



## 碎屑锆石UPb年代学定量物源分析的基本原理与影响因素

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# 碎屑锆石 U-Pb 年代学定量物源分析的基本原理与影响因素

## ——以现代河流砂为例

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**摘要** 碎屑锆石 U-Pb 年代学数据获取快, 物源对比精确度高, 还可以估算源区剥蚀量, 在定量物源分析方面具有显著优势, 广受沉积学界青睐。但由于采样、实验过程中的不确定性, 常常导致一些物源判别结果存在多解性, 甚至产生了很多争议。从碎屑锆石 U-Pb 年代学定量物源分析的原理入手, 综述了由于沉积水动力、母岩锆石产率、沉积再旋回、人类活动、以及数据获取与处理 5 方面因素对年龄谱可能产生的影响。结果表明, 河流砂相比地层中的沉积岩, 物源区母岩性质明确, 运移路径非常清晰, 可以进行锆石产率的准确测定, 并能够同时开展混合模型正演和反演, 是理想的定量物源分析研究对象。对开展基于现代河流砂的定量物源分析机理研究进行了展望, 指出应用新技术、新方法开展小流域碎屑锆石 U-Pb 年代学研究是揭示锆石侵蚀、搬运和沉积过程行为机理的重要手段、也是构建定量物源分析方法的重要基础, 将为规范开展沉积地层的物源研究提供重要的理论依据。

**关键词** 碎屑锆石; 定量物源分析; 现代河流砂; 锆石产率; 混合模型

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## 0 引言

当前源—汇系统研究倍受国内外地球科学家关注, 已经成为指导能源矿产资源勘探<sup>[1-2]</sup>、重建深时地质与气候过程<sup>[3-4]</sup>、揭示人类生存环境与表层系统协同演变<sup>[5-6]</sup>等前沿领域的重要研究内容<sup>[7-10]</sup>。作为源—汇系统研究的核心手段, 物源分析近年来得以快速发展。据 Scopus 数据库最新的统计数据, 相关的研究文献达到 800 篇/年(图 1a)。从 Pettijohn 砂岩分类模式的建立<sup>[11]</sup>, 到 Dickinson 基于砂岩模式的大地构造单元的划分<sup>[12]</sup>, 再到现今单颗粒、多指标物源综合判别<sup>[13]</sup>, 物源分析正逐步从“定性”走向“定量”<sup>[14-18]</sup>, 应用范围也从传统的盆地分析<sup>[19-21]</sup>, 扩展到古地理、古气候、古地貌等众多领域<sup>[22-25]</sup>。

定量物源分析(quantitative provenance analysis)是物源分析发展的重要趋势。早期的物源分析借助砂岩的碎屑组分<sup>[12]</sup>、重矿物组合特征<sup>[26]</sup>、粉砂或黏土的元素地球化学<sup>[27]</sup>等指标可以区分物源区的大地构造单元, 但由于受到风化、后期成岩等影响, 这些指标难以保留原始的源区信号, 而且在造山带尺度的空间分辨率之下, 这些方法也很难准确判定提供碎屑的岩石单元, 更无法确定源区的贡献量的大小<sup>[28-29]</sup>(图 1b)。Molinaroli *et al.*<sup>[30]</sup>最早提出了定量物源分析的概念, 即不仅可以定量估计不同源区的贡献量, 还能够根据贡献量确定源区剥蚀量。然而, 定量物源分析直到最近十余年才开始快速发展, 这得益于碎屑单矿物分析技术的广泛应用<sup>[31-32]</sup>。利用该技术可以对沉积物(岩)中的特征重矿物(如锆石、磷灰石等)进

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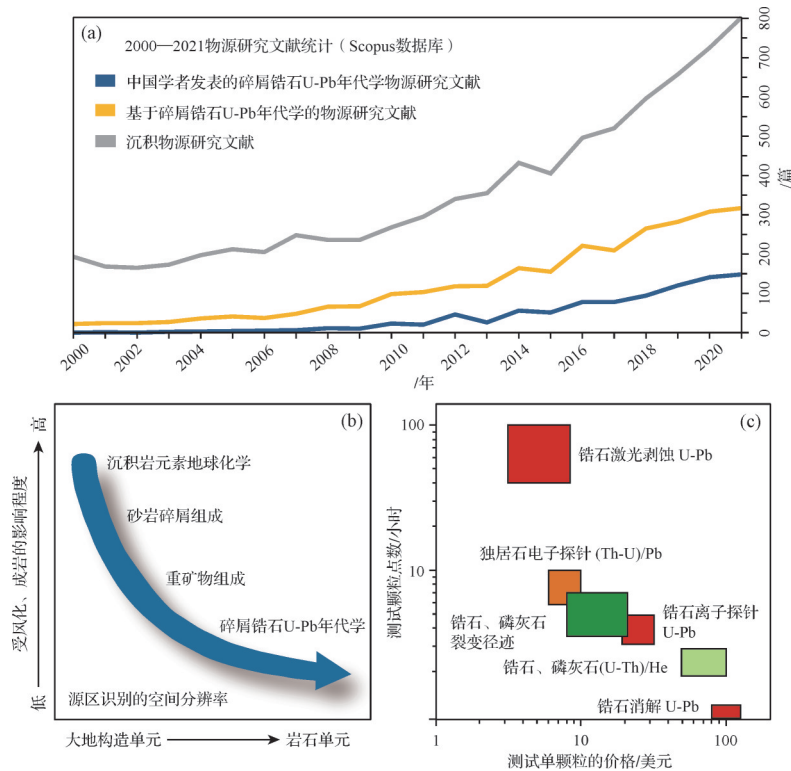


图1 沉积物源分析研究的现状和趋势

(a)近20年物源研究文献统计;(b)物源分析方法的优劣对比;(c)单颗粒碎屑年代学技术的成本对比(据文献[33]修改)

Fig.1 Research status and trend in sediment provenance

(a) number of papers published in the last 20 years; (b) comparison of different methods for provenance study; (c) costs for different single-grain detrital geochronology (modified from reference [33])

行快速的年代学或同位素分析,能够与源区岩石建立准确的联系<sup>[33-34]</sup>。由于锆石的化学稳定性强,抗风化、受成岩作用影响小<sup>[35-36]</sup>,且在地壳中广泛存在<sup>[37]</sup>,碎屑锆石U-Pb年代学方法发展最快、应用最广。

