

UNIVERSIDADE DE SÃO PAULO
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**Quartz Luminescence Sensitivity as a Sediment Provenance tracer:
Perspectives from the Northern Andes Orogenic basins**

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ABSTRACT

In recent years, the luminescence of quartz has emerged as a potential tool for provenance analysis, particularly through the application of optically stimulated luminescence (OSL) sensitivity. While OSL sensitivity in quartz crystals from igneous and metamorphic rocks is typically low, previous findings indicate that OSL sensitivity can vary within five orders of magnitude within Quaternary sediments. These observations imply that quartz sensitization, in natural settings, occurs at some stage between the sediment source areas and the depositional sites. Laboratory experiments have demonstrated that sensitization of the quartz OSL fast component can be prompted by irradiation-bleaching cycles or by heating at temperatures above 300 °C. These sensitization mechanisms could be driven by surface processes that expose quartz grains to ionizing radiation and sunlight or to high temperatures, including sediment reworking, soil mixing, and wildfires. Thus, the longer sediments remain on the Earth's surface, the higher the probability of those grains undergoing sensitization through surface processes. In this way, the extended residence of sediments on the surface increases the opportunities for quartz grains to experience natural irradiation, bleaching by sunlight exposure, and heating, increasing the probability of sensitization. This hypothesis establishes a connection between OSL sensitivity and surface processes occurring mainly in the source areas, ultimately enabling the tracing of the provenance of sediments released from particular source areas characterized by a specific combination of source rock and surface processes.

OSL sensitivity has been effectively utilized for provenance analysis in modern deposits, but its application in ancient basin-fill sequences has been limited. This dissertation aims to address this gap by investigating the OSL sensitivity of pure quartz sand grains extracted from sandstones of a ~10 km-thick exhumed crustal section of the Northern Andes in the Colombian Eastern Cordillera, as well as from one of its adjacent basins. The studied section encompasses rocks from the entire Phanerozoic eon that have a well-established provenance history determined through various methods including low-temperature thermochronology, detrital geochronology, and sandstone petrography. This comprehensive array of information provides a strong foundation to establish a proof-of-concept for the applicability of quartz OSL sensitivity

as a sediment provenance tracer. To achieve this, in this work the OSL sensitivity, measured in sandstones from the Colombian Eastern Cordillera, is compared with proxies such as detrital zircon U-Pb geochronology and sandstone petrography. Here, two different scenarios within the same basin in the Northern Andes were tested. Firstly, changes in quartz OSL sensitivity were demonstrated to be correlated with changes in sediment source areas characterized by contrasting conditions related to the residence time that quartz grains spend on the Earth's surface. Low-sensitivity quartz is associated with orogenic source areas experiencing rapid exhumation and denudation, whereas quartz exhibiting higher sensitivity is correlated with stable cratonic source areas. Finally, the highest-sensitivity quartz is linked to the recycling of sedimentary rocks that exhibit enhanced quartz OSL sensitivity. These observations allow for the construction of a hypothetical frame that serves as a basis for using the quartz luminescence sensitivity as a provenance tracer not only on quaternary and modern sediments but also in ancient basin-fill sequences.

Keywords: Quartz, OSL sensitivity, Northern Andes, Provenance, Source areas

RESUMO

Nos últimos anos, a luminescência do quartzo surgiu como uma ferramenta potencial para a análise da proveniência, particularmente através da aplicação da sensibilidade da luminescência opticamente estimulada (OSL). Embora a sensibilidade OSL em cristais de quartzo de rochas ígneas e metamórficas seja tipicamente baixa, descobertas anteriores indicam que a sensibilidade OSL pode variar dentro de cinco ordens de magnitude em sedimentos quaternários. Estas observações implicam que a sensibilização do quartzo, em ambientes naturais, ocorre numa determinada fase entre as áreas fonte dos sedimentos e os locais de deposição. Experimentos laboratoriais já demonstraram que a sensibilização da componente rápida OSL do quartzo pode ser provocada por ciclos de irradiação-esvazamento ou por aquecimento a temperaturas superiores a 300 °C. Estes mecanismos de sensibilização podem ser provocados por processos superficiais que expõem os grãos de quartzo à radiação ionizante e à luz solar ou a temperaturas elevadas, incluindo o retrabalhamento de sedimentos, a mistura de solos e os incêndios florestais. Assim, quanto maior for o tempo de permanência dos sedimentos na superfície terrestre, maior será a probabilidade desses grãos sofrerem sensibilização através de processos superficiais. Desta forma, a permanência prolongada dos sedimentos à superfície aumenta as oportunidades de os grãos de quartzo sofrerem irradiação natural, esvazamento por exposição à luz solar e aquecimento, aumentando a probabilidade de sensibilização. Esta hipótese estabelece uma ligação entre a sensibilidade OSL do quartzo e os processos superficiais que ocorrem principalmente nas áreas fonte, permitindo, em última análise, o rastreio da proveniência de sedimentos erodidos de determinadas áreas fonte caracterizadas por uma combinação específica de rochas fonte e processos superficiais

