and shoreface bars in deltaic settings (Figure 6 in Reference [34]) [34].

Violations of the settling-equivalence principle offer additional clues to provenance diagnoses. If one mineral or group of minerals shows anomalous textural relationships with another mineral or mineral group, then we may suspect mixing of detrital populations derived from different sources or transported in different modes or by different media (e.g., wind versus ephemeral streams in deserts, wind versus waves in deltas, local rivers versus longshore drift along high-energy coasts (Figure 8 in Reference [34]; Figures 22 and 26 in Reference [63]; Figure 6 in Reference [64]; Figures 9 and 10 in Reference [65]) [34,63–65].

Anomalous roundness relationships may convey useful information on both provenance and depositional environment. As a first approximation, we may consider the roundability of heavy minerals as inversely related to their hardness measured by the empirical Mohs' scale [66] (p. 299), [67], [68] (p. 13). Grain roundness has long been held to be suggestive of eolian sand transport [69,70], because rounding is far more effectively achieved by strong grain-to-grain impacts in air than during transport under water [23,71,72]. Association of heavy mineral species with similar hardness but markedly different roundness is another clue revealing mixing of detrital populations from different sources (e.g., angular local-river sand and rounded longshore-drifting grains) (Figure 10 in Reference [65]) [65].

6. Illusions, Shortcuts, and Logical Traps

Geological problems are intricate. Nature's products typically result from the interplay of several controlling factors, and we seldom have adequate tools to detangle the relative contribution of each. Laboratory experiments can hardly reproduce natural complexities at the prohibitively huge spatial and temporal scales of geological processes. Our equations thus generally remain with far too many unknowns and poorly constrained knowns to solve. In face of difficulty and with the desire to hit ground, it may be tempting to recur to instinct, which often merely amounts to prejudice, and fix ourselves on one preferred plausible solution. But plausibility, and worse fashion, seldom paves the way of scientific progress [73].

For instance, considering grain-size classes as transport-invariant sub-populations [74] may lead to the widely shared but wrong assumption that narrowing the size-window in heavy mineral analysis leads to increased consistency, whereas in fact, it maximizes bias (Figure 3). As seen above, high-density minerals settle at the same velocity of, and hence are deposited together with, coarser low-density or platy minerals [27]. Therefore, the different grain-size classes of sorted sediment deposited by a traction current have invariably a notably different heavy mineral suite, and bulk-sample point-counting or multiple-window grain-counting or point-counting represent the only options to estimate the volume percentages of detrital minerals in the sample correctly [30].

6.1. How Could Zircon Be Enough?

Owing to its great durability, detrital zircon is preserved widely in polycyclic sands and in ancient sandstones, where it commonly represents one of the few minerals that survived chemical dissolution during diagenesis. Moreover, zircon grains can be dated robustly and routinely at a reasonable cost. These unique features have made zircon the preferred target of provenance research in the last decade. Zircon-age spectra, especially if coupled with Hf isotopic fingerprinting, represent indeed a powerful tool to identify sources characterized by specific age-windows of magmatism and crustal growth. Using detrital-zircon data alone to extract provenance information, and even worse to calculate sediment budgets, is however a risky business [75,76].

First, it requires handling the thorny fertility issue [77–79], which is hard to do precisely and robustly [80]. Zircon-free sources, including mafic and ultramafic rocks, limestone and chert, will remain unseen, and worse, not looked for [81] (p. 85). Second, durable zircon is likely to undergo recycling even several times [82,83]. The information it carries, therefore, may not relate to igneous or metamorphic rocks exposed during the sedimentary cycle in question, but to the igneous or metamorphic sources of the parent sandstones, or even to the igneous or metamorphic sources of the sandstones that sourced the parent sandstones, and so on in a recursive iteration that may climb backwards an unknown number of steps.