最早开展碎屑锆石U-Pb年代学研究的案例见于20世纪60年代<sup>[38-39]</sup>,并在80年代开始应用于沉积物源分析<sup>[40-41]</sup>。然而,限于化学消解实验效率,早期应用范围非常有限。直到2000年前后,二次离子探针(SHRIMP、SIMS)和激光烧蚀等离子体质谱(LA-ICPMS)技术开始应用于碎屑锆石U-Pb年代学<sup>[42-44]</sup>,该方法才开始真正推广,并一跃成为物源分析的“新宠”(图1c)。很多学者开始尝试借助此方法,期待取得物源判别的突破,然而结论却仍然存在很大的争议。例如,松潘—甘孜复理石盆地是最早开展碎屑锆石U-Pb年代学物源研究的区域之一<sup>[45]</sup>。一些学者认为锆石主要来自远源的秦岭,甚至大别山,包括大量华北和华南板块的贡献<sup>[45-49]</sup>;还有一些学者认为主要来自近源地块(如昆仑、羌塘等)<sup>[50-55]</sup>或者呈现多源的特征<sup>[56-58]</sup>。再如,古金沙江是否曾经南流的问题也是近年的研究

热点。通过对新生代地层开展的碎屑锆石U-Pb年代学物源分析,一些结果支持古金沙江南流<sup>[59-64]</sup>,但是也有不少证据反对南流<sup>[65-67]</sup>,还有学者甚至对碎屑锆石U-Pb年代学方法的科学性提出了质疑<sup>[68]</sup>。

随着采用碎屑锆石U-Pb年代学的物源研究不断增多,研究区域相同但解释结果不一致甚至相互矛盾的现象也时有发生,部分研究也暴露出数据泛化、年龄谱重现率低、信息挖掘不足、物源解释主观性大等问题,使得一些地质问题的解释更加不确定,一定程度上也影响了该方法在定量物源分析的应用效果<sup>[68]</sup>。造成这种局面的原因,一方面是对碎屑锆石在侵蚀、搬运和沉积过程中的行为机理了解不够深入,另一方面,对样品采集、处理、测试和数据处理等流程中可能对数据解释产生影响的关键因素缺乏有效的量化约束。鉴于此,本文基于现代河流砂的研究成果,系统总结了碎屑锆石U-Pb年代学定量物源分析的原理,分析了5方面因素对定量物源分析结果可能造成的偏差,在此基础上,对未来开展碎屑锆石U-Pb年代学定量物源分析提出了展望和建议。

# 1 碎屑锆石 U-Pb 年代学定量物源分析的基本原理

通常情况下,碎屑锆石U-Pb年龄值呈离散分布,根据年龄值相近程度,可以划分成不同的组分,构成多组分统计分布,可以用直方图、饼图、概率密度图(PDP)、核密度估计图(KDE)等形式表示<sup>[69-71]</sup>,即U-Pb年龄谱。相对于传统的碎屑成分、重矿物、元素地球化学等分析,碎屑锆石U-Pb年代学的优势在于可以依据单矿物的年龄与源区对比,获得精确的源区信息,并在此基础上开展统计分析和定量计算。基于年龄谱对物源进行定量分析基本原理如图2所示,具体应用体现在以下三个方面。

## 1.1 目视判别源区相对贡献量

通过碎屑锆石U-Pb年龄谱与源区母岩的U-Pb年龄相对比,根据经验判别源区,估算不同源区的相对贡献量。Cawood *et al.*<sup>[72]</sup>对澳大利亚西南Frankland流域(长度320 km,面积4 630 km<sup>2</sup>)的河流砂进行了系统的碎屑锆石研究,验证其对河流所流经的岩石单元的响应情况。结果表明,随着流经地基岩从太古代变质岩到元古代花岗岩的变化,在约100 km的范围内,太古代碎屑锆石年龄组分从大于90%下降到20%左右,表现出年龄组分的高空间分辨率的物源响应特征。这种思路普遍被用于现代河流的物源示踪研究中,包括对长江<sup>[73]</sup>、雅鲁藏布江<sup>[74]</sup>的源区贡献量进行估算。