A sensibilidade OSL tem sido efetivamente utilizada para a análise da proveniência em depósitos modernos, mas a sua aplicação em sequências antigas de de bacias tem sido limitada. Esta dissertação visa colmatar esta lacuna, investigando a sensibilidade OSL de grãos de areia de quartzo puro extraídos de arenitos de uma secção crustal exumada com cerca de 10 km de espessura dos Andes do Norte, na Cordilheira Oriental da Colômbia, bem como de uma das suas bacias adjacentes. A secção estudada abrange rochas de todo o éon Fanerozoico, as quais têm uma

história de proveniência sedimentar bem estabelecida, determinada através de vários métodos, incluindo termocronologia de baixa temperatura, geocronologia detrítica, e petrografia de arenito. Este conjunto de informações fornece uma base sólida para estabelecer uma prova de conceito para a aplicabilidade da sensibilidade OSL do quartzo como um traçador de proveniência sedimentar. Para tal, neste trabalho a sensibilidade OSL medida em arenitos da Cordilheira Oriental Colombiana é comparada com proxies como a geocronologia U-Pb de zircão detrítico e a petrografia de arenitos. Aqui, foram testados dois cenários diferentes dentro da mesma bacia nos Andes do Norte. Em primeiro lugar, demonstrou-se que as mudanças na sensibilidade OSL do quartzo estão correlacionadas com mudanças nas áreas fonte dos sedimentos, caracterizadas por condições contrastantes relacionadas com o tempo de residência que os grãos de quartzo passam na superfície da Terra. O quartzo de baixa sensibilidade está associado a áreas fontes orogénica com rápida exumação e denudação, enquanto o quartzo com maior sensibilidade está correlacionado com áreas fontes cratónicas estáveis. Por fim, o quartzo de maior sensibilidade está associado à reciclagem de rochas sedimentares que apresentam uma sensibilidade OSL do quartzo mais elevada. Estas observações permitem a construção de um quadro hipotético que serve de base à utilização da sensibilidade da luminescência do quartzo como traçador de proveniência, não só em sedimentos quaternários e modernos, mas também em sequências antigas de bacias.

Palavras-chave: Quartzo, Sensibilidade OSL, Andes do Norte, Proveniência, Areas fonte

INTRODUCTION

The sediment provenance analysis is an important field in sedimentary geology and has been widely used to associate sediment compositional signatures with sedimentary processes, including weathering, erosion, and transport (Haughton et al., 1991). Advancements in the plate tectonics theory and the development of technology to acquire, process, and analyze massive datasets allow for linking sand detrital modes not only to sedimentary processes but also to geodynamic settings, particularly, regarding the evolution of orogens and their associated sedimentary basins (Najman, 2006; Caracciolo, 2020). Meanwhile, provenance analysis has usually been performed employing classic techniques such as sandstone petrography (e.g., Garzanti, 2016) or using accessory mineral phases in sediments such as heavy mineral assemblages (Garzanti and Andò, 2019) and detrital geochronology, especially U-Pb in zircon (e.g., Gehrels, 2014).