Last but not least, the opportunities offered by zircon grains should not lead us to disregard the wide spectrum of other detrital minerals. The expected average zircon content in a siliciclastic sediment is about 1 grain out of 5000, considering that the average zirconium concentration in the upper continental crust is estimated as 190–193 ppm [84,85], that zircon in sediments does not occur exclusively as sand-sized grains but also commonly as tiny inclusions within micas and other minerals [22], and that not all zirconium is hosted in zircon. If the fascination exerted by zircon grains leads us to forget everything else, then we are bound to miss all of the information potentially offered by the other 99.98% of framework grains in the sample (Figure 6).

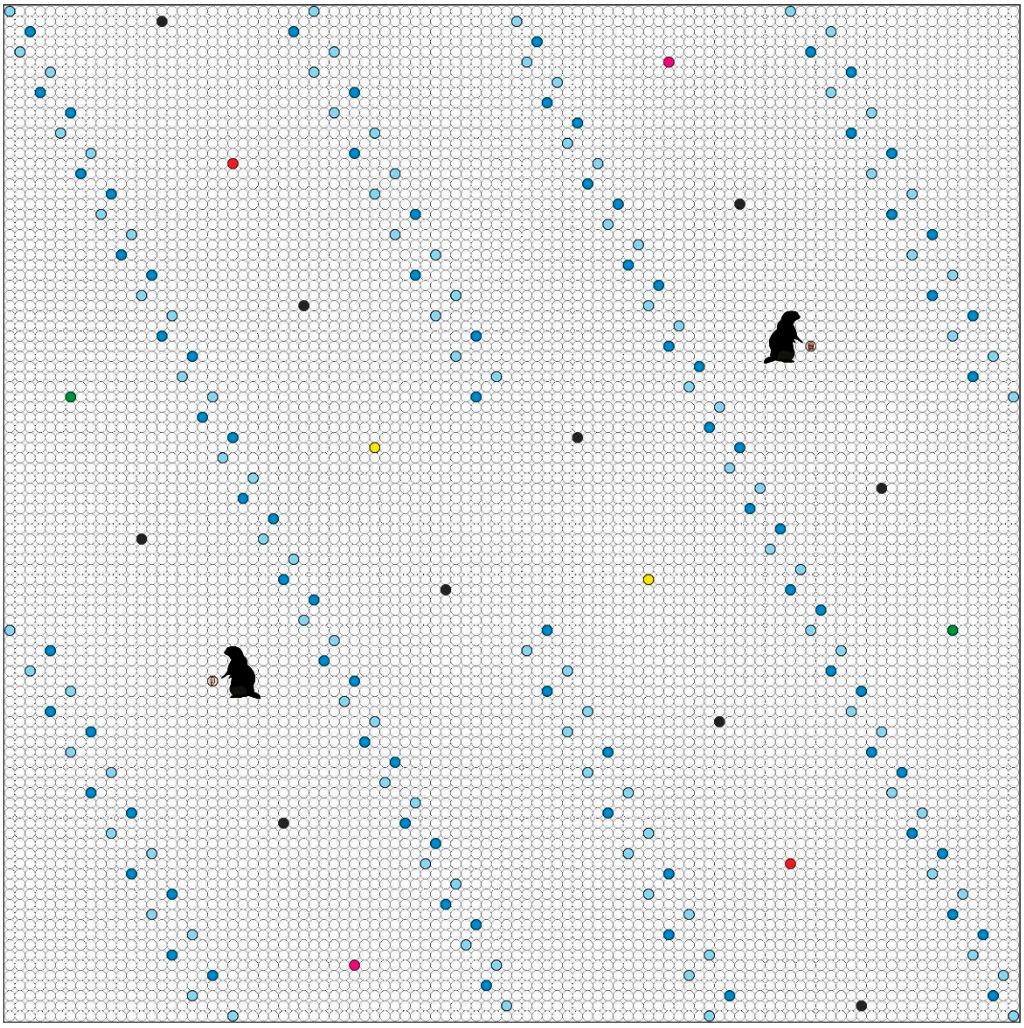

Figure 6. Wonder zircon is hardly enough. This schematic picture represents the quartzose composition typical of an ancient quartzarenite or recycled sand as that carried by the Congo River. Quartz is 98% (white), K-feldspar 1% (light blue) and plagioclase 0.8% (darker blue); opaque (black) and transparent heavy minerals (colourful) are 0.1% each. In such cases, zircon is undoubtedly the most precious vehicle of provenance information, although it represents only 2 grains (both spotted by brave woodchucks) out of 10,000.