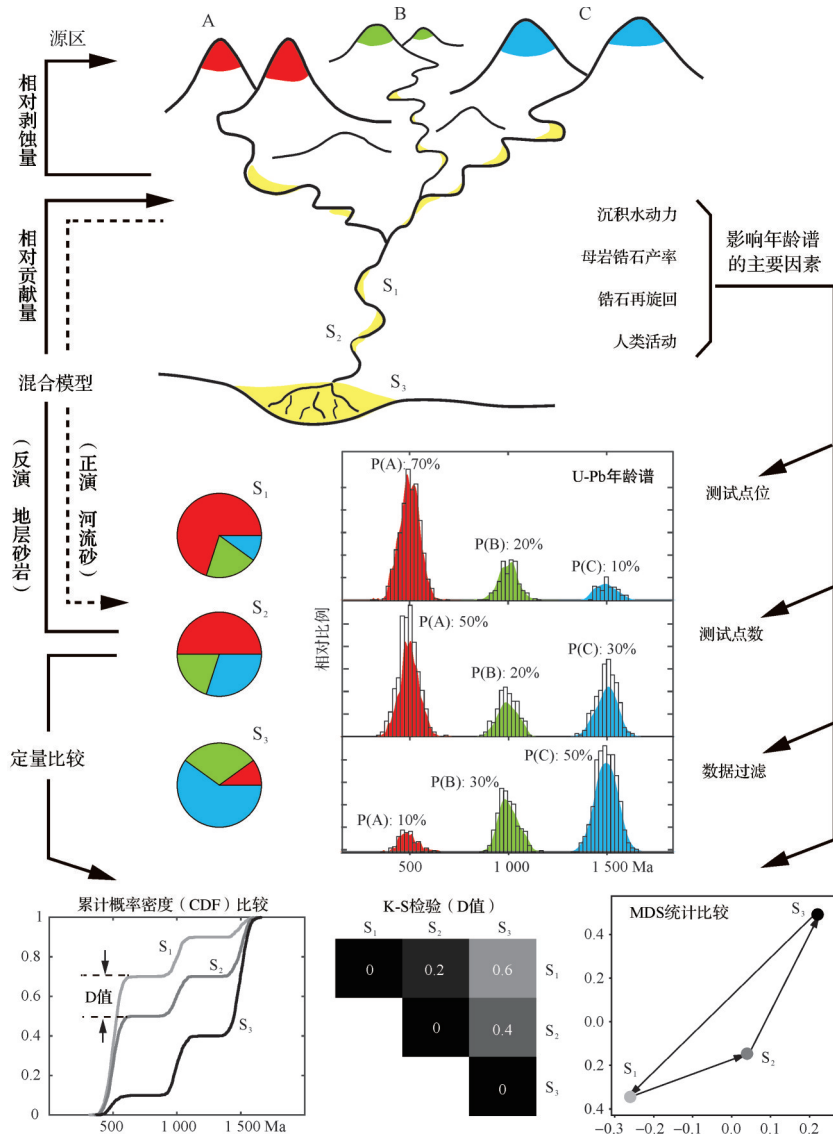


图2 碎屑锆石U-Pb年代学定量物源分析的基本原理与影响因素

Fig.2 Principle of quantitative provenance analysis and major effects from the result

## 1.2 U-Pb 年龄谱定量比较

目视判别对于年龄谱相近、源区可辨识度低的情况,无法获得有效的结果。近年来的研究表明,利用统计学方法对不同样品的碎屑锆石 U-Pb 年龄谱进行定量比较,可以大幅提高源区对比的准确度。常用的统计学方法包括累计概率密度法(CDF)<sup>[75]</sup>、Kolmogorov-Smirnov 统计检验(K-S 检验)<sup>[76-77]</sup>、相似度参数法<sup>[78-79]</sup>、多维定标法(MDS)<sup>[80-81]</sup>,以及贝叶斯方法<sup>[82]</sup>。不论哪种方法,它们都基于特定形式的年龄谱,而非直接的碎屑锆石年龄数据的比较。在得到的比较结果中(图2),如K-S检验的D值,代表了统计上两件样品来源的相似程度(D值越小越相似),而MDS则更为直观地用二维或三维图示距离来表示,适用于复杂源区与沉积碎屑的对比<sup>[83]</sup>。目前此种方法已经被应用于多种沉积环境之中,如河流沉积<sup>[24-25,84]</sup>、边缘海沉积<sup>[85-86]</sup>、风成沉积<sup>[87]</sup>等,取得了良好的效果。

## 1.3 混合模型计算相对贡献量

采用碎屑锆石 U-Pb 年龄谱混合模型,正演(Mixing)或反演(Unmixing)不同源区的相对贡献量和剥蚀量。相对于上述直接利用 U-Pb 年龄谱与母岩 U-Pb 年龄对比估算源区的相对贡献量的方法,这种方法能够规避复杂源区误判带来的误差,且正演和反演也能够相互验证,提高了定量物源分析的准确度。Amidon *et al.*<sup>[88-89]</sup>对尼泊尔 Marsyandi 流域河流砂的碎屑锆石 U-Pb 年代学的研究开创了该方法的先例。作者建立了沉积物正演混合模型(公式1):假设仅有三个源区的情况,其中  $P(A)$ 、 $P(B)$  和  $P(C)$  为三个源区的年龄谱, $P(S)$  为混合后沉积碎屑的年龄谱, $\phi_a$  为 A 源区的贡献率, $\phi_b$  为 B 源区的贡献率,而  $1-\phi_a-\phi_b$  是 C 源区的贡献量,在已知  $P(A)$ 、 $P(B)$ 、 $P(C)$  和  $P(S)$  的情况下,采用 Monte Carlo 近似算法对  $\phi$  进行迭代,寻找最佳的  $\phi$  值,以满足公式 1 能够通过 K-S 检验。

$$\phi_a P(A) + \phi_b P(B) + (1 - \phi_a - \phi_b) P(C) = P(S) \quad (1)$$

后来的研究者又引入了多种方法,如 bootstrap 方法<sup>[90]</sup>、最小二乘方法<sup>[91]</sup>、非负矩阵分解法<sup>[92]</sup>替代 Monte Carlo 方法,并且可以选取多种统计检验形式对公式 1 进行逼近<sup>[93]</sup>,但思路与 Amidon *et al.*<sup>[88-89]</sup> 基本一致。反过来,对地质历史时期的沉积岩,采用相似的思路,在流域未知的情况下对碎屑锆石 U-Pb 年龄谱进行反演,估算源区的贡献量。目前该技术在黄土高原<sup>[94-95]</sup>、三角洲地区<sup>[96]</sup>等复杂源区判别方面已经取得了不少重要进展。

## 2 碎屑锆石 U-Pb 年代学定量物源分析的影响因素

针对可能造成物源解释分歧的原因,近年来学术界也开始关注碎屑锆石 U-Pb 年代学物源分析的方法学研究。例如在澳大利亚西南 Frankland 流域<sup>[72]</sup>、尼泊尔喜马拉雅 Marsyandi 流域<sup>[88]</sup>、北美阿巴拉契亚 French Broad 流域<sup>[97]</sup>、欧洲的阿尔卑斯 Po 流域<sup>[98]</sup>、南美安第斯 Mendoza 流域<sup>[99]</sup>等造山带的小流域建立了定量物源分析的实验区,开展了一系列碎屑锆石 U-Pb 年代学物源分析的方法验证和机理探索,并发现了诸多可能造成年龄谱解释和物源判别偏差的影响因素,主要认识体现在以下几个方面。

### 2.1 沉积水动力影响年龄谱解释

经典的沉积水动力学研究表明,在牵引流的作用下,颗粒沉降受到密度、大小及形态等因素的控制<sup>[100-101]</sup>。根据普遍接受的沉降等效(settling equivalence)原理<sup>[102]</sup>(图3),重矿物颗粒发生沉降时的粒径大小与密度相关。特定的重矿物,如锆石,将会富集在样品特定的粒度区间<sup>[103-105]</sup>。因此,选择合

