



## Research paper

# Quartz OSL sensitivity as a proxy for storm activity on the southern Brazilian coast during the Late Holocene

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## ABSTRACT

Natural cycles of irradiation during burial and bleaching due to solar exposure during transport increase the Optically Stimulated Luminescence (OSL) sensitivity of quartz sand grains. The relationship between the OSL sensitivity and sediment transport allows to discriminate quartz sand grains with different depositional histories. In this paper, we evaluate the variation of OSL sensitivity in quartz grains deposited during the progradation of the Ilha Comprida barrier on the southern Brazilian coast. Changes in sand sensitivity recorded by barrier growth since 6 ka ago are controlled by the variation in the proportion of low versus high sensitivity quartz grains. Low sensitivity grains with short sedimentary history are supplied by the Ribeira de Iguape River and reach the barrier through southward alongshore currents during fair weather conditions. Storm conditions shift the alongshore currents to northeast and permit the transport of high sensitivity grains with long sedimentary history from distal southern coastal sectors to the barrier. Therefore, the input of distal sediments for the Ilha Comprida barrier depends on the frequency and intensity of storms. Thus, the OSL sensitivity can be used as proxy for storm activity. The variation of OSL sensitivity through time indicates that the Ilha Comprida barrier changed from a relatively stable to an unstable storm pattern around 2 ka ago. Periods with increased storm activity peaked around AD 500, AD 1500 and AD 1850, approximately on the boundaries of the Medieval Climate Anomaly and the Little Ice Age.

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## 1. Introduction

Coastal barriers formed by well-sorted fine sands dominated by quartz are common features in tropical or subtropical zones such as that from Brazil and Australia. Finding paleoclimate proxies for these sedimentary deposits has been challenging due to the monotonous mineralogical composition and absence of organic materials. This is the case for the southern Brazilian coast that presents large Holocene prograded barriers with well-preserved sets of sand ridges. Although paleoclimate records for onshore (Behling et al., 2004; Cruz et al., 2006) and offshore (Chiessi et al., 2008) regions of Southern Brazil have been obtained in the last couple of years, paleoclimate proxies for coastal sands would fill

a gap and allow a more complete reconstruction of past climates in Southern Brazil. In general, geological paleoclimate proxies would correspond to components within sediments able to record changes in climate sensitive systems such as lakes, forests and caves. Currently, the wave climate which determines the intensity and direction of alongshore currents in the southern Brazilian barriers has a pronounced change when climate conditions shift from fair weather to storm weather (Pianca et al., 2010). Therefore, the sand provenance in these barriers would be climate sensitive and suitable to be used as a paleoclimate proxy.

Thermoluminescence (TL) and optically stimulated luminescence (OSL) are promising quartz provenance indicators. A provenance indicator based on quartz luminescence combined with sediment OSL dating would allow coastal barriers to be used as paleoclimate records. Efforts to find a relationship between the TL/OSL characteristics and the provenance of quartz have been made during the last decades (Hashimoto et al., 1989; Rink et al., 1993;

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Rink, 1999, 2003; Preusser et al., 2006; Choi et al., 2006; Chithambo et al., 2007; Keizars et al., 2008; Pietsch et al., 2008; Westaway, 2009; Tokuyasu et al., 2010; Sawakuchi et al., 2011; Fitzsimmons, 2011; Tsukamoto et al., 2011; Lü and Sun, 2011). However, the development of reliable TL/OSL provenance proxies has been difficult due to ambiguity in the geological significance of the luminescence properties of quartz.

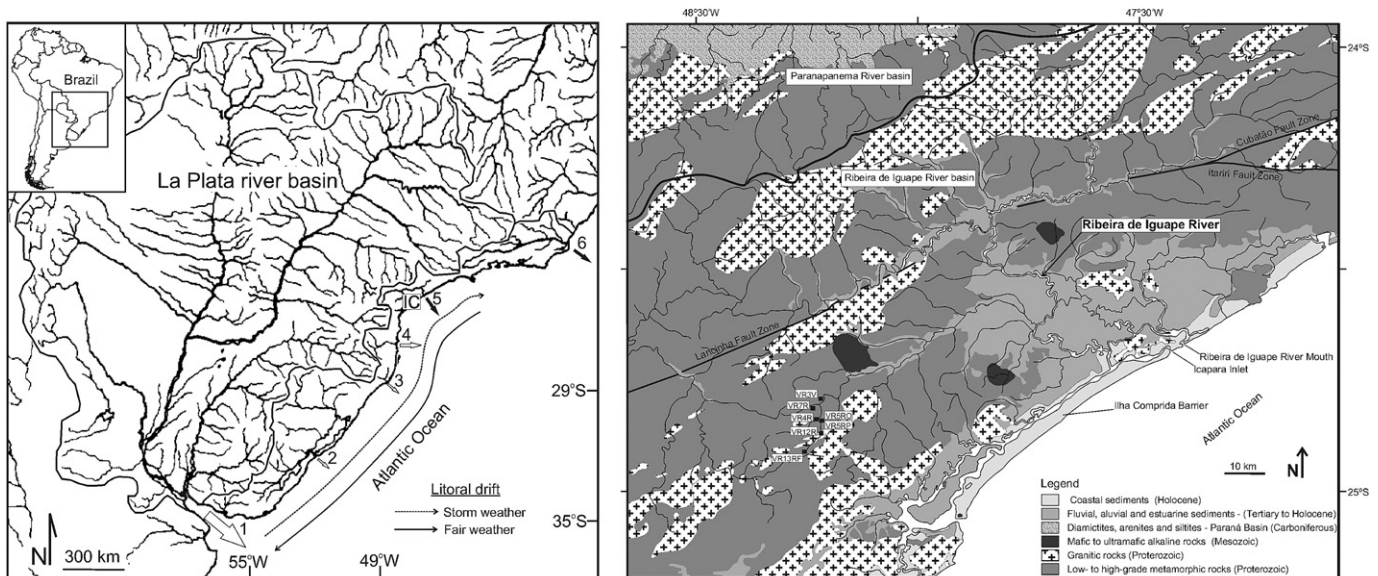
Pioneering studies by Rink (1999, 2003) attempted to use the residual TL signal decay as a tracer for sediment transport in coastal systems. The gradual decay of the natural TL signal of quartz along transport routes allows one to determine the direction of coastal currents as well as the source of sediment grains since their last burial episode (Rink, 1999, 2003). Keizars et al. (2008) also estimated the residence time of sand grains in the swash zone through the calibration of the residual TL signal decay using laboratory bleaching experiments. In spite of the suitability to identify the source of sediments with a natural TL signal acquired during temporary burial, this method is limited by the depletion of the TL signal. In the coastal zone, this depletion would occur on timescales from hours to centuries (Rink, 2003). The residual TL signal is useful to outline the direction and source of sediments under transport in active depositional systems but it is not suitable for sediments that are not under transport or with completely depleted TL signal. Another approach is based on the increase of the quartz OSL sensitivity during sediment transport (Pietsch et al., 2008). This increase is supposed to be cumulative and irreversible in sedimentary systems, and is therefore supposedly able to record the sedimentary history of quartz grains.