6.2. Jumping to Conclusions: the Plausibility Trap

To handle complexity, we have to make assumptions about those controlling factors that we believe to be commanding. This is legitimate, provided that our starting assumptions are not rapidly forgotten, thus increasing the risk of moving in a circle to make discoveries that are merely tautological.

The development of sequence stratigraphy [86,87] provides an archetypal example illustrating the pitfalls related to such a faulty conceptual procedure. Although we are well aware that stratigraphic architecture is the product of complexly interacting local autocyclical and regional to global allocyclical factors including tectonics, climate, and sediment supply [88], at a certain point most of the scientific community found advantageous to believe that the stratigraphic record could be almost mechanically interpreted as the univocal response to eustatic change [89]. Eustasy, however, has no plausible cause at the required amplitude and frequency through geological time [90,91], and thus ended up being used as a convenient ruling factor reinforced by circular reasoning rather than by observational evidence [92,93]. A similar illusory way to cut the Gordian knot is to assume that "*detrital modes of sandstone suites primarily reflect the different tectonic settings of provenance terranes*" [46] (p. 333), although they rather reflect the mineralogy of source rocks, which is not univocally related to a geodynamic setting, and are variously affected by environmental and diagenetic bias.

Because the controlling processes and modifying factors are far too many and too poorly constrained to be evaluated with fair accuracy, paleogeographic reconstructions based on detrital modes of ancient sandstones are inevitably as simplistic as "spherical cows" [94]. If on the one hand, our cartoons appear too often as hopeless exercises when observed with a critical mind, on the other hand exposing ourselves to failure may represent the only path to progress in the long run [95]. Rather, the game is really lost when we choose to defend a comfortable theory by dismissing observations. Examples include the widely held belief that mafic detrital minerals such as olivine and pyroxene, or even amphibole and feldspar, can be rapidly eliminated during transport, a fact long demonstrated untrue by studies of natural river systems and laboratory experiments [23,63,73,96,97]. Another idea that "*owes its popularity to plausible reasoning rather than to observational evidence*" is that sand-sized detrital minerals get effectively rounded during fluvial transport, thus enabling us to guess whether the source was near or far [73] (p. 1348).

6.3. The Maturity Misconcept

When confronted with natural phenomena that we do not understand, we humans are prone to believe in mythical narrative. Myths are nice to hear, credible, fearsome, and appeasing. Geological literature is full of mythical scenarios plunged in deep time. Some are skilfully reconstructed (e.g., the dinosaur-inhabited Earth) [98], but others are simply born in our mind rather than inferred by induction. An example is the maturity concept, extrapolated from biology to numerous other disciplines including psychology, sociology and economics, which conveys a positive feeling of natural progress and betterment. Under such a teleologic spell [99], sediments are expected to become cleaner and purer with time and destined to reach a final stage of perfection in both texture (well sorted and rounded "supermature" sandstone) [100] and mineralogy (quartzarenite with "ultrastable" heavy mineral suite) [101]. Quartzose sand containing only zircon, tourmaline and rutile may indeed be the result of polycyclicity [102], but recycling may even produce lithic sands poorer in quartz and less "mature" than their parent sedimentary rocks [13,103,104]. Pure quartzose sand was apparently more common on Earth during the Proterozoic and Lower Paleozoic [70,105] than today [106].

7. The Chemical Moloch

Chemical processes are far more efficient than mechanical processes in removing unstable grains such as ferromagnesian silicates. Dissolution occurs while fluids circulate through soil profiles and more extensively when acting through geological time at the progressively higher temperatures reached during burial diagenesis.

