METHODS

Prior to the grain size, bulk mineralogical, and geochemical analyses, samples (<2 mm) were pretreated with 10 ml of hydrogen peroxide (H₂O₂; 30%) and 10 ml of hydrochloric acid (HCl; 0.5 N) for at least 72 hours to remove organic matter and biogenic carbonate. Next, an aliquot of this sediment sample was ground using a micronizing mill (McCrone) and 5 ml of ethanol for approximatively 10 minutes until a homogeneous mixture of grain <10 µm was obtained. The mixture was oven-dried at 60°C and ground again in an agate mortar to prevent aggregations of fine particles due to drying. Aliquots of these homogenized sediment samples were used for bulk mineralogical and geochemical analyses.

The observations of tephra using a JEOL 6460LV SEM were performed on the 300–63 μ m and <63 μ m fractions of the BV01, BV06, and BC11 samples. Before sieving, the samples were pretreated with 10 ml of hydrogen peroxide (H₂O₂; 30%) for 24 hours and rinsed with distilled water at least five times. Finally, the samples were oven-dried at 60°C.

GRAIN-SIZE ANALYSIS

The grain-size analysis of sediment samples was carried out on detrital fraction using a Beckman Coulter Particle Size Analyzer LS 13 320 (0.04-2,000 µm). Deflocculation was performed by successive washing with distilled water and the samples were mechanically shaken for 12 hours before measurement. Grain-size distribution and statistical parameters were processed with the GRADISTAT software using the geometric (µm) and logarithmic (φ) method of moments (Blott and Pye, 2001). The end-member modeling algorithm (EMMA; Dietze et al., 2012) was applied to the grain-size data. The EMMA algorithm is used to unmix the original grain-size distribution (Figure 2b) in order to extract meaningful EM grain-size distributions and thus to estimate the processes (transport conditions) related to the EM grain-size distributions (Dietze et al., 2012). The characteristics and detailed procedures of the EMMA method used for this study are presented in Dietze et al. (2012) and Dietze and Dietze (2013).

SOURCE SAMPLES

In order to quantify potential sources of sediment in the GSJ, 13 terrestrial and riverine samples were retrieved in August 2016 (Figure 1a,b); nine of them were collected on the shore of the GSJ (topsoil and beach sediments), two at the mouths of the Chubut and Deseado Rivers (river bank and beach sediments, respectively), and two in the Bajo de Sarmiento area (topsoil sediments from the dry bed of Lake Colhué Huapí [NS1] and 10 km to the east of the lake, close to dune fields formed by the southern westerlies [NS2; Montes et al., 2017]). A marine surface sediment sample (0-2 cm sediment depth) from Magellan Strait was also analyzed. The latter was recovered with a CASQ gravity corer (MD07-3131) during the MD 159 PACHIDERME expedition on board R/V Marion Dufresne II (Figure 1a; February 2007).

BULK MINERAL ANALYSIS

The random powder samples were sideloaded into holders and analyzed by X-ray diffraction (XRD) using a PANalytical X'Pert Powder diffractometer. The samples were measured between 5° and 65° 2θ in steps of 0.02° and a counting time of 2 seconds per step. Bulk mineral associations were analyzed following the quantitative method developed by Eberl (2003) and Eberl and Smith (2009) and used in other Late Quaternary marine studies that deal with sediment mineralogy (e.g., Andrews et al., 2015, 2016; Andrews and Vogt 2014). For the quantification of the major mineralogical components, the XRD scans were converted into mineral

weight percentages (wt.%) using the standardless option of the Excel macro-program Rockjock v11 (Eberl, 2003; Eberl and Smith, 2009). This program uses a full-pattern fitting method that permits the quantification of the whole-sediment mineralogy with an error of approximatively ±3 wt% at 50 wt% of a mineral (Eberl, 2003). To verify the quality of this fitting procedure, a degree-of-fit (DOF = minimum absolute difference) statistic was calculated between the measured and simulated XRD patterns. The DOF values obtained with our samples were satisfying. They were within a range of 0.072 to 0.442 with an average of 0.165 for the 95 samples. Polytypes of illite, smectite, plagioclase feldspars, and K-feldspars are reported as a total amount (e.g., Andrews and Vogt, 2014; Andrews et al., 2015, 2016). This qXRD method came in second in the international "Reynolds Cup" quantitative mineral analysis competition (Omotoso et al., 2006). The calculated total mineral wt% was normalized to sum to 100%. We present the wt % data for 12 minerals (Table S2), but we focus on 11 minerals and exclude carbonates and biotite because of their restricted occurrence (Table S5).