Over the last decade, applied dosimetry in earth sciences has witnessed a significant expansion of frontiers beyond sediment dating, including novel applications that rely on luminescence properties to track sediment movement (e.g., Rhodes and Leathard, 2022) and fingerprint provenance (e.g., Sawakuchi et al., 2018; Capaldi et al., 2022). One of these properties is optically stimulated luminescence (OSL) sensitivity, defined as the luminescence response of a material to an applied radiation dose (Mineli et al., 2021). Extensive research for OSL dating purposes has documented substantial variability of luminescence sensitivity in quartz across different geomorphic and geological settings (Hu et al., 2020). Indeed, quartz grains originally released from crystalline rocks tend to exhibit low sensitivity (e.g., Sawakuchi et al., 2011; Guralnik et al., 2015) and are potentially sensitized through multiple bleaching and irradiation cycles by exposing quartz grains to light, heat, and ionizing radiation (Moska and Murray, 2006). As a result, quartz sensitivity from sedimentary deposits can vary at least five orders of magnitude (Mineli et al., 2021). Although it is not completely understood, in natural settings, quartz sensitization cycles could be driven by surface processes such as wildfires (e.g., Zhang et al., 2023), sediment reworking (e.g., Pietsch et al., 2008), and soil mixing (e.g., Gray et al., 2019). Thus, the rates at which those surface processes occur, which modulate sediment generation and production conditions, would link quartz OSL sensitivity with the residence time of sediments on

the Earth's surface (e.g., Sawakuchi et al., 2018) and long-term sedimentary recycling (e.g., Fitzsimmons, 2011; del Río et al., 2021) allowing for the capacity to discriminate the sediment provenance from contrasting source regions.

Fingerprinting sediment provenance using quartz luminescence signals offers numerous advantages, highlighting its potential for further research investment and refinement. These advantages lie in the abundance of quartz, a ubiquitous mineral on the Earth's surface and the most common component of sands and sandstones. Consequently, sample preparation requires smaller volumes, and limitations related to source fertility are minimal. Additionally, luminescence measurements offer high analytical sensitivity, are cost-effective, and are easier to conduct compared to other measurement techniques (Sawakuchi et al., 2020). Furthermore, the potential of quartz OSL sensitivity to track changes in sediment production conditions including sediment residence time in source areas and long-term sedimentary recycling, coupled with other provenance proxies, enhances interpretations regarding sediment routing systems.

Quartz OSL sensitivity in provenance analysis has typically been explored in active sediments (e.g., Bartyik et al., 2021; Capaldi et al., 2022) and quaternary deposits (e.g., Liu and Li, 2006; Lü and Sun, 2011; Nian et al., 2019), but it has only been applied a few times in basin-fill sequences (e.g., del Río et al., 2021) and never in orogenic settings. Therefore, no record represents a robust basis for extending the applicability of quartz OSL sensitivity as a provenance tracer in ancient basin-fill sequences. Additionally, the role of the quartz sensitization mechanisms and the sensitization process itself remains unknown over geological time. One natural experiment to address these concerns involves characterizing the quartz OSL sensitivity in basins where multiple physical and geological variables are well-known and other provenance proxies are available. This approach allows for assessing the effects of the addition and depletion of sediments released from contrasting source regions on quartz OSL sensitivity measured in sedimentary basin-fill sequences. Following this line of reasoning, the Colombian Northern Andean basins stand out as a suitable scenario for conducting such an experiment, as (i) its most external mountain range in Colombia, the Eastern Cordillera, is a Mesozoic back-arc basin which underwent moderate tectonic inversion during Andean orogenesis, that resulted in exposure and recycling of abundant siliciclastic sedimentary rocks into foreland basin strata until the present;

and (2) there is robust control over the major shifts in their sediment source regions through geological time from various provenance datasets. In these basins, multiple proxies, including sandstone and conglomerate petrography (e.g., Parra et al., 2010; Bayona et al., 2013), detrital zircon U-Pb geochronology (e.g., Horton et al., 2010; Saylor et al., 2011), U-Th/He thermochronometry in detrital apatites and zircons (e.g., Horton et al., 2010a; Saylor et al., 2012; Odoh et al., 2019), and reworked palynomorphs (e.g., De La Parra et al., 2015), allow for the recognition of different source regions that experienced contrasting sediment generation and production conditions such as the Amazon Craton and the Andean Magmatic Arc.