The definition of OSL sensitivity is the light intensity emitted per unit radiation dose. As demonstrated by laboratory experiments, cycles of irradiation and bleaching enhance the OSL sensitivity of quartz (McKeever et al., 1996; Li, 2002; Moska and Murray, 2006; Koul and Chougaonkar, 2007). During the sedimentary transport of quartz grains, these cycles correspond to periods of irradiation during burial after deposition, followed by bleaching due to solar exposure during erosion. Independently of the effect of other process on quartz OSL sensitivity, we hypothesize that in depositional environments with a dominant mode of transport and mixing between sediments with contrasting degree of reworking,

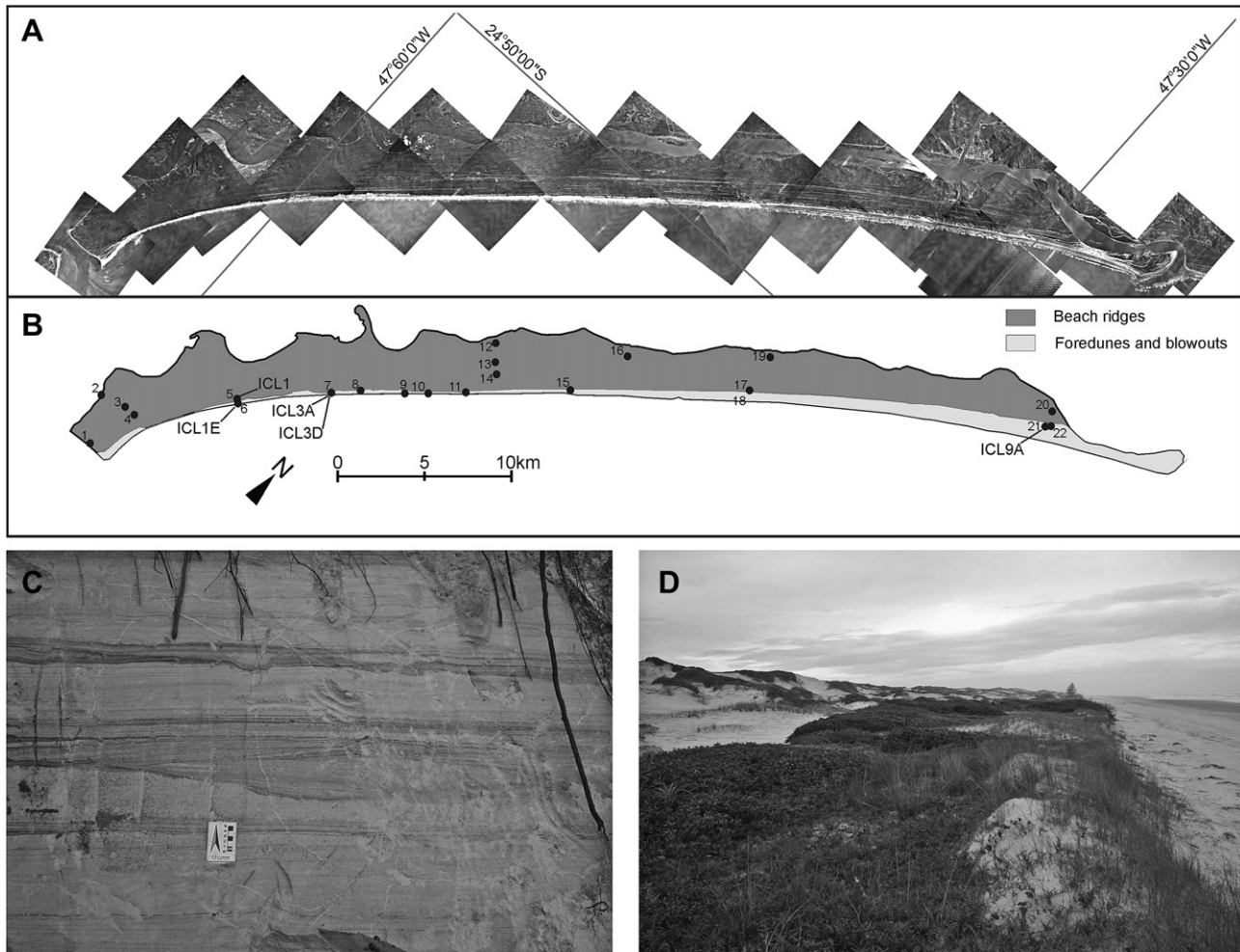
the quartz OSL sensitivity could be a provenance indicator able to differentiate sediments with distinct depositional histories. The Ilha Comprida barrier, on the southern Brazilian coast (Guedes et al., 2011a), comprises sands whose OSL sensitivity of their quartz grains vary in many orders of magnitude (Sawakuchi et al., 2011). The huge variation in the OSL sensitivity of these sands is attributed to the mixture of grains with different depositional histories regardless of the source rock for the quartz grains (Sawakuchi et al., 2011). This would allow the discrimination between sediments with short and long depositional histories. In this paper, we evaluate the quartz OSL sensitivity in sands from the Ilha Comprida barrier. This barrier is composed of sets of beach ridges and dune ridges deposited by the progradation of a wave-dominated coastal system since the Middle Holocene (Giannini et al., 2009). Its well-defined depositional chronology (Sawakuchi et al., 2008; Guedes et al., 2011a) and sands with known source rocks (Guedes et al., 2011b) make the Ilha Comprida barrier suitable to use the quartz OSL sensitivity as a proxy for provenance. The connection between sediment provenance and shifts in the coastal currents correlated to the swap from fair weather to storm weather will allow us deduce past climate changes through the variation of quartz OSL sensitivity during the development of the Ilha Comprida barrier.

## 2. Study setting

According to Giannini et al. (2009), the Ilha Comprida is a Holocene sand barrier developed in a microtidal setting (Fig. 1). It corresponds to a prograded barrier formed by beach and dune ridge alignments (Fig. 2) deposited since the Middle Holocene (Giannini et al., 2009; Guedes et al., 2011a). The climate in the Ilha Comprida barrier region is wet subtropical, with a mean annual precipitation of 1583 mm and a mean annual temperature of 21.1 °C (IBGE, 1992; IPCC-DCC, 2000). The barrier is located in the region of the South Atlantic Convergence Zone (SACZ) where tropical air masses meet with polar air masses, forming cold fronts that advance northward (Satyamurti et al., 1998). Trade winds from east and northeast and associated swell wave systems prevail during the action of tropical air masses. The northward migration of cold fronts takes bad



**Fig. 1.** At the left, the Ilha Comprida barrier in the context of the major alongshore transport pathways and fluvial systems that supply sediments to the southern Brazilian coast. The numbers indicate the major rivers delivering sediments to the coastal system: 1. La Plata River; 2. Guaíba River; 3. Tubarão River; 4. Itajaí-Açu River; 5. Ribeira de Iguape River; 6. Paraíba do Sul River. At the right, the Ilha Comprida barrier and the rocks drained by the Ribeira de Iguape River. The black squares show the location of the source rock samples.