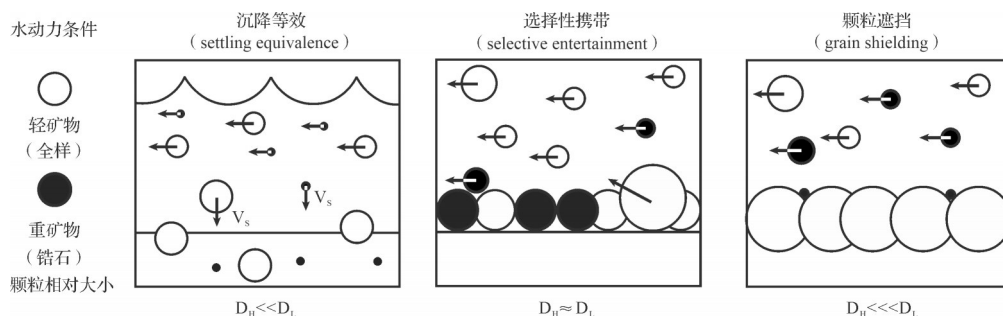


图3 不同水动力条件下重矿物(锆石)沉积的机制(据文献[102]修改)

Fig.3 Three scenarios considering the hydrodynamic condition for relative sizes of heavy and light mineral grains (modified from reference [102])

适的粒度区间进行碎屑锆石U-Pb年代学分析,对于获得准确的、有代表性的年龄谱就显得至关重要。然而,实际情况往往是,由于采样过程中沉积微环境的差别,以及锆石提取、制靶和测试过程中不可避免的人为干扰,很难使用到真实合理的粒度区间,可能会造成年龄谱失真。

对于碎屑锆石样品采样,普遍建议选择沉积环境相似、分选较好的中砂或细砂<sup>[29,91]</sup>。然而实际研究表明,即便在同一处采样点附近,水动力分选也会造成年龄谱较大的偏差。Lawrence *et al.*<sup>[106]</sup>在亚马逊河对波长20 m,波高1.25 m的单个沙丘选取5个不同位置采样,获得了5件粒度不同的样品,它们的年龄谱存在显著差异。Ibañez-Mejia *et al.*<sup>[107]</sup>在Rio Orinoco三角洲的同一河道截面的心滩、边滩等不同位置采取的3件样品,平均粒度分别是75 μm、130 μm和220 μm,结果显示碎屑锆石的年龄谱也明显不同。类似的现象在地层样品中也曾多次报道<sup>[108-109]</sup>,反映了采样的沉积微环境可能造成的影响。

另外,在锆石的提取过程中,当前采用较多的浮选法虽然效率较高,但很可能损失一些粒径小(<63 μm)的锆石<sup>[110]</sup>;在制靶过程中,如采用较多的人工选粘锆石的方法,也更倾向选择粒径大、形态好的颗粒<sup>[71,110-111]</sup>。在使用LA-ICPMS测试过程中,受束斑大小的影响,一些小颗粒、长宽比高的锆石无法测试,也会导致年龄谱倾向大颗粒的锆石<sup>[112]</sup>。

究竟粒度偏差会怎样影响年龄谱解释?很多学者开展了相关的机理研究。Lawrence *et al.*<sup>[106]</sup>利用亚马逊河的现代河流砂进行了系统的碎屑锆石粒度一年龄的关系研究,结果表明,细的锆石颗粒往往年龄较老,而粗的颗粒则相对年轻。Yang *et al.*<sup>[113]</sup>分析了长江的现代河流砂的粒度一年龄关系,也得到了类似的分布特征。这种关系同样也表现在一些地层样品中<sup>[114]</sup>,可能反映了老的锆石颗粒由于多次旋回,在磨蚀作用下粒径变小的趋势。据此推测,如果在采样、测试过程中人为倾向大颗粒锆石,年龄谱将会偏向年轻的年龄组分。然而,近期的研究却指出,锆石年龄和粒度之间不存在明显的相关性<sup>[109,115-117]</sup>。例如,Leary *et al.*<sup>[116]</sup>的研究结果表明,近源的锆石粒径偏大且分选差,而远源的锆石的粒径较小且分选较好,即粒度只是在一定程度上反映了源区的距离,而锆石年龄取决于相对应的源区,不存在锆石越老、粒度越小的普遍趋势。依此结论,如果在采样、测试过程中

人为倾向大颗粒锆石,则可能丢失一部分远源的信息。

造成对粒度一年龄关系不同认识的原因可能有两个方面。一方面,可能来自测量方法的差异。一些研究对粒径的测量采用等效球径方法<sup>[106,113]</sup>,而另一些则采用长、短轴统计的方法,不仅能约束颗粒大小,还可以利用长短轴比值近似计算磨圆度<sup>[114,116]</sup>。由于这些方法都是在已经制靶并且抛光的锆石截面上进行的,计算结果受颗粒产状和截面位置的影响。最近的研究也开始采用三维的几何约束方法,包括进行锆石形态的定性分类<sup>[118-119]</sup>以及使用更先进的三维测量技术<sup>[115]</sup>。Markwitz *et al.*<sup>[115]</sup>利用高分辨率的显微CT,对澳大利亚的Murchison河流砂碎屑锆石的形态进行了大量的三维形态分析,发现不同粒度锆石年龄谱差异主要反映的是源区距离,而不是再旋回的特征。另一方面,锆石颗粒的沉积过程复杂,除了颗粒密度控制的沉降以外,还有选择性携带(selective entrapment)、颗粒遮挡(grain shielding)等多重因素<sup>[102]</sup>,可能打乱粒径分布的规律性<sup>[120]</sup>。针对这一现象,Cantine *et al.*<sup>[102]</sup>采用正演模拟的方法,基于不同源区的锆石粒度差异建立了考虑粒度的混合模型(图4),并施加了不同的沉积水动力条件,模拟结果充分说明沉积水动力对于年龄谱存在明显的控制作用。