7.1. Pre-Depositional and Post-Depositional Dissolution

Minerals found in igneous and metamorphic rocks grew at temperature and pressure conditions very different from those existing at the surface of the Earth. In sediments, therefore, they find themselves "out of place" by different degrees, as empirically indicated by the Bowen series:

olivine > pyroxene > amphibole, biotite > muscovite, plagioclase > K-feldspar > quartz [107]. Chemical reactions, however, proceed slowly in the lack of circulating fluids and/or at low temperatures, and dissolution rates tend therefore to be lower in arid and cold climates [108,109]. In humid and warm climates, instead, prolonged feldspar hydrolysis can transform granitoid and gneissic source rocks into residual sand consisting almost entirely of quartz [110]. Tourmaline [111], zircon [112,113], and even quartz may be extensively leached [114–116], although generally slower than other minerals. No detrital species is chemically stable in all conditions.

Two important aspects are emphasized in van Loon and Mange [111] (p. 189). First: "*the effects of extreme chemical weathering on a heavy mineral assemblage differ fundamentally from those of burial diagenesis*". Apatite and garnet are widely considered among the most durable species, which is generally true only during diagenesis [12]. Apatite can be highly unstable even in mildly acidic conditions [26], and garnet is rapidly leached out in lateritic soils of equatorial regions, where it may prove to be far more unstable than amphibole (Figure 9 in Reference [44]) [44], thus reversing the stability sequence observed in most ancient siliciclastic successions.

Second: "*the joint occurrence of fresh and strongly weathered grains (with the same chemical composition) of one heavy mineral species indicates that the degree of chemical weathering is a statistical rather than a fixed parameter*". In ancient sandstones, deeply etched or skeletal grains of pyroxene, amphibole, or staurolite may coexist with surviving grains of the same mineral that are only weakly corroded. This "statistical" aspect must be kept in mind while trying to assess the degree of pre-depositional or post-depositional chemical dissolution from surface textures of heavy minerals [19].

7.2. Diagenetic Bias: What You See is not All There Was

The conviction that what is observed is all that matters is typical of naive "fast" thinking [81]. This easygoing attitude proves to be often misleading in ordinary life as in psychological or economic issues, and the potential pitfalls are not less insidious in geological reconstructions. For instance, although we are aware that most was lost from the sedimentary record, we prefer to believe "*that the stratigraphical column in any one place is a long record of sedimentation with occasional gaps*", rather than "*a long gap with only very occasional sedimentation ... a lot of holes tied together with sediment*" [88] (p. 35). Since Steensen [117], paleontologists have long realised that most fossils represent a part of the hard skeleton and only a fraction of the living body, but it is much harder to realize that the very same is true in heavy mineral research. We know that most detrital minerals do not resist prolonged diagenesis in most cases (Figure 7), and yet, because we can work only with what we have and cannot retrieve what has been lost, the "*what you see is all there is*" pitfall [81] is difficult to avoid.

Many sedimentary geologists, asked about the most common heavy minerals, will name zircon, tourmaline, and rutile. Probably a minority only will mention amphibole, epidote, garnet, or pyroxene, which are by far the most abundant species in igneous and metamorphic rocks and consequently in detritus derived from them. Researchers working with ancient sandstones indeed find zircon, tourmaline, and rutile far more commonly than other minerals in their heavy mineral mounts, but this is just because zircon, tourmaline, and rutile are the durable ones that stand the best chance to survive chemical attack through multiple sedimentary cycles.