Likewise, we used the nonlinear unmixing Excel macro program SedUnMixMC (Andrews and Eberl, 2012; Andrews et al., 2015, 2016) to obtain a quantitative understanding of the surface changes in sediment provenance. We ran SedUnMixMC on the normalized (100%) data for the 11 key minerals (Table S5) that represented more than 99% of the overall mineral concentration in the sediment samples. SedUnMixMC allows up to six sources to be examined as potential contributors to sediment composition. Based on surface geology (e.g., Pankhurst and Rapela, 1995; Pankhurst et al., 1998; see Physical Setting section in main text) and sediment transport pathways (e.g., Matano

et al., 2010; Montes et al., 2017; see Physical Setting and Sedimentation sections in main text), we suggest that the surface sediment compositions in the GSJ are potentially the mixing product of five source areas: (S1) Marine park sector (i.e., rhyolitic rocks outcrop), (S2) Other coastal areas of the GSJ (i.e., cliffs and beaches of Eocene-Miocene sedimentary rocks and/or Quaternary fluvio-glacial deposits), (S3) rivers (i.e., Chubut and Deseado, as well as Strait of Magellan), (S4) dust from continental Patagonia (i.e., Bajo de Sarmiento area), and (S5) continental shelf (i.e., southern South America influence and northward shelf Patagonian Current). Under this context, SedUnMix was run using five sources, and each source is represented by two to five of the source area samples (Table S5). The samples were attributed to potential sources according to their locations coupled with geological and environmental context, and also with the results from the mineralogical analysis (Figure 3).

CLAY MINERAL ANALYSIS

In this paper, clay minerals were quantified in the bulk sediment fraction (<2 mm) using the Excel macro program RockJock. However, nearly all previous clay-mineral provenance studies of the Argentinean continental shelf used oriented mounts of the <2 µm sediment fraction to identify and semi-quantify the clay-mineral abundance, notably illite, kaolinite, chlorite, and smectite (e.g., Petschick et al., 1996; Diekmann et al., 2000; Dominguez et al., 2008). Therefore, in this study, the clay-size fraction of all sediment samples was isolated and analyzed in this manner for comparison. Clay mineral associations were studied using XRD following established protocols (Bout-Roumazeilles et al., 1999). The clay-sized fraction (<2 µm) was isolated by settling according to Stoke's Law, concentrated by centrifugation, and oriented on glass slides. For each sample, analyses were performed on three subsamples under different conditions: (1) untreated sample (normal run); (2)

ethylene-glycol saturation applied for 12 hours (glycol run); and (3) sample heated at 490°C for two hours (heating run). The analyses were measured on a PANalytical X'Pert Powder diffractometer, between 2.49° and 32.49° 2 θ for the normal and glycol runs and between 2.49° and 14.5° 2θ for the heating run, with steps of 0.02° and a counting time of three seconds per step for all the runs. Semi-quantitative estimation of clay mineral abundances (smectite, illite, chlorite, kaolinite, and illite/smectite mixed layer) based on peak areas was performed using the MacDiff[®] 4.2.5 software (Petschick, 2000). The error on the reproducibility of measurements is estimated to be 5% for each clay mineral, as verified with analyses on replicate samples. Note that the comparison of both RockJock and oriented mounted methods yielded similar results within analytical uncertainty (Figure S5). This highlights the robustness of the qXRD method used in our study.

BULK ELEMENTAL GEOCHEMISTRY

Concentrations of 14 major and minor elements (Al₂O₃, SiO₂, K₂O, MgO, CaO, TiO₂, MnO, Fe₂O₃, P₂O₅, Sr, V, Cr, Zn, and Zr) were measured by energy dispersive X-ray fluorescence spectrometry (EDXRF) using a PANalytical Epsilon 3-XL. Before EDXRF analysis, loss on ignition (LOI) was determined gravimetrically by heating the dried samples up to 950°C for two hours. Subsequently, the glass disks were made by melting ~0.6 g of samples with a mixture of lithium tetraborate and metaborate (49.75% Li2B4O7, 49.75% LiBO2, and 0.5% LiBr, CLAISSE) in an automated fusion furnace (CLAISSE M4 Fluxer) prior to being analyzed with the spectrometer. Acquired XRF spectra were processed with the standardless Omnian software package (PANalytical). Analytical accuracy, based on both an international standard (USGS SDC-1) and analysis of replicate samples, was about 1%-5% for major elements and 5%–10% for the other elements.

Likewise, the elemental composition

analysis of filters and tephras was performed using an INCA X-sight energy dispersive X-ray spectrometer (Oxford Instruments) coupled to a JEOL 6460LV scanning electron microscope. X-ray spectra were measured from 15 randomly chosen particles for filters and 10 glass shard fragments for tephras, with two or three spectra per particle. Each spectrum was acquired for 60 (filters) and 120 (tephras) seconds of live time at an accelerating voltage of 20 kV. System quantitative optimization was made using copper as standard.

The relative proportions of the three most abundant detrital elements (Si, Al, and Fe) of the GSJ samples are presented in the form of a ternary diagram with respective mean grain sizes using the phiscale to determine the influence of grain size on major element composition. In addition, the ternary plot Fe-K-Ca was used here to understand the contributions of various Patagonian sedimentary sources to the elemental geochemistry of sediments in the GSJ (see Bulk Mineralogy section in the main text). In this diagram the GSJ samples (excluding marine park sites) are illustrated according to their locations in the gulf.