This work aims to provide a proof-of-concept of the potential of quartz OSL sensitivity for tracing provenance in sedimentary basin-fill sequences by comparing sensitivity data with other well-established proxies for provenance used in the Northern Andes basins. The structure of this dissertation consists of four chapters describing all the work performed during the master's degree including two manuscripts to be submitted to international peer-reviewed journals, which are the core of this work. The first chapter is the introduction of the dissertation presenting the fundamentals of optically stimulated luminescence in quartz and its previous uses in provenance analysis. In the second chapter, the response of quartz OSL sensitivity to documented changes in contrasting sediment source regions (e.g., cratonic vs orogenic) is assessed, providing insights into the potential of this technique as a provenance proxy. The third chapter presents long-term sedimentary recycling as a quartz sensitization mechanism in geological timescales and how this characteristic is useful in the provenance analysis within sedimentary basin-fill sequences. Finally, the fourth chapter summarizes the main conclusions of all the work carried out in the frame of this master's degree, emphasizing the uses of quartz OSL sensitivity in the provenance analysis and the future work to be developed to establish a robust method in different geological settings.

CHAPTER 4: MAIN CONCLUSIONS AND PERSPECTIVES

For the first time, in this work, quartz OSL sensitivity data was acquired in an almost complete supracrustal section spanning rocks from the whole Phanerozoic eon. This dataset represents an archive for assessing changes in this luminescence characteristic over geological time.

In the second chapter, quartz OSL sensitivity is demonstrated to correlate with previously constrained shifts in sediment source areas. In this case, the source area is characterized by being composed mainly of crystalline rocks either orogenic (Andes) or cratonic (Guyana Shield). Therefore, regardless of the source bedrock, low-sensitivity quartz is associated with orogenic source areas, either in extensional or compressional settings, experiencing rapid denudation. Conversely, quartz that exhibits enhanced sensitivity is correlated with distal and stable cratonic source areas. This observation, now supported by other proxies such as detrital zircon U-Pb geochronology, set the base for the use of this technique to study changes in quartz OSL sensitivity in ancient basin-fill sequences and correlate them with changes in contrasting source areas. Thus, this proxy along with other tools can be very useful in tracing sedimentary provenance.

In the third chapter, quartz OSL sensitivity is also demonstrated that varies according to previously constrained shifts in sediment source areas. Nonetheless, the logic here is different, the source bedrock changes from crystalline and volcanic rocks in an orogenic source area, such as the Andean magmatic arc emplaced in the Colombian Central Cordillera to recycled sedimentary rocks that characterize the Eastern Cordillera. In this case, quartz with enhanced OSL sensitivity is correlated with the recycling of rocks triggered by the uplift and erosion of the Eastern Cordillera. One interesting observation is that quartz grains derived from the recycling of the sedimentary rocks that were sourced in the Guyana Shield exhibit significantly higher sensitivity than those derived from the recycling of the Andean first-cycle sedimentary rocks. These observations complement the conclusions obtained in the second chapter and allow for the construction of a hypothetical frame to assess variations in quartz OSL sensitivity not only in modern and quaternary sedimentary covers but also in rocks from the entire phanerozoic eon (Figure 11).