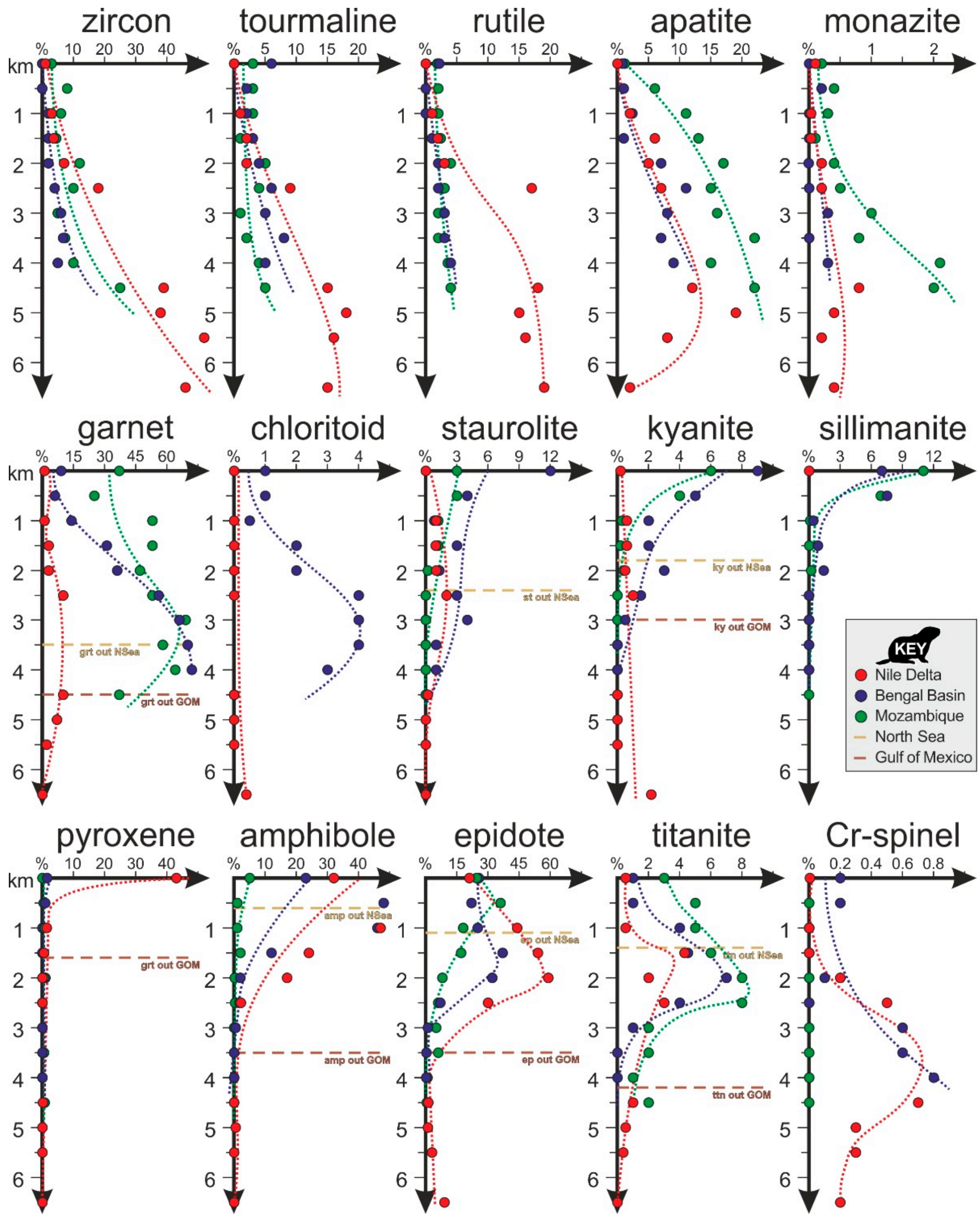

Figure 7. Diagenetic bias. Comparison between the burial-depth distribution of transparent heavy minerals in subsurface sedimentary successions of the Gulf of Mexico, North Sea, Bay of Bengal, Nile Delta, and coastal Mozambique. Data after Figure 2 in [4], Figure 2 in [12], [19], and [118,119]. Heavy minerals are progressively dissolved during burial diagenesis and finally leached out at depths varying notably from basin to basin depending on original mineral abundance, pore-fluid composition, and geothermal gradient (18–26.5 °C/Km for the Nile Delta, 20–30 °C/Km for the Gulf of Mexico, 30–40 °C/Km for the North Sea) [120–122]. The indicative order of relative mineral durability would be zircon ≥ rutile ≥ tourmaline ≥ Cr-spinel ≥ apatite ≥ monazite > chloritoid ≥ garnet > staurolite > kyanite ≥ titanite > epidote > sillimanite > amphibole > pyroxene.