## 2.2 母岩锆石产率影响相对剥蚀量估算

通常情况下,母岩区提供的锆石在沉积物碎屑锆石U-Pb年龄谱中的占比,即源区相对贡献量 $\phi$ ,与源区母岩中锆石的产率呈正相关,如公式2所示。

$$\phi \propto Zrc_{\text{yield}} \times A \quad (2)$$

式中: $A$ 是母岩或流域的面积( $\text{m}^2$ ),是单位面积锆石的产率( $\text{kg} \cdot \text{Ma}^{-1} \cdot \text{m}^2$ )。Zrc<sub>yield</sub>又可以表示为锆石含量 $C_{\text{zrc}}$  ( $10^{-6}$ )和剥蚀速率 $E$  ( $\text{m} \cdot \text{Ma}^{-1}$ )的函数,如公式3。由于母岩的密度 $\rho_{\text{bulk}}$  ( $\text{kg} \cdot \text{m}^{-3}$ )可知,因此可以根据贡献量 $\phi$ 和锆石含量 $C_{\text{zrc}}$ 估计剥蚀速率 $E$ 的相对值,即相对剥蚀量。

$$Zrc_{\text{yield}} = C_{\text{zrc}} \times E \times \rho_{\text{bulk}} \quad (3)$$

研究表明,不同岩性的锆石含量 $C_{\text{zrc}}$ 可以从 $10^{-3}$ 到 $10^{-5}$ 不等<sup>[121]</sup>。对于火成岩而言,锆石通常在 $\text{SiO}_2$ 含量大于60%的中酸性岩浆中结晶,而在基性岩浆中非常少见;对于沉积岩,只在碎屑岩中广泛存在,而在碳酸盐岩中几乎没有锆石;对于变质岩,变质程度只有达到角闪岩相到麻粒岩相,才能产生新的变质

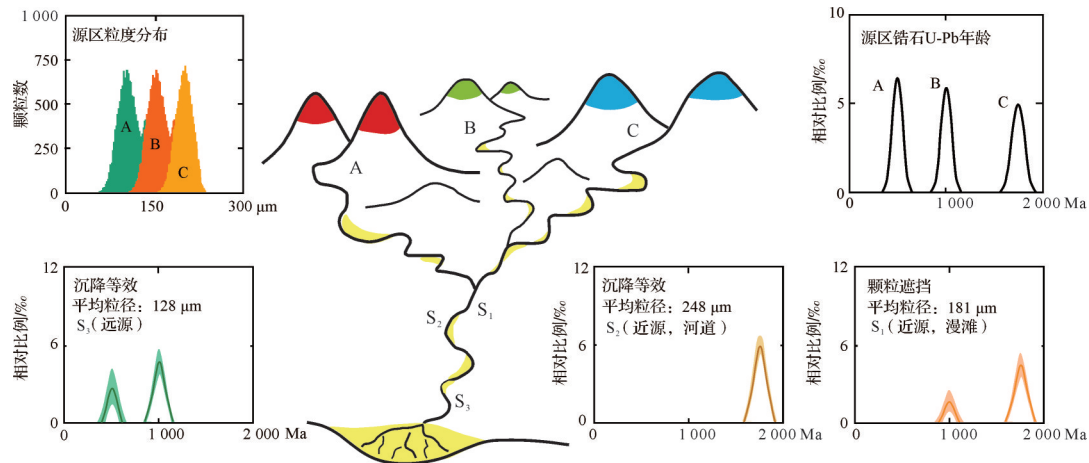


图4 不同沉积位置(远源、近源)、不同水动力条件下(沉降等效、颗粒遮挡)碎屑锆石年龄谱和平均粒径的数值模拟结果(据文献[102]修改)

Fig.4 Age spectra and grain size of detrital zircon at three sampling locations (distal and proximal) for the two hydrodynamic condition (settling equivalence and grain shielding). Results are based on numerical modeling (modified from reference [102])

锆石<sup>[37]</sup>。即使对于同一种岩性,锆石含量可以相差5倍之多<sup>[122]</sup>,这将严重影响物源的解释和贡献量的估算<sup>[110,123-124]</sup>。

在以往的研究中,往往忽略母岩锆石产率(含量)的影响或假设含量相近<sup>[73-74,125]</sup>。这样做虽然可以省去统计锆石含量的复杂流程,但代价是在剥蚀量的估算时就会偏向锆石含量高的母岩源区。因此,相对剥蚀量的估算必须首先确定锆石的含量。然而,直接统计锆石含量需要非常严格的重矿物分选和统计流程<sup>[98]</sup>,过程十分复杂。Amidon *et al.*<sup>[88]</sup>使用元素地球化学方法测定河流砂中Zr元素含量,来近似计算锆石含量,如公式4所示。式中 $mm_{zrc}$ 是锆石( $ZrSiO_4$ )的摩尔质量( $g \cdot mol^{-1}$ ), $mm_z$ 是Zr元素的摩尔质量( $g \cdot mol^{-1}$ ), $Zr_{bulk}$ 是母岩中Zr元素含量( $10^{-6}$ )。Amidon *et al.*<sup>[88]</sup>将Zr元素法与颗粒统计法得到的锆石含量相对比,发现两者基本相当。很显然,测定母岩Zr元素含量更为快捷,而且往往可以通过河流砂的Zr元素含量代替上游流域母岩的平均Zr元素含量,使得计算更为简便。

$$C_{zrc} \approx Zr_{bulk} \cdot \frac{mm_{zrc}}{mm_z} \quad (4)$$

事实证明,在考虑锆石含量后,将相对贡献量转化为剥蚀量(公式2和3联立),估算的相对剥蚀量与实际情况更为接近。例如在阿尔卑斯Po流域,根据河流砂碎屑锆石年龄谱计算的贡献量,经过锆石含量校正后,得到的剥蚀量与宇宙成因核素<sup>10</sup>Be估计的侵蚀速率趋于一致<sup>[98]</sup>。在澳大利亚西南Frankland流

域,一些早期认为可能是侵蚀速率差异影响造成的年龄谱差异<sup>[72]</sup>,在经过校正后,被重新解释为锆石的含量差异所致<sup>[124]</sup>。

然而,对于公式4中锆石含量的近似,目前仍然存在不同的认识。早期使用的都是河流砂全样的Zr元素含量<sup>[88,122-123]</sup>,而近期一些研究则采用细粒成分( $<63 \mu m$ )进行Zr元素含量分析<sup>[90]</sup>。最近,也有学者指出这种近似方案存在风险,例如Malusà *et al.*<sup>[98]</sup>采用非常严格的重矿物分选和统计流程,对比了重矿物统计和元素地球化学分析两种方法得到的锆石含量,发现二者的差异仍然比较显著(图5)。后者得到的锆石含量明显偏高,原因可能是除锆石以外的其他很多矿物也可能提供Zr元素,例如斜锆石、磷钇矿、甚至火山玻璃都可能造成统计的误差。