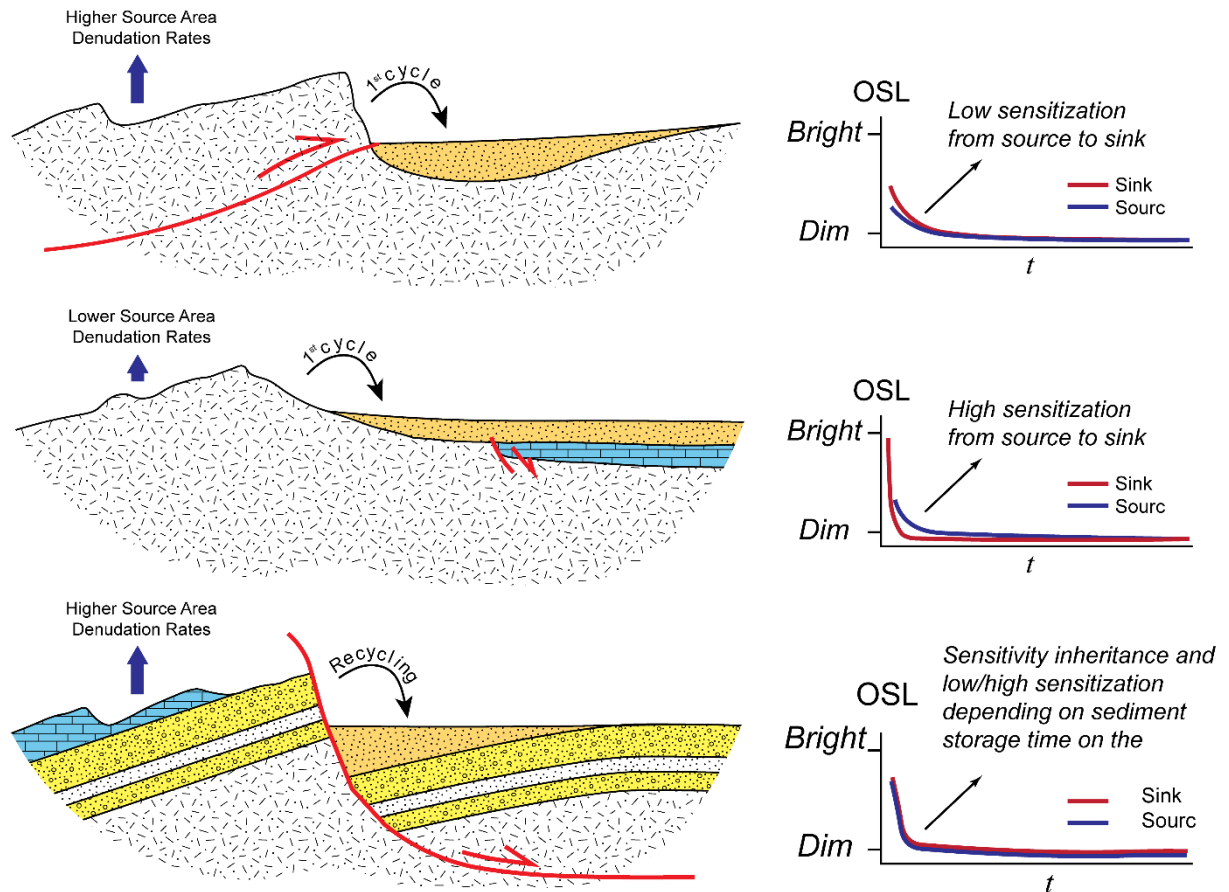


Figure 11. Hypothetical frame to understand changes in quartz OSL sensitivity. Source area denudation rates are correlated with sediment residence time on the Earth's surface (see Sawakuchi et al., 2018).

The first observation to consider in this hypothetical frame is that quartz derived from crystalline and volcanic rocks generally exhibits low-sensitivity fast components (e.g., Alexanderson, 2020 Guralnik et al., 2015; Mineli et al., 2021; Sawakuchi et al., 2011) and that quartz sensitization in natural settings occurs mainly on the Earth surface due to exposure of multiple bleaching and irradiation cycles or by heating induced by wildfires (e.g., Pietsch et al., 2008). Following this line of reasoning, the greater the time that sediments spend on the Earth's surface, the greater the probability of enhancing sensitivity on the surface. This hypothesis correlates with source area denudation rates (e.g., Sawakuchi et al., 2018). Furthermore, quartz grains derived from recycled sedimentary rocks exhibit higher quartz OSL sensitivity when compared with those released from crystalline rocks. Since recycling means that grains go through at least twice on the Earth's surface, they can be potentially sensitized in the subsequent cycles they spend on the surface. All these premises are supported by the dataset studied in this dissertation and previous observations made on quaternary

deposits. In this way, sediment source areas characterized by certain types of bedrock (more crystalline or sedimentary) and contrasting denudation rates (slow or rapid) would exhibit a distinctive OSL sensitivity imprint. Therefore, changes in quartz OSL sensitivity can be correlated with changes in these sediment source areas. Instead of tracing provenance using minerals or characteristic ages of a group of rocks, as is the case with detrital zircon U-Pb geochronology or heavy minerals, the sensitivity of quartz allows for fingerprinting the provenance of unique source regions that are differentiated by the type of exposed rock and the processes of sediment generation, production, and erosion that ultimately control the residence time that quartz grains spend on the surface. This perspective opens a new opportunity to establish quartz luminescence sensitivity as a sediment provenance tracer in rocks and sediments.

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