7.3. How to Deal with Ancient Sandstones?

Heavy mineral investigations are important for the industry [123] and not of academic interest only. In unfossiliferous strata, heavy minerals may represent one of the most dependable tools for correlation [6]. In the common unlucky case that only durable minerals have been preserved, one good way to extract information on the ultimate metamorphic or igneous sources is to consider the ratio between couples of minerals having similar density and thus hydraulic behaviour. To this goal, Morton and Hallsworth [124] formally defined the ATi [100 × %apatite/(%apatite + %tourmaline], GZi [100 × %garnet/(%garnet + %zircon)], RZi [100 × %rutile/(%rutile + %zircon)], and CZi [100 × %Cr-spinel/(%Cr-spinel + %zircon)] indices. Because garnet and apatite are particularly sensitive to dissolution in soils [26,44,111,125], the ATi and GZi indices are affected by weathering during the sedimentary cycle, and thus may provide information on paleoclimatic conditions. Care should be taken in handling these indices, which generally reflect selective diagenetic dissolution as well, not all durable minerals being equally durable (e.g., garnet generally far less than zircon) [12] (Figure 3 and p. 239 in reference [12]).

The classical ZTR index (sum of zircon, tourmaline, and rutile over total transparent heavy minerals) [101], widely used to evaluate the durability of a heavy mineral assemblage, integrates the effects of both pre-depositional weathering and post-depositional dissolution accumulated through an undetermined number of sedimentary cycles, and thus it is fundamentally a rough estimator of the extent of recycling. Other indicators such as the Hornblende Colour Index, the Metasedimentary Mineral Index, or the Sillimanite Index (Table 1) can seldom be used, because of the insufficient occurrence of these minerals in ancient sandstones. Varietal studies on durable (e.g., tourmaline, rutile, Cr-spinel, apatite) [126–129] or semi-durable detrital minerals (e.g., garnet) [130] represent another good viable option [131].

Table 1. Heavy mineral indices to be used in modern sediments or sedimentary rocks poorly affected by diagenetic dissolution (based on data from modern Alpine sands) [16,132].

Mineral Index	Greenschist Facies	Amphibolite Facies			Granulite Facies	
		Lower	Middle	Upper	Metasediments	Metagabbro
Hornblende Colour Index	-	≤10	10–30	30–60	>60	≥90
Metasedimentary Mineral Index	0	50	75	100	100	-
Sillimanite Index	-	-	0	≤30	>80	-

Hornblende Colour Index: HCI = (1/3 green + 2/3 green/brown + brown hornblende)/total hornblende × 100; Metasedimentary Mineral Index: MMI = (St/2 + Ky/2 + And/2 + Sil)/(chloritoid + staurolite + kyanite + andalusite + sillimanite) × 100; Sillimanite Index = prismatic sillimanite/total (fibrolitic + prismatic) sillimanite × 100.