**Fig. 2.** (A) Geomorphology of the Ilha Comprida barrier in aerial photographs from 1962. (B) Sampling sites for OSL sensitivity measurements. OSL sensitivities of quartz single-grains were measured in samples identified in the map by ICL1, ICL1E, ICL3A, ICL3D and ICL9A. The shaded areas differentiate the outer zone with eolian ridges (foredunes and blowouts) from the inner zone with beach ridges. (C) Sands with plan-parallel and low-angle cross stratification from beach ridges at sampling site 2. (D) Foredunes and blowouts at sampling site 6.

weather conditions throughout the southern Brazilian coast, turning to south and southeast winds and swell wave systems (Seluchi and Marengo, 2000; Rodrigues et al., 2004). These two swell wave systems generate alongshore currents with opposite directions (Fig. 1). Yet, northeastward litoral drift dominated the barrier growth during the Holocene as indicated by heavy minerals indexes, grain size parameters and sandy ridge morphology (Tessler, 1988; Nascimento Jr. et al., 2008; Guedes et al., 2011a). OSL ages recently obtained by Guedes et al. (2011a) indicate that the Ilha Comprida barrier progradation began about  $6041 \pm 504$  years ago. Wind conditions able to build eolian dunes appeared only in the late stage of barrier progradation, around  $575 \pm 47$  years before AD 2006 (Sawakuchi et al., 2008). The main eolian forms comprise foredunes and associated blowouts with lobes to NNW. Both forms developed in the seaward backshore zone, with short eolian transport of sands from the foreshore. A small dunefield with irregular barchanoid chains migrating to NNW evolved in the northern portion of the barrier. OSL ages indicate that the development of eolian dunes by winds from south was episodic and followed an event of coastal erosion as indicated by beach ridges truncation. This points to a transient morphodynamic shift of the barrier, which is attributed to the intensification of cold fronts since the Little Ice Age (Sawakuchi et al., 2008). This is in agreement with the northward expansion of the southern westerly wind belt in the

Holocene (Lamy et al., 2010), since the westerlies stimulate the northward advance of polar air masses.

The drainage system of southern South America comprises the La Plata River and minor coastal rivers (Fig. 1). The inland area of the Ilha Comprida barrier is part of the Ribeira Belt (Campanha and Sadowski, 1999; Faleiros et al., 2011), the major geological province of the onshore zone bordering the southeastern Brazilian coast. The Ribeira Belt comprises low to high grade metamorphic rocks, such as schists, mylonites, orthogneisses, paragneisses and granulites, and subordinate granites. Hydrothermal quartz veins occur within all these rocks. The Ribeira de Iguape River is the major watershed inland from the Ilha Comprida barrier. It has an area of approximately 25,400 km<sup>2</sup> and an annual average water flow of 375 m<sup>3</sup>/s at its mouth (DAEE, 1998). Thus, sediments derived from the Ribeira Belt reach the northern end of the Ilha Comprida barrier through the Ribeira de Iguape River. The Ilha Comprida sediments are well-sorted very fine to fine sands with a high content of quartz (>90%). These sands have a heavy minerals assemblage with elevated proportions of amphibole, pyroxene, epidote, sillimanite, kyanite, staurolite, garnet, zircon and rutile (Nascimento Jr. et al., 2008; Guedes et al., 2011b), pointing to a primary provenance compatible with the rock assemblage outcropping in the adjacent onshore zone. Yet, variations in the RZi (rutile to zircon ratio) heavy minerals index imply changes in sand provenance during the Ilha

Comprida barrier evolution (Guedes et al., 2011b). On the south side of the Ilha Comprida barrier, the Itajaí-Açu River and its tributaries are the major watershed. This watershed has an area of approximately 15,500 km<sup>2</sup> (Schettini, 2002) and drains rocks of the Luis Alves Terrane, which is a small cratonic remnant that occurs south of the Ribeira Belt (Faleiros et al., 2011). These rocks are composed of Archean–Paleoproterozoic orthopyroxene-bearing mafic to leucocratic orthogneisses, dominantly of intermediate composition (metaenderbite, metaopdalite, metaleuconorite), metamorphosed under granulite facies conditions (Basei et al., 1998; Hartmann et al., 2000). Minor quartzite and banded iron formation lenses are also present. The Itajaí-Açu River discharges in the Atlantic Ocean at around 230 km southward of the Ilha Comprida barrier. The coastal zone between the Itajaí and Ribeira de Iguape rivers comprises only minor watersheds and the Babitonga, Guaratuba and Paranaguá bays, which act as hydraulic obstacles for the alongshore bedload transport.

### 3. Material and methods

#### 3.1. Experiment and sample preparation

For this study, OSL single-grain measurements were especially valuable for investigating the variability of OSL sensitivity within and between sediments and source rocks. This technique allows us to characterize differences in sensitivity among grains of the same sedimentary bed or source rock, allowing a better evaluation of the effect of sedimentary transport on sensitivity through the comparison between sediments and source rocks. Two sediment samples from beach ridges (ICL1 and ICL3A) and three sediment samples from foredune ridges (ICL1E, ICL3D and ICL9A) of the Ilha Comprida barrier were prepared for OSL sensitivity measurements in quartz single-grains. These samples were selected to be representative of the sedimentary facies and position within barrier length. OSL sensitivity data from seven major types of source rocks that outcrop inland from the barrier were used as a reference for the sensitivity of quartz before its release to sedimentary systems. The analyzed source rocks comprised hydrothermal quartz vein (VR3V), granite (VR12R), orthogneiss (VR5RO), paragneiss (VR4R), kyanite–garnet–muscovite–quartz schist (VR7R), migmatitic schist (VR5RP) and sericite–chlorite schist (VR13RF). The luminescence characteristics and the geological context of the samples used for single-grain measurements were previously presented in Sawakuchi et al. (2011). Figs. 1 and 2 show the location of sediments and source rocks samples used for quartz single-grain measurements.

Thirty-six sediment samples collected from twenty-two pits or natural outcrops (Fig. 2) in the Ilha Comprida barrier were prepared for OSL sensitivity measurements in multigrain aliquots. The aliquots used for multigrain measurements in samples ICL1, ICLE, ICL3A, ICL3D and ICL9A were different from that used in single-grain measurements to avoid sensitivity changes due to laboratory procedures. All sediment samples represent well-sorted fine sands from beach and stabilized or active dune ridges. These samples were previously dated using the Single-Aliquot Regenerative protocol (Murray and Wintle, 2000; Wintle and Murray, 2006), with the results presented in Sawakuchi et al. (2008) and Guedes et al. (2011a). The OSL sensitivity measurements presented in this study were performed in aliquots different from that used in these dating studies. The OSL permitted evaluation of changes in the luminescence sensitivity of sediments accumulated in the barrier through time.

Firstly, crushed rock samples and sediment samples were wet sieved. Quartz separates with 180–250 µm grain size for single-grains and 120–150 µm grain size for multigrain aliquots were prepared by treatment with 27% H<sub>2</sub>O<sub>2</sub>, 3.75% HCl and 48–51% HF

for 40 min. Then, density separation in sodium polytungstate solution with densities of 2.75 g/cm<sup>3</sup> and 2.62 g/cm<sup>3</sup> were performed for removal of heavy minerals and remaining feldspars, respectively. The weight percent of heavy minerals was measured in the very fine sand fraction (0.0625–0.125 mm) through heavy liquid separation (bromoform, CHBr<sub>3</sub>, 2.85 g/cm<sup>3</sup>) as described in Guedes et al. (2011b). Heavy minerals concentration was measured in subsamples that were not submitted to chemical treatments.