### 2.3 再旋回碎屑锆石造成物源判断不唯一

由于超强的抗风化能力,锆石一旦进入沉积系统就可以被反复埋藏、剥蚀,最终无法分清其“最初”的来源<sup>[126]</sup>。据估算,碎屑矿物中来自沉积岩的“再旋回”比例可能高达80%<sup>[29]</sup>。而在针对碎屑锆石的大量实际研究中也发现,再旋回的锆石在沉积物中非常普遍。例如,Campbell *et al.*<sup>[127]</sup>在对印度河和恒河的河流砂研究中,运用了U-Pb和(U-Th)/He双定年的方式对再旋回的锆石进行检验,发现只有约10%的锆石直接来自基底岩石。Anderson *et al.*<sup>[128-129]</sup>对南非现代沉积物开展了大量的碎屑锆石U-Pb年代学和Hf同位素分析,证明这些锆石全部来自新元古代或古生代砂岩中锆石的再旋回,很难与“最初”的源区

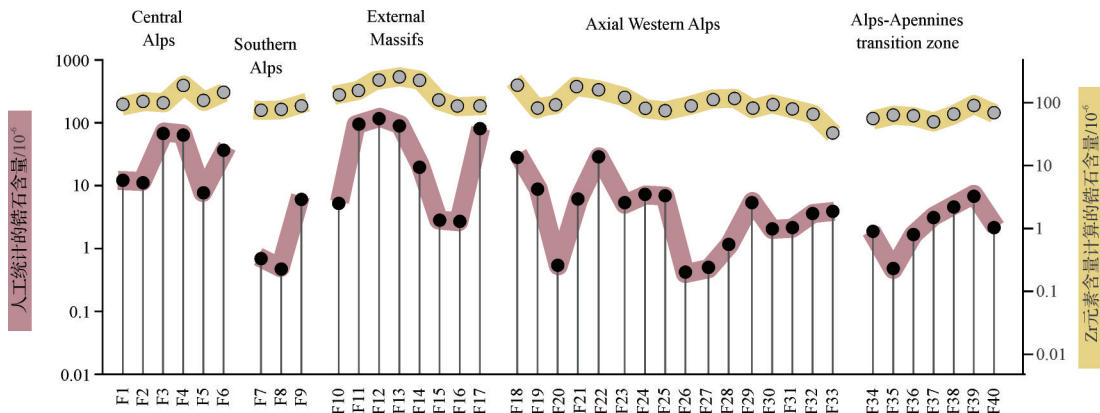


图5 重矿物统计和元素地球化学分析计算得到的锆石含量差异(据文献[98]修改)  
横坐标为样品编号,黑色圆点代表重矿物统计得到的锆石含量,灰色圆点代表元素分析得到的锆石含量

Fig.5 Comparison between heavy mineral counting and geochemical analysis for zircon fertility (modified from reference [98])  
samples are listed on horizontal axis, black dots are based on heavy mineral counting and gray dots on geochemical analysis

建立直接的源—汇关系。对于地质历史时期的沉积岩,例如美国亚利桑那白垩系河流相砂岩的碎屑锆石, Dickinson *et al.*<sup>[130]</sup>发现它们主要来自科罗拉多高原东部中—上侏罗统的风成石英砂岩中锆石的再旋回,但由于古地理的不确定性,确定这些再旋回锆石“最初”的来源则非常困难。

因此,简单地将盆地内碎屑锆石年龄谱与周缘的造山带基底岩石进行对比显然会忽略沉积岩再旋回对物源的贡献,容易造成物源的误判。传统方法根据定性的锆石形态学<sup>[118-119]</sup>,也很难达到区分再旋回锆石的目的。当前采用一些多重约束方法,例如,利用锆石的Hf同位素、微量元素等信息揭示锆石原始的岩浆性质,配合U-Pb年龄可以对锆石再旋回提供指示,但仍然很难介入到具体的沉积过程,从而判断哪些是单旋回,而哪些是再旋回锆石。近年来,针对单颗粒锆石采用U-Pb和(U-Th)/He双定年<sup>[131]</sup>,借助其他碎屑矿物(如楣石U-Th-Pb<sup>[132]</sup>、钾长石Pb同位

素<sup>[133]</sup>)协助鉴别再旋回和单旋回沉积物等等。然而,双重约束意味着成本的增加,而且上述方法的测试效率在短时间都难以赶上锆石U-Pb年代学,还没有得到广泛的应用。

#### 2.4 人类活动影响年龄谱解释

人类活动对碎屑锆石U-Pb年龄谱的影响体现在两个方面。首先,人类对河流进行改造和治理,例如建设水坝,阻挡沉积物向下游运移,使下游沉积物中的源区锆石U-Pb年龄峰值降低。具体过程是,当河流搬运沉积物进入水库,粗碎屑立即在水库边缘沉积<sup>[134]</sup>,细碎屑随即沉降到水库底部,因此在坝体出水口处沉积物被“过滤”,而基本不含源区的碎屑锆石颗粒(图6)。但是,从坝体排出的水会对坝体前端的河道沉积物进行强烈冲刷,被冲刷的河道沉积物被搬运至下游,在下游二次堆积,源区锆石的U-Pb年龄峰会重新出现。然而,由于没有源区沉积物的持续补给,下游沉积物中源区锆石U-Pb年龄峰值相比