7.4. The Recycling Nightmare

In J.L.Borges's novel "*The writing of God*" [133], the Aztec priest Tzinacan is imprisoned in a stone-walled cell in the threatening company of a jaguar. While striving to decipher the divine formula inscribed on the animal's skin (Figure 8), he falls into a noteworthy sedimentological nightmare "*I dreamt there was a grain of sand on the floor of the prison. Indifferent, I slept again. I dreamt I awoke and that on the floor there were two grains of sand. I slept again. I dreamt that the grains of sand were three. They went on multiplying in this way until they filled the prison and I lay dying beneath that hemisphere of sand. I realized that I was dreaming; with a vast effort I roused myself and awoke. It was useless to awake; the innumerable sand was suffocating me. Someone said to me: You have not awakened to wakefulness, but to a previous dream. This dream is enclosed within another, and so on to infinity, which is the number of grains of sand. The path you must retrace is interminable and you will die before you ever really awake*". This poetical prose describes the sense of doom felt when trying to retrace the ultimate provenance of recycled sand grains. How could we know whether the information carried by a zircon grain does refer to the present cycle, or instead to the previous one, or perhaps to the one before, and so on, and so on? How could we tie such information with any of the many panoramas created and destroyed during the successive evolutionary episodes that shaped the face of our planet? The path is fraught with difficulties, but the junior woodchuck will not be discouraged to take the few hundred million steps needed to reach the moon!

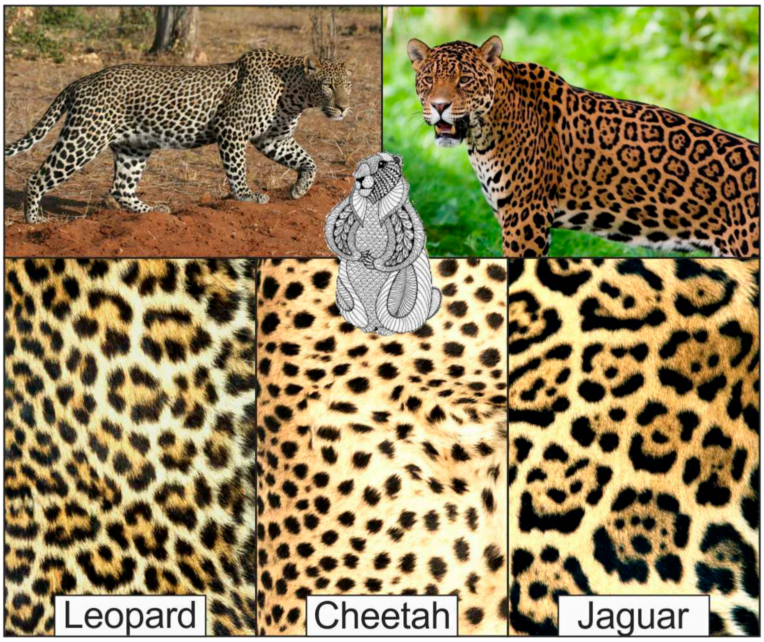

Figure 8. Messages encrypted in animal's skin as bar codes. A few years after Jorge Luis Borges wrote "*The writing of God*" fascinated by the jaguar's fur at Buenos Aires zoo, Alan Turing [134] envisaged a reaction-diffusion model of colour pigments to explain patterns on animal skins [135,136].

8. Conclusions

In this playful little handbook, we have tried to condense the experience acquired in twenty years of heavy mineral research and to provide practical and conceptual advice pointing at those mistakes most commonly made in heavy mineral analysis. The most important piece of advice is not to use heavy minerals alone (or worse a single heavy mineral species only), but to combine heavy mineral and petrographic analysis in thin section routinely. This also helps to visualize the heavy mineral concentration in our sample, the outer message that provides the key to understand the inner message carried by the heavy mineral suite. Our suggestions are based largely on modern case studies, which allow combining information on sources, processes, and products with theoretically complete control of all factors potentially affecting detrital mineralogy. In the study of modern sediments, we can evaluate, and in optimal cases quantify, the relative importance of each process. Moreover, we do not have to tackle the thorny problem of diagenesis, which becomes a nightmare to solve if combined with recycling, because the effect of recycling basically equates to multiple events of chemical dissolution cumulated through successive sedimentary cycles. Sooner or later, the junior woodchuck will find the answer, as Tzinacan eventually deciphered the divine design encrypted in the spots of the jaguar skin.

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