Single-grains of quartz were deposited on aluminum discs with a 10 × 10 grid of pits that were 300 µm deep with a 300 µm diameter. Three single-grain discs per sample were prepared. The grains within each pit were checked under a stereoscopic microscope and only pits with pure quartz grains were used in the analysis. Empty pits and pits with non-quartz grains were excluded from the data analysis. This procedure ensured that all analyzed OSL signals were provided by single-grains of quartz. 121–268 grains per rock sample and 190–291 grains per sediment sample were analyzed.

For the multigrain aliquots of quartz from sediments, subsamples of the same volume comprising about 400 grains were measured with a micro spoon and fixed to stainless steel cups using Silkospray. Four quartz aliquots, totalizing around 1600 grains, were used to determine the OSL sensitivity of each sediment sample.

#### 3.2. Single-grain measurements

Single-grain measurements were carried out on a Risø TL/OSL DA-15 system with single-grain attachment in the Los Alamos National Laboratory. A preliminary assessment of the OSL sensitivity of quartz single-grains from these samples is discussed in Sawakuchi et al. (2011). The Risø reader used for single-grain analysis is equipped with a 532 nm green laser, nominally 50 W/cm<sup>2</sup> at the sample position, for optical stimulation of grains and a built-in beta source with a dose rate of 0.0139 Gy/s. The OSL signal was detected using a Hoya U-340 filter with transmission wavelength between 290 and 370 nm. Initially, each quartz grain was bleached at 125 °C (5 °C/s) for 1 s with 90% laser power. Then, Linear Modulated OSL (LM-OSL) curves were acquired for 10 s of stimulation, with a resolution of 0.2 s per data point, after 100 Gy of beta radiation. A relatively high radiation dose was applied to improve the signal of low sensitivity grains and facilitate the discrimination among grains with different sensitivities, and a pre-heat was not used to avoid laboratory-induced changes in sensitivity.

LM-OSL measurements were used to identify the component controlling the luminescence signal of the studied quartz grains. The OSL sensitivity of each quartz grain was obtained by integrating the total LM-OSL curve. The background was obtained from the LM-OSL curves of empty disc positions, which yielded mean integral of 564 counts and standard deviation of 41 counts. The value of 687 counts that corresponds to the mean background plus three standard deviations was used to correct the LM-OSL curves and to identify dim and low sensitivity grains. Grains with OSL signals smaller than 687 counts were considered as dim grains since they cannot be discriminated from the background. Grains with signals less than 1374 counts, which correspond to two times the background, were classified as low sensitivity grains. The proportions of dim and low sensitivity were assessed for sediments and source rocks samples.

#### 3.3. Multigrain aliquots measurements

The multigrain aliquots of quartz were measured using an automated Risø TL/OSL DA-15 system equipped with a bialkali PM tube (Thorn EMI 9635QB) and Hoya U-340 filters (290–370 nm) in the Radiation Dosimetry Laboratory at Oklahoma State University.

The built-in  $^{90}\text{Sr}/^{90}\text{Y}$  beta source gives a dose rate of  $0.0996 \pm 0.0041$  Gy/s. The quartz aliquots were bleached through Continuous Wave (CW) stimulation at  $125^\circ\text{C}$  ( $5^\circ\text{C}/\text{s}$ ) for 1000 s with blue LEDs (470 nm). Afterwards, optical stimulation was carried out at  $125^\circ\text{C}$  ( $5^\circ\text{C}/\text{s}$ ) during 100 s, with blue LEDs delivering  $31\text{ mW}/\text{cm}^2$  to the sample. A pre-heat was not performed to keep the OSL sensitivity as natural as possible, in the same way adopted in the single-grain measurements. Stimulation was carried out immediately after the irradiation of each aliquot, minimizing the loss of unstable OSL components. CW-OSL was measured instead of LM-OSL since future protocol development for sensitivity measurements will probably use data from routine dating protocols. A feldspar test with infrared stimulation performed on all samples ensured the effectiveness of quartz separation. The OSL sensitivity was obtained by integrating the first two seconds of the OSL decay curve while the background was determined by integrating the last ten seconds. The average OSL sensitivity of the four aliquots of each sample was evaluated in relation to its deposition age and was used to build interpolation maps using the inverse distance weighted method (Shepard, 1968). The OSL sensitivity errors were calculated through the Gaussian law of error propagation.

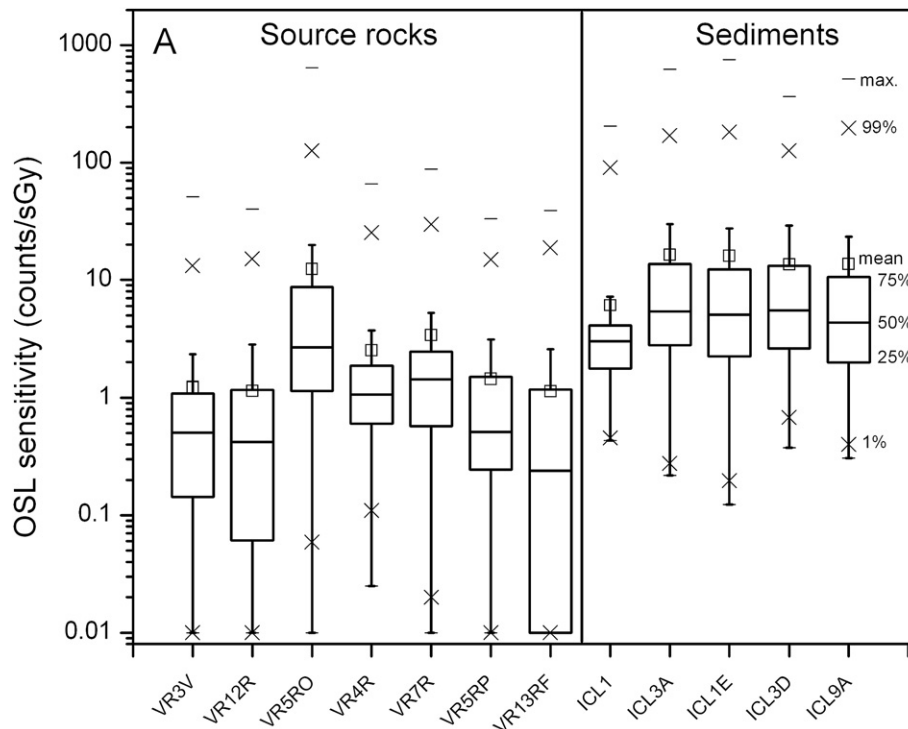
## 4. Results

### 4.1. Sensitivity of quartz single-grains

The OSL sensitivities of quartz single-grains extracted from the Ilha Comprida sands and inland source rocks have frequency distributions with positive asymmetry that vary by five orders of magnitude, ranging from  $10^{-2}$  to  $10^3$  counts/sGy. Despite the high variability within samples, the percentiles of quartz sensitivity from the Ilha Comprida barrier sediments are approximately one order of magnitude higher than the percentiles of source rocks

(Fig. 3). Orthogneiss (VR5RO) is the only source rock with high sensitivity similar to the sensitivity of sediments. However, this relatively high sensitivity source rock is diluted among all other low sensitivity source rocks. Thus, the comparison between total assemblages of quartz single-grains from source rocks and sediments shows that the sensitivity of sediments is significantly higher than the sensitivity of source rocks. Regarding the sediment samples, no significant difference was observed in the OSL sensitivity of wave- and wind-deposited sands.