STATISTICAL APPROACH

The mineralogical and geochemical data are of a compositional nature, that is, they are vectors of non-negative values subjected to a constant-sum constraint (usually 100%). This implies that relevant information is contained in the relative magnitudes, so statistical analysis must focus on the ratios between components (Aitchison, 1986). Under this framework, the discriminant scatter plots based on mineralogical and geochemical data were represented here as log ratios. Note that a log transformation will reduce the very high values and spread out the small data values and is therefore well suited for right-skewed distributions (van den Boogaart and Tolosana-Delgado, 2013). Thus, compared to the raw data, the log-ratio scatter plots exhibit better sediment discrimination. Likewise, in order

to visualize the main tendencies of mineralogical enrichment in the gulf, the spatial distribution of the major mineral groups were represented as mineralogical balances (i.e., b{(Quartz+Feldspars)/Clays}). Note that mineralogical balances are log contrasts resulting from a log ratio of two geometric means of two non-overlapping mineralogical groups (Egozcue and Pawlowsky-Glahn, 2005). Furthermore, a principal component analysis (PCA) was performed on the elemental geochemical data set in order to highlight elemental associations with similar relative variation patterns that may be interpreted from an environmental standpoint (e.g., Montero-Serrano et al., 2010). Prior to all multivariate analyses, a log-centered (clr) transform was applied to the data (Aitchison, 1990). The clr transform is derived by dividing each variable (e.g., mineral percentage, element concentrations) by the geometric mean of the composition of the individual observations and then taking the logarithm. This operation removes statistical constraints on compositional variables, such as the constant-unit sum, and allows the valid application of classical (Euclidean) statistical methods to compositional data (Aitchison, 1990; Montero-Serrano et al., 2010). Statistical calculations were conducted with CoDaPack v2.02.04 (Thió-Henestrosa and Martín-Fernández, 2005) and "R" software using the packages "compositions" (van den Boogaart and Tolosana-Delgado, 2008) and "vegan" (Oksanen et al., 2016). Finally, the spatial interpolations of the results from bulk and clay mineralogy and elemental geochemistry were generated using the inverse distance weighting (IDW) algorithm available in ArcGIS® Spatial Analyst Tools.

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FIGURE S1. Spatial distribution of the mean grain-size for the surface sediments in (a) the Gulf of San Jorge (GSJ) and (b and c) marine park areas.



FIGURE S2. Spatial distribution of tephra (i.e., volcanic rhyolitic tephra) concentrations in the GSJ.



FIGURE S3. (a) Comparison of measured X-ray diffractogram and calculated best-fit curve obtained from RockJock v11 on a representative sample (BV01). The quartz and rhyolitic tephra (Hekla-4) standard used in the XRD analysis are also shown for comparison. (b–c) Scanning electron microscopy (SEM) images of the 300–63 μ m fraction of surface sediment sample BV01. (d) K₂O vs. SiO₂ classification diagram for glass shards from BV01, BV06, and BC11 surface samples. The geochemical composition field of different volcanic provinces of the southern part of the SVZ (Carel et al., 2011, and references therein) and ashes from the 2008 Chaitén eruption (Watt et al., 2009; Ruggieri et al., 2012) are presented for comparison.



FIGURE 54. (a) Weight % Si plotted vs. weight % quartz. (b) Weight % Ca plotted vs. weight % plagioclase. (c) Weight % Al plotted vs. weight % clays. (d) Weight % Fe plotted vs. weight % Fe-bearing + chlorite + clays.



FIGURE S5. (a) Box plot illustrating the smectite (S), illite (I), and chlorite (C) relative concentrations of GSJ sediment samples as yielded by RockJock and oriented mounted methods (<2 μ m; MacDiff). (b) Box plot of ratios S+I/C and S/I+C showing the relative clays compositions of GSJ sediment samples as yielded by RockJock and oriented mounted methods (<2 μ m).

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FIGURE S4. (a) Weight % Si plotted vs. weight % quartz. (b) Weight % Ca plotted vs. weight % plagioclase. (c) Weight % Al plotted vs. weight % clays. (d) Weight % Fe plotted vs. weight % Fe-bearing + chlorite + clays.

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 TABLE S1. Mean grain size and End-member (EM) scores for surface sediments of the Gulf of San Jorge (GSJ).

TABLE S2. Mineral composition of bulk and clay fraction of sediment samples.

TABLE S3. Chemical composition of sediment samples.

TABLE S4. Sources contributing to surface samples.

TABLE S5. (a) Minerals identified in RockJock v11, (b) consolidated list of minerals, (c) minerals used in SedUnMix, and (d) samples associated to sources in SedUnMix.