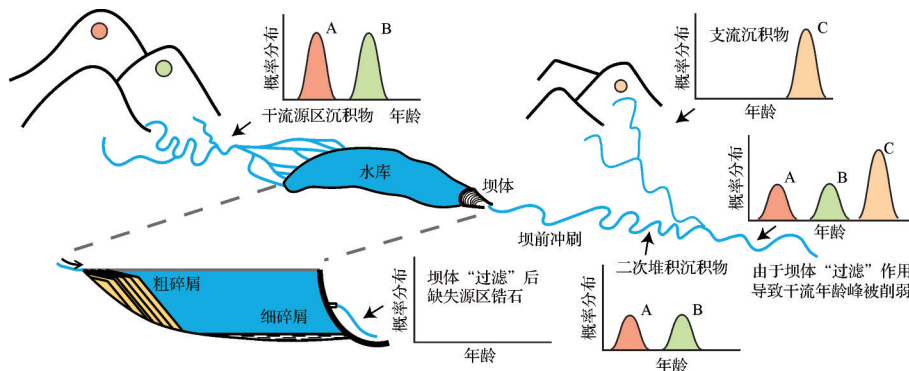


图6 建设水坝对下游碎屑锆石U-Pb年龄谱可能产生的影响示意图(据文献[135]修改)  
Fig.6 Effect of dams on the downstream detrital zircon age signals (modified from reference [135])



坝前沉积物有所降低。例如, Thomson *et al.*<sup>[135]</sup>使用混合模型对密苏里河流域及其主要支流流域进行了相对贡献量的计算,结果表明,在建设有水坝的河流中,支流的相对贡献量比干流更大。因此,水坝的建设,会对下游新堆积的河流沉积物的碎屑锆石年龄谱的解释造成一定偏差,具体表现为年龄峰值的削弱。

此外,人类过度的土地利用,会加速基岩或其上覆风化壳的侵蚀,造成局部地区剥蚀量的增加,以至于影响年龄谱的峰值形态。He *et al.*<sup>[73]</sup>对长江及其主要支流的碎屑锆石 U-Pb 年龄谱进行对比,并根据年龄组分估算了不同支流的相对剥蚀量,认为长江上游(如嘉陵江、汉江)的剥蚀量有限,而长江下游(如湘江、赣江)的剥蚀量较大,下游剥蚀量的增加是由人类活动所致。然而,如何准确评估由人类活动造成的剥蚀量增加对碎屑锆石年龄谱的影响,仍然具有挑战性。例如, Wissink *et al.*<sup>[136]</sup>对 He *et al.*<sup>[73]</sup>发表的 U-Pb 年龄谱重新进行了定量比较和混合模型的研究,发现长江干流的锆石贡献仍然主要来自上游(如嘉陵江流域),反映了青藏高原东缘构造活动的强剥蚀,并不支持下游人类活动导致剥蚀量增加的说法。

## 2.5 数据获取与处理过程影响年龄谱解释

碎屑锆石 U-Pb 数据获取与分析过程中测试点数、测试点位、数据过滤方法等条件的选择,都可能引起年龄谱的偏差。

### (1) 测试点数

由于往往无法将样品中所有碎屑锆石都进行分析,为了获得有代表性的 U-Pb 年龄谱,采取的方式类似统计学上的“抽样调查”。因此,测试的颗粒越多,越能反映真实的源区特征。但是对于具体的“抽样”数量,即样本量,仍然存在不同的认识。早期的一些学者认为,大约 60 颗锆石就可以反映物源特征<sup>[41]</sup>。然而,经过严格的统计学计算, Vermeesch<sup>[137]</sup>认为,要保证贡献量小的源区(5%)也能在年龄谱中被检测到,至少需要 117 颗锆石,而要想获得更小的源区信号(2%),样本量则需要增加到 300 颗<sup>[138]</sup>。最近一些研究又进一步提升了测试数量,例如采用单个样品大于 1 000 颗的大样本量(large-n)碎屑锆石研究。通过对结果采用 K-S 检验方法的定量比较发现,大样本量能够更好地指示物源,提高分析结果的重现性,尤其适合复杂年龄谱的物源分析<sup>[107,139]</sup>。

### (2) 测试点位

由于碎屑锆石的来源多样,其最初的成因可能是岩浆锆石、变质锆石、热液锆石或蛻晶化锆石等,并且可能含有继承性锆石或者包裹体,因此,在单颗粒锆石的不同位置进行 U-Pb 年龄测试,例如生长边和继承核,得到的年龄结果可能相差很大<sup>[140-142]</sup>。在对碎屑锆石进行测试分析时,由于点数多、成本高、颗粒小等因素,往往每个颗粒只分析一次,选择“边”还是“核”都可能对年龄谱造成影响。例如, Hietpas *et al.*<sup>[97]</sup>对流经阿巴拉契亚的 French Broad 流域的河流砂样品进行了测试点位的对比,在只分析“核”的情况下年龄谱中的年轻组分的占比很低;在既分析“核”又分析“边”的情况下,年轻组分的占比提升了近 10 倍。Bonich *et al.*<sup>[143]</sup>在加州东南花岗岩出露区的小流域进行了实验研究,发现有继承核的锆石是导致源区信息偏差的主要原因。Zimmermann *et al.*<sup>[144]</sup>对东南亚和阿尔卑斯的地层样品进行了更为详细的“核”、“边”对比研究,结果表明只分析“核”或者“边”都无法获得准确的年龄谱信息,并建议对单颗粒锆石采用“核一边”双点测试的策略,以保证物源解释的可靠性。最近, Liu *et al.*<sup>[145]</sup>在对北美阿巴拉契亚古生代前陆盆地的碎屑锆石 U-Pb 年代学研究过程中,采用了深度剖面方法对“核一边”双年龄进行了测试,这些核部年龄均为 Grenville 期,边部则出现了两组年龄,可以有效地区分源区。

### (3) 数据过滤

虽然碎屑锆石分析得到的 U-Pb 年龄结果精度较高,但就单颗粒而言,仍然存在由于普通 Pb 含量高、Pb 的丢失、U 的过剩等现象造成的年龄误差<sup>[146]</sup>,例如  $^{206}\text{Pb}/^{238}\text{U}$  年龄与  $^{207}\text{Pb}/^{235}\text{U}$  或者  $^{207}\text{Pb}/^{206}\text{Pb}$  年龄的“不谐和”现象。常用的处理方法是先计算年龄误差百分比,再采用谐和度阈值(5%~30%)将此类数据过滤掉<sup>[147]</sup>。另外,受测试精度影响,最优年龄的取值通常也受阈值控制,通常年龄较大(>1 000 Ma)的锆石选用  $^{207}\text{Pb}/^{206}\text{Pb}$  年龄,而年龄较小时则选用  $^{206}\text{Pb}/^{238}\text{U}$  年龄。如果这些数据过滤的标准或阈值选取不当,也可能造成年龄谱的误差<sup>[34,148]</sup>。最近的研究建议,采用二次计算得到的谐和年龄作为碎屑锆石的最优年龄,可以规避阈值风险<sup>[108,149]</sup>,而且新的谐和度计算方法,也会使数据过滤更加趋于合理<sup>[149]</sup>。