Despite the higher sensitivity, the Ilha Comprida sands comprise a mixture of relatively dim and bright grains. The percentages of low sensitivity grains within the Ilha Comprida sands vary from 1% (ICL3D) to 6% (ICL1E). Dim grains were not observed. Bright grains with sensitivity higher than 100 counts/sGy represent less than three percent of the grains but can have a significant contribution to the sensitivity of multigrain aliquots. Fig. 4 shows the distributions of light output among quartz single-grains from source rocks and sands. Around 10% of the quartz grains from the Ilha Comprida sands contribute approximately 50–60% of the total luminescence. Similar relationships were observed for the source rocks, where 10% of the grains produce 55–65% of the total light output. Thus, a small proportion of quartz grains is responsible for a significant part of the luminescence signal of the studied sands and rocks, being in agreement with observations of previous studies (Duller et al., 2000; Murray and Wintle, 2002; Rhodes, 2007). Duller et al. (2000) demonstrated that only 5% of quartz grains in sands from coastal dunes in southern Africa provide 95% of the total light sum. In the studied sediment samples, 50% of the grains give approximately 85–90% of the luminescence. For the source rocks, 50% of the grains generate around 90–100% of the luminescence, indicating a higher proportion of dim or low sensitivity grains. The proportion of dim grains in source rocks was from 0 to 34%, while low sensitivity grains represent from 13% to 66% of the measured



**Fig. 3.** Boxplots of OSL sensitivities of quartz single-grains from the Ilha Comprida barrier sediments and their main source rocks types. Source rock types are represented by hydrothermal quartz vein (VR3V), granite (VR12R), orthogneiss (VR5RO), paragneiss (VR4R), kyanite–garnet–muscovite–quartz schist (VR7R), migmatitic schist (VR5RP) and sericite–chlorite schist (VR13RF). Samples ICL1 and ICL3A correspond to wave-deposited sands (beach ridges) and samples ICL1E, ICL3D and ICL9A are from wind-deposited sediments (foredunes and blowouts). The number of quartz grains of each sample is presented in Table 2.

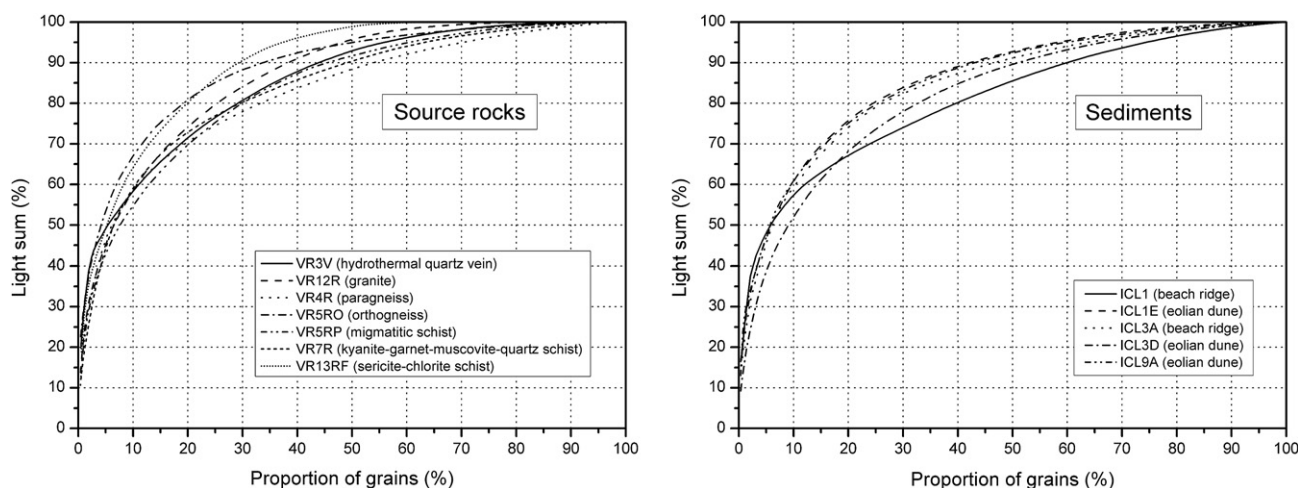


Fig. 4. Percentage of light sum originated from the proportion of decreasing bright grains from sediments and source rocks.

grains. Table 1 summarizes the proportion of dim and low sensitivity quartz grains in the studied sediments and source rocks.

#### 4.2. Sensitivity of quartz multigrain aliquots

The mean OSL sensitivities of the multigrain aliquots reflect a relatively high variation within the barrier, ranging from 7676 to 20,130 counts/sGy, with errors from 2 to 10% of the mean. The OSL sensitivities of eolian and wave-deposited sands are similar (Fig. 5), even though averaging effects can mask subtle differences. Samples with higher mean sensitivities also display a higher mean heavy minerals concentration (Fig. 6). Despite the significant variation of OSL sensitivity within samples, we observe a decreasing tendency in sensitivity toward the northern and younger portion of the barrier (Fig. 7). As a consequence, the spatial variation in sensitivity is also temporal. Table 2 summarizes the OSL sensitivities, heavy minerals contents and deposition ages of the studied sediment samples. The evaluation of sensitivity regarding the age of sediment deposition shows a shift at 2 ka from a pattern of relatively stable sensitivity to highly varied sensitivity. Cyclic and high amplitude changes in the mean sensitivity are observed from 2 ka to present. An abrupt change in sensitivity is observed at around AD 1450, which corresponds to the transition between the Medieval Climate Anomaly (MCA) and the Little Ice Age (LIA). Both changes are also observed in the concentration of heavy minerals (Fig. 7).

## 5. Discussion

### 5.1. Natural controls on quartz OSL sensitivity

The quartz luminescence depends on the type and content of point defects formed by vacancies of oxygen or silicon ions and by the incorporation of impurities such as aluminum and titanium (Preusser et al., 2009). The formation of point defects and their effectiveness as luminescence centers is regulated by many geological factors. There is no consensus regarding the most important factor controlling the OSL sensitivity of quartz in nature. Studies attempting to interpret a provenance signal through quartz TL or OSL have been attributed the observed luminescence to three main factors: 1) the composition of fluids and melts from which quartz crystallizes (Rink et al., 1993). This defines the availability of impurities to be incorporated by quartz during crystallization; 2) the temperature of quartz formation and posterior heating events (Bøtter-Jensen et al., 1995; Poolton et al., 2000; Polymeris et al., 2007; Sawakuchi et al., 2011). Higher temperatures favors the incorporation of impurities as well as the formation of intrinsic defects such as oxygen vacancies; 3) the irradiation history, including the irradiation during the lifetime of quartz in the parent rocks (Tsukamoto et al., 2011) and cycles of irradiation and bleaching during the sediment transport (Pietsch et al., 2008), both cases would activate non-luminescent centers as well as generate new ones. In summary, the OSL sensitivity depends on factors

Table 1  
Percentage of dim and low sensitivity grains in the studied source rocks and sediments.

Sample type	Sample ID	Sample description	Number of grains	% of dim grains	% of low sensitivity grains
Source rocks	VR03R	Hydrothermal quartz vein	210	7	58
	VR04R	Granite	246	0	33
	VR05RO	Paragneiss	229	1	13
	VR05RP	Orthogneiss	121	3	57
	VR07R	Migmatitic schist	215	1	31
	VR12R	Kyanite–garnet–muscovite–quartz schist	255	16	61
	VR13RF	Sericite–chlorite schist	182	34	66
Sediments	ICL1	Beach ridge	190	0	2
	ICL1E	Eolian dune	285	0	6
	ICL3A	Beach ridge	284	0	3
	ICL3D	Eolian dune	291	0	1
	ICL9A	Eolian dune	291	0	4

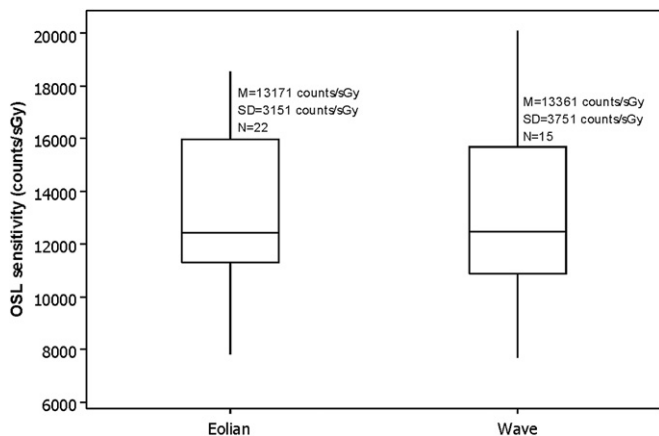


Fig. 5. OSL sensitivities measured in multigrain aliquots of quartz from the Ilha Comprida barrier sands. There is no significant difference between the OSL sensitivities measured in wind- and wave-deposited sands. M = mean, SD = standard deviation, N = number of samples.

acting in the primary parent rocks (igneous and metamorphic rocks) before their weathering and release of quartz grains to surface systems and factors acting in quartz grains during their transport in sedimentary systems. The key point in order to use the OSL sensitivity as an indicator of depositional history of quartz is to evaluate if the sensitivity increase due to sediment transport can be discriminated from the sensitivity inherited from parent rocks.

The primary origin of quartz has been evoked to explain the variability of quartz OSL sensitivity observed in sediments from New Zealand (Preusser et al., 2006) and China (Zheng et al., 2009). According to Westaway (2009), the OSL emissions relate with the quartz origin, which would influence the OSL sensitivity detected in specific wavelengths. The number of cycles of irradiation during temporary burial and subsequent bleaching by solar exposure when sediment is transported increases the OSL sensitivity of

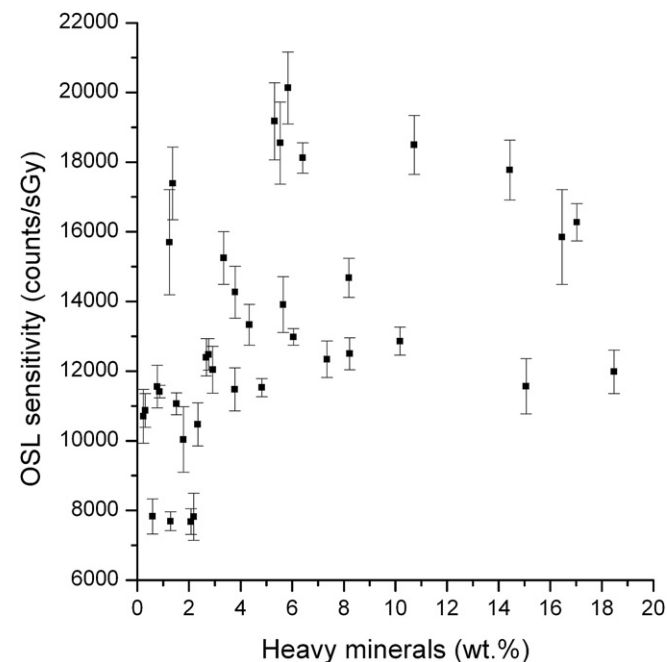


Fig. 6. OSL sensitivities of multigrain aliquots in relation to heavy minerals concentration.

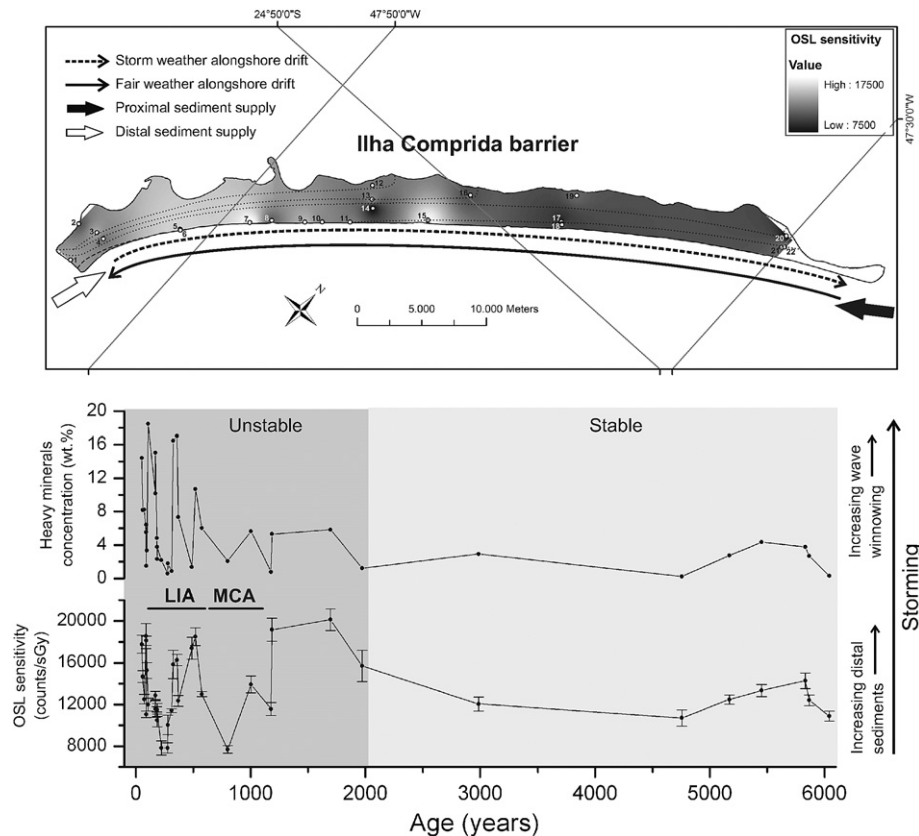
quartz. This conclusion was reached by Pietsch et al. (2008) through the study of the OSL sensitivity of quartz grains from the Castlereagh River that drains an area dominated by few rock types (volcanic and sedimentary rocks), minimizing the effect of quartz origin on the OSL sensitivity. In this setting, dim grains represented 75% of these grains while grains with initial detectable luminescent signal comprised 25% of the grains. The downstream increase in sensitivity in the Castlereagh River is represented by the conversion of dim to bright grains as well as by an increase in the sensitivity of grains with initial detectable luminescent signal. Only 10% of the grains do not increase their luminescence sensitivity while the sensitivity of 62% of the initial luminescent grains (around 15% of all grains) increased linearly by two orders of magnitude until reaching a limit and becoming the brightest grains.