### 3 展望与建议

碎屑锆石 U-Pb 年龄谱除了包含了源区的信息,也包含了很多物源以外的信息,如前所述的沉积水动力、源区剥蚀效率、人类活动等等,甚至实验过程中产生的偏差。前人通过河流砂的碎屑锆石 U-Pb 年代学研究,已经认识到这些因素的影响,并逐步开始通过改进实验方法或施予合理的校正,实践证明可以获得定量、可靠的分析结果<sup>[90,98]</sup>。河流砂相比地层中的沉积岩,物源区母岩性质明确,运移路径非常清晰,可以进行锆石产率的准确测定,并能够同时开展混合模型正演和反演,是理想的定量物源分析研究对象。

近年来,国内学者针对现代河流砂也开展了大量碎屑锆石 U-Pb 年代学物源研究,集中在两个应用领域;一是针对现代流域的源—汇过程研究,主要集中在长江<sup>[73,113,150]</sup>、黄河<sup>[151-154]</sup>、雅鲁藏布江<sup>[74]</sup>等大河流域,二是针对现代样品的年龄谱中的峰值,开展的流域构造演化分析<sup>[151,155]</sup>。相比之下,真正关注河流砂定量物源分析机理的研究则很少。此外,大河流域的岩石单元出露多样,但很难获知全流域的母岩锆石产率,复杂的构造—沉积演化历史产生了大量的再旋回锆石,且流域内的构造、气候造成的显著的剥蚀差异,多重因素叠加在一起,不利于开展定量物源分析的方法验证。反之,一些小流域可能更适合开展定量验证和机理探索。近年来,利用造山带小流域河流砂开展的碎屑锆石 U-Pb 年代学研究初见成效。例如, Deng *et al.*<sup>[156]</sup>对台湾的浊水溪和兰阳溪两个小流域进行河流砂碎屑锆石 U-Pb 年代学研究,利用正演混合模型估算了来自大陆不同地块的贡献量。Guo *et al.*<sup>[125]</sup>针对西藏拉萨河、年楚河和朋曲河三个小流域的河流砂进行了包括锆石在内的多种矿物的 U-Pb 年代学分析,估算了不同源区母岩的贡献量。

由此可见,小流域的现代河流砂碎屑锆石 U-Pb 年代学研究,不仅是揭示锆石侵蚀、搬运和沉积过程行为机理的重要手段,也是构建定量物源分析方法的重要基础,将为规范开展沉积地层的物源研究提供重要的理论依据。通过上述的原理和因素分析,结合最新的技术发展,针对碎屑锆石 U-Pb 年代学定量物源分析提出以下建议。

(1) 采样环节。需要确保采集的样品碎屑锆石 U-Pb 年龄谱能客观、真实地反映物源特征。包括避

开可能的人为干扰区,并收集样品的沉积微环境、粒度、分选性等沉积结构信息。采用一些新的技术,如三维形态测量技术(如显微 CT)能够更为准确地获取锆石颗粒的粒度、磨圆度等几何信息<sup>[115]</sup>,建立更为可靠的单颗粒年龄—性状(如粒度)之间的关系。

(2) 测试环节。需要确保从颗粒挑选、分析测试到数据处理的规范化、标准化。最近,采用改进的 LA-ICPMS 进样系统使得短时间内获得大样本量 (large-n) 碎屑锆石数据成为可能<sup>[139,157]</sup>,即可以提升统计数据的质量,也可以保证物源定量比较的准确度;采用“核一边”双年龄测试,不但可以区分源区,还可能指示源区发生的多期构造过程<sup>[144-145]</sup>。针对单矿物,采用多种测试方法相结合,例如锆石的(U-Th)/He 分析,还可能区分多旋回和单旋回锆石<sup>[131]</sup>。

(3) 定量分析环节。需要确保能够快速、准确地计算出源区母岩的锆石产率,以建立年龄谱与剥蚀量之间的定量关系。除了传统的元素分析法,还可以利用扫描电镜矿物自动识别系统(如 TIMA、QEMSCAN)计算样品的锆石含量(产率)<sup>[104,158]</sup>,对相对贡献量实施校正,得到更为准确的剥蚀量估计。此外,将锆石与其他重矿物的 U-Pb 年代学相结合,例如榍石、金红石等,可以对贡献量进行对比分析,避免单矿物分析可能造成的误差<sup>[125]</sup>。

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## Principles and Biases of Quantitative Provenance Analysis Using Detrital Zircon U-Pb Geochronology: Insight from modern river sands

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**Abstract:** Detrital zircon U-Pb geochronology is a powerful tool for deciphering Earth's sedimentary archive, answering a large amount of research questions including sediment provenance and palaeogeographic correlations. Although sound reasons exist to conduct qualitative analysis, the advent of large-n acquisition techniques and readily available statistical tools have provoked a shift towards quantitative analysis as the preferred approach during the last two decades. The assumption for a geologically meaningful interpretation of inter-sample comparison through detrital zircon age distributions is that the analyzed samples are a true reflection of the sediment sampled, and this can be used as a proxy for the relative proportion of crystalline rocks in the source region. However, this foundational assumption may be undermined by a number of biases, leading to controversial interpretations and a risk of abuse of the method. In this paper, we demonstrate the principles of the quantitative provenance analysis using detrital zircon U-Pb geochronology and review five major factors that can add bias to the age spectrum and influence the provenance interpretation. The results show the significance to using modern river sand for testing the fidelity of the detrital zircon geochronology because the zircon fertility, hydrological sorting, and sediment mixing can be better constrained than ancient fluvial sequence. Based on the new technologies, including large-n sampling, rim-core dating, and three-dimensional zircon morphology, the quantitative relationship between sedimentary hydrodynamics and age spectra are established, providing further instruction for the provenance study of deep-time sedimentary strata.

**Key words:** detrital zircon; quantitative provenance analysis; modern river sand; zircon fertility; mixing model