Igneous and metamorphic rocks rich in quartz crystallize form melts with high contents of Al and Ti, which are the main impurities substituting for Si in quartz crystal lattice to form extrinsic point defects. Thus, the increase in quartz crystallization temperature that favors the incorporation of impurities would be the most important factor acting on parent rocks. It has been demonstrated that the increase in sensitivity due to sedimentary cycles can exceed the sensitivity inherited from parent rocks (Pietsch et al., 2008) even in sediments derived from multiple lithologies (Sawakuchi et al., 2011). Exceptions should be made to sedimentary parent rocks, whose quartz grains will have a sensitivity inherited from previous sedimentary cycles (Fitzsimmons, 2011), and to sediments eventually heated by contact with lava flows or by fault movement. These possibilities should be addressed before the interpretation of the OSL sensitivity signal of quartz from sediments.

The mode of sediment transport, which determines the extent of sunlight exposure and possibly influences the increase in the OSL sensitivity of quartz, also has been evoked to explain the OSL sensitivity of quartz grains (Li and Wintle, 1992). However, Fitzsimmons (2011) concluded that the nature of sunlight exposure during sediment transport is less important than source lithology for the OSL sensitization of quartz. Yet, it is important to note that the source lithology is represented by sandstones whose quartz grains were submitted to previous sedimentary cycles before their return to active sedimentary systems.

Regarding the Ilha Comprida barrier, our data indicate that the type of sediment deposition does not affect significantly the OSL sensitivity of quartz. There is no significant difference between the sensitivities of wind- and wave-deposited sands, which is compatible with the short transport from the foreshore zone to the foredunes and blowouts from the backshore zone.

For provenance purposes, the quartz grains can be classified into two end members: 1. Grains with a short depositional history whose sensitivity is still dominated by sensitivity inherited from parent rocks; 2. Grains with long depositional history with sensitivity surpassing that inherited from parent rocks. The “short” and “long” sedimentary histories are relative definitions that will depend on the sensitivity inherited from parent rocks and capacity of the sedimentary system to increase the sensitivity. The Ilha Comprida barrier is supplied by sands with contrasting depositional histories, which are suitable for the discrimination using the quartz OSL sensitivity. Our single-grain data indicate that the top 30% of the grains in terms of brightness are responsible for more than 70% of the total luminescence of the Ilha Comprida sands. These brightest grains control the observed variation of OSL sensitivity of the Ilha Comprida barrier through time and therefore should more reliably record the relative contributions of the distal and proximal sediment sources.



**Fig. 7.** At the top, variation in OSL sensitivities from multigrain aliquots within the Ilha Comprida barrier. Sites with multiple samples are represented by the mean sensitivity. The arrows indicate the main sediment sources and directions of alongshore transport. The dotted lines show the main phases of barrier growth. At the base, variation in OSL sensitivities and heavy minerals concentration in relation to ages of sediment deposition. OSL ages by Sawakuchi et al. (2008) and Guedes et al. (2011a). Heavy minerals concentration by Guedes et al. (2011b). The shaded areas differentiate stable from unstable periods regarding wave climate. The time intervals of the Little Ice Age (LIA) and Medieval Climate Anomaly (MCA) are marked by the black bars.

### 5.2. The provenance signal of quartz OSL sensitivity in the Ilha Comprida barrier and its use as a proxy for storm activity

Dating studies have demonstrated that the Ilha Comprida barrier sands are significantly bright being since even small equivalent doses around  $0.040 \pm 0.005$  Gy can be recovered (Sawakuchi et al., 2008). As it has been interpreted for the bright Australian sands (Fitzsimmons et al., 2010), the high sensitivity of the Ilha Comprida sands is due to their long sedimentary history. However, quartz single-grains from the Ilha Comprida barrier sands have sensitivity varying in four orders of magnitude, with the sensitivity of multigrain aliquots differing by a factor of 3. Thus, the Ilha Comprida sands would comprise a mixture of grains with different sedimentary histories and their high sensitivity is due to the presence of a significant content of bright grains. The sum sensitivity of quartz single-grains from these sands is significantly higher than the sensitivity of quartz from their primary source rocks, as would be expected due to the presence of grains with long depositional history. Single-grain data show that the sensitivity distributions of quartz single-grains from sediments and source rocks are positive skewed, suggesting a power law relationship similar to that observed by Rhodes (2007) in eolian and fluvial sands from Australia. This points out that the sedimentary reworking increases the sensitivity but keep the luminescence distribution inherited from source rocks.

In summary, single-grain data indicates that the Ilha Comprida barrier sands are made of a mixture of grains with contrasting depositional histories since their crystallization and weathering out

from primary parent rocks to sedimentary systems. The sediment transport in the Ilha Comprida barrier is controlled by a drift system of alternating alongshore currents to southwest during fair weather times that shift to northeast during storm events. Geomorphological and sedimentological features indicate that the storm drift to northeast is more effective for sediment remobilization and deposition (Suguio and Martin, 1978; Giannini et al., 2009; Guedes et al., 2011a). The Holocene barriers of southern Brazil comprise distal sediments with high coastal sedimentary reworking as well as proximal sediments with short sedimentary history delivered to the coastal barriers directly by river catchments (Sawakuchi et al., 2009). Sands with long sedimentary history can also derive from offshore zones in the outer shelf (Viana et al., 1998a). Storm waves on the Brazilian shelf can rework sediments in water depth around 100 m (Viana et al., 1998b), making possible the transport of sands from the outer shelf to the shoreface and foreshore zones. For the Ilha Comprida barrier, proximal sediments are supplied by the Ribeira de Iguape River, the greatest drainage basin in the hinterland of the studied coast. The Ribeira de Iguape river mouth reaches the South Atlantic at the northern end of the Ilha Comprida barrier. Its sands are delivered to the Ilha Comprida barrier by the alongshore fair weather drift to southwest. These fluvial sands have a relatively short depositional history because they are derived mainly from the weathering of igneous and metamorphic rocks. On the other hand, storm currents to northeast allow that distal sediments from southern coastal sectors and/or from the outer shelf reach the barrier. Sediments from the south can be transported for hundreds of kilometers of coastal segments, suffering multiple



**Table 2**

Mean OSL sensitivities measured in multigrain aliquots and their ages of deposition (OSL-SAR) as determined by Sawakuchi et al. (2008) (\*) and Guedes et al. (2011a) (+). Heavy minerals concentration from Guedes et al. (2011b).

Sampling site	Sample code	Mean sensitivity (counts/sGy)	Heavy minerals (wt.%)	OSL-SAR age (years)
1	IC-1-CI <sup>+</sup>	11,555 ± 617	0.77	1179 ± 92
2	IC-3-FML-2 <sup>+</sup>	10,874 ± 484	0.30	6041 ± 503
3	IC-4-CI-5 <sup>+</sup>	14,267 ± 742	3.78	5831 ± 432
4	IC-4-CI-7 <sup>+</sup>	12,044 ± 675	2.92	2986 ± 221
5	ICL1*	13,910 ± 804	5.65	1004 ± 88
6	ICL1A*	15,849 ± 1365	16.46	325 ± 31
	ICL1B*	11,984 ± 626	18.47	108 ± 10
	ICL1C*	15,252 ± 756	3.34	97 ± 11
	ICL1D*	18,124 ± 435	6.41	91 ± 9
	ICL1E*	18,553 ± 1180	5.53	89 ± 11
7	ICL3A*	20,130 ± 1034	5.83	1697 ± 159
	ICL3B*	12,341 ± 524	7.35	370 ± 60
	ICL3C*	16,275 ± 534	17.02	360 ± 31
	ICL3D*	17,774 ± 860	14.43	53 ± 8
8	ICL2A*	19,176 ± 1106	5.32	1186 ± 98
	ICL2B*	11,528 ± 259	4.82	184 ± 25
9	ICL4A*	18,493 ± 851	10.72	520 ± 38
	ICL4B*	12,856 ± 403	10.17	172 ± 17
	ICL4C*	11,564 ± 790	15.06	172 ± 18
10	ICL4D*	14,679 ± 562	8.20	60 ± 8
	ICL6A*	12,979 ± 240	6.05	575 ± 47
11	ICL7A*	11,474 ± 618	3.78	188 ± 16
	ICL7B*	12,502 ± 462	8.23	74 ± 16
12	IC-24-CI <sup>+</sup>	12,397 ± 534	2.67	5866 ± 425
13	IC-24-CI-1 <sup>+</sup>	13,331 ± 586	4.34	5451 ± 487
14	IC-24-CI-CI-3 <sup>+</sup>	7676 ± 368	2.08	801 ± 68
15	ICL5*	17,391 ± 1041	1.37	489 ± 48
16	IC-31-CI <sup>+</sup>	12,478 ± 451	2.76	5169 ± 422
17	ICL8A*	7828 ± 503	0.59	278 ± 22
18	ICL8B*	10,474 ± 619	2.34	187 ± 17
	ICL8C*	11,062 ± 313	1.52	91 ± 12
19	IC-39-CI <sup>+</sup>	10,703 ± 775	0.23	4755 ± 398
20	IC-55-CI-1 <sup>+</sup>	15,700 ± 1514	1.24	1971 ± 154
21	ICL9A*	11,412 ± 180	0.86	313 ± 26
22	ICL9B*	10,037 ± 938	1.78	279 ± 22
	ICL9C*	7821 ± 675	2.18	222 ± 23

cycles of transport and temporary storage in the shelf and coastal plains until reach the Ilha Comprida barrier. This northward long distance sediment transport in the southern Brazilian coast is supported by stratigraphical (Lessa et al., 2000), isotope (Mahiques et al., 2008) and mineralogical (Sawakuchi et al., 2009) data. Thus, sediments from south are transported for longer distances in wave-dominated coastal systems until their deposition in the Ilha Comprida barrier. Sediments from north are transported for shorter distances, with this transport being dominated by fluvial processes until their deposition in the barrier. The mixture of these sands with different depositional histories dictates the variability of the OSL sensitivity in the barrier. The multigrain aliquots in the Ilha Comprida barrier show a higher OSL sensitivity when heavy minerals concentration gets higher. Heavy minerals concentration in coastal sand barriers has been related to storm events (Woolsey et al., 1975; Roy, 1999; Buynevich et al., 2004; Dougherty et al., 2004). Thus, the relationship between heavy minerals concentration and OSL sensitivity in the Ilha Comprida sands suggests that the supply of brighter sands is related to storm currents. As a result, the OSL sensitivity of the Ilha Comprida barrier sands would be a proxy for storming in the southern Brazilian coast.

The variation of OSL sensitivity through time shows a striking shift in sand provenance at around 2 ka ago. The time from 6 to 2 ka is characterized by the input of sands with mean sensitivity from 10,000 to 12,000 counts/sGy, which is followed by a period with high variation in sand provenance as indicated by sensitivity varying from 7500 to 20,000 counts/sGy. This shift in sand

provenance would represent a transient change from a relatively stable dynamics to a condition of submillennial high variability in the frequency and/or intensity of storms. Abrupt increase in storm activity would be occurred during the transition from the Medieval Climate Anomaly (MCA) to the Little Ice Age (LIA) at around AD 1500 and after the LIA at around AD 1850. Currently, storm patterns along the southern Brazilian coast are influenced by the northward advance of polar air masses. Increased activity of polar air masses since 2 ka ago correlates with the strengthening and/or northward expansion of the westerlies wind belt in southern South America (Gilli et al., 2005; Lamy et al., 2010), suggesting that the provenance variation in the Ilha Comprida barrier results from regional sub-millennial paleoclimate changes.

## 6. Conclusions

The high variation in quartz OSL sensitivity of the Ilha Comprida barrier sands results from the mixture of sediments with different depositional histories, corresponding to the mixture of two end members regarding the sensitivity of quartz grains. Low sensitivity quartz grains would be supplied by the Ribeira de Iguape River, which are transported to the barrier by fair weather southward alongshore currents. The shift to northward alongshore currents during storm conditions permits the input of high sensitivity quartz grains from distal southern coastal sectors and/or from offshore zones. Thus, the OSL sensitivity of quartz grains records the storm activity in the Ilha Comprida barrier.

The OSL sensitivity of the Ilha Comprida barrier displays a relatively short variation from 6 ka to 2 ka. Huge variation in the sensitivity occurs from 2 ka to present. This shift in the OSL sensitivity records the onset from a relative stable coast to an unstable condition characterized by more frequent and/or intense storms. The increase in storm patterns at 2 ka is consistent with other regional paleoclimate changes in South America such as the strengthening of the westerly wind belt since the Middle Holocene. In the South Atlantic, the westerlies drive the activity of polar air masses whose migration northward and encounter with tropical air masses generate storm conditions in the Brazilian coastal zone.

The relationship between OSL sensitivity and the sedimentary history of quartz is a promising tool for the provenance analysis of sediments, especially in contexts comprising the mixing between sediments with short and long depositional histories. The quartz OSL sensitivity coupled with luminescence dating is a valuable paleoclimate proxy for southern Brazilian barriers as well as for other sedimentary systems whose sediment provenance is climate dependent.

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