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GEOSCIENCES

Stable-isotope ratios (δ^{18}O and δD) in a firn core from West Antarctica

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Abstract: A 22.48 m long ice core (BR-IC-4) was collected in the West Antarctic Ice Sheet (at 83°58'59.4" S, 80°07'01.4" W, 1,295 m above the sea level) during the Austral summer of 2004–2005, as a contribution to the International Trans-Antarctic Expedition program. The isotopic composition (δD and δ^{18}O) of 599 samples, corresponding to the upper 12.98 m of the ice core, was determined by gas source mass spectrometry and cavity ring-down spectroscopy. Relative dating was based on the isotopic ratios and major ions (MS^-, Na^+, $nssSO_4^{2-}$) and trace elements (Na, S, Sr) concentrations. The record covers approximately 13 years – from 1990 to 2003. The mean accumulation rate of 0.48 ± 0.09 m water equivalent per year (m eq H_2O a^{-1}) is relatively high for the geographical area and possibly results from snowdrifting from near areas, as attested by ice glaze surfaces in other sites in the region. The stable isotope δD content varies between -367.90‰ and -256.30‰ (mean -314.42 ± 19.01‰); and δ^{18}O ranges from -44.96‰ to -35.08‰ (mean -39.95 ± 2.05‰). Deuterium excess values (mean 3.70 ± 1.54‰) indicate episodic intense oceanic evaporation and high relative humidity in the moisture sources.

Key words: Antarctica, firn core, glaciology, stable isotopes

INTRODUCTION

The history of Antarctica's climate provides the setting for understanding present-day environmental changes. It allows researchers to determine the processes that led to the development of our present interglacial period and to define the ranges of natural environmental variability on timescales from decades to millennia that have prevailed over the past millions of years (Convey et al. 2009). Recognizing the importance of polar regions, in addition to atmospheric monitoring programs, national and international programs have drilled and extracted numerous deep ice cores from Greenland and Antarctica (Legrand & Mayewski 1997).

Climate monitoring through continuous instrumental measurement and reliable global satellite monitoring – associated with historical records – enables us to interpret the climate. However, this kind of observation is relatively new; they began in the 19th and 20th centuries, respectively. Therefore, paleoclimatic proxies are needed to cover a large temporal scale, and ice cores are some of the best tools to carry information on temperature, precipitation, atmospheric composition, volcanic activity, and wind patterns.

Studies aiming to collect environmental information from ice cores began in the 1960s (Jouzel 2013). Investigating their physical and chemical properties in deep drillings from the Arctic and Antarctica (*e.g.*, EPICA Community Members 2004, 2006, NorthGRIP Members 2004, Jouzel 2013) has brought considerable progress in paleoclimatology in the recent decades

(Schlosser et al. 2008, Wolff 2012). Over the last 30 years, these investigations have become frequent, mainly due to the international effort to analyze and predict future changes in the climate (Schwanck et al. 2016).

Ice cores from polar regions and low-latitude high-elevation areas, such as glaciers from mountainous tropical and temperate zones, provide a unique natural archive of past atmospheric conditions at high temporal resolution (Legrand & Mayewski 1997, Wolff 2012). The Antarctic ice sheet is perfect for various environmental studies due to its geographical position and meteorological conditions. Antarctica is located far away from continents (where local sources such as wind-blown dust, biogenic emissions from soils and vegetation, or anthropogenic emissions dominate); the region provides the cleanest atmospheric environment in the South Hemisphere. Antarctica's meteorological conditions are characterized by well-marked seasonal cycles with a long polar night, very low temperatures, and air dryness. Consequently, the chemical reactivity of the polar atmosphere is considerably reduced (*e.g.,* the quasi absence of photochemistry reactions during the winter months and the reduction of other chemical reactions at low temperatures) compared with the situation at midlatitudes. Also, only solid precipitation accumulates on the Antarctic plateau, and no seasonal melting of the snow surface occurs. Assuming we understand the relationship linking the composition of the snow cover to the atmosphere, polar ice cores offer the possibility to reconstruct past atmospheric chemistry and climate of the Earth over periods as long as several thousand years (Legrand & Mayewski 1997).

The interpretation of stable isotope concentrations (δD and $\delta^{18}O$) in ice layers successively deposited on Antarctic ice sheets is a powerful tool for reconstructing climate change in this region. These profiles contain temperature-related information since there's a linear relationship between surface temperature and oxygen and deuterium stable isotope ratios in snow and ice (Schlosser et al. 2008).

To contribute to studies about the variability of stable isotope content in the West Antarctic Ice Sheet (WAIS), the BR-IC-4 firn core was collected during the austral summer of 2004–2005 Chilean-Brazilian ITASE (International Trans-Antarctic Expedition) traverse. The firn core had its hydrogen/deuterium (δD) and oxygen-18/oxygen-16 ($\delta^{18}O$) ratios determined, interpreted, and correlated to data previously obtained in the area, aiming to help reconstruct its paleoclimatic history. Data like these are scarce in literature and represent an essential contribution to science. ITASE (International Trans-Antarctic Scientific Expedition) is a 20-nation scientific program making multiple traverses of the Antarctic continent to collect information about the variability of a wide range of environmental and climate indicators. Due to the remote location and difficulty of access, Antarctica has very few instrumental records of its climate, unlike the highly populated areas of the Northern Hemisphere. On the other hand, because of its pristine isolation, it is an ideal environment to monitor changes in the global climate. Therefore, ITASE aims to build a continent-wide database of factors relating to the environment over the last 200 years.

Stable isotopes background

Isotopes are atoms whose nuclei contain the same number of protons but a different number of neutrons. They can be divided into two fundamental kinds, stable and unstable (radioactive) species (Hoefs & Hoefs 1997). Oxygen occurs in nature predominantly in the form of ^{16}O isotope with a natural abundance of 99.76%. Moreover, it occurs as ^{17}O and ^{18}O isotopes with abundances of 0.04% and 0.2%, respectively. Hydrogen also has three isotopes: ^{1}H, ^{2}H (also known as deuterium, or D), and ^{3}H (tritium). Their natural abundances are 99.98%, 0.014% and $7x10^{-16}$%, respectively. Water molecules may occur in any isotopic combination and molecules of the same chemical formula that have different isotopic species will have different physical properties, such as density, melting and boiling point and vapor pressure (Hoefs & Hoefs 1997). However, the most common and important natural isotopic fractionation studies are $H_2^{16}O$, $HD^{16}O$, and $H_2^{18}O$.

The isotope's relative concentration can be measured more precisely than the absolute concentration. Therefore, the method for inferring paleoclimatic conditions by the content of stable isotopes uses a ratio (R) between the heavy stable isotopes and the lighter stable isotopes ($^{18}O/^{16}O$ or D/H). Differences in the abundance of stable isotopes between samples are minor; hence concentrations are measured in terms of a deviation between samples and an international standard, mostly the Vienna Standard Mean Ocean Water (VSMOW). This standard is valid because oceans are the most significant water vapor sources and have a well-known and practically uniform stable isotopic composition (Paterson 1994). Since the VSMOW standard was exhausted in 2006, the International Atomic Energy Agency (IAEA) utilizes the VSMOW2 standard sample. Results of $\delta^{18}O$ are still presented relative to the VSMOW standard. The equation is defined by Craig (1961):

$$\delta D \text{ or } \delta^{18}O \text{ ‰} = \frac{Rsample - Rstandard}{Rstandard} \times 1000c$$

Delta values are expressed in per mille (‰). The natural isotopic composition of water covers a wide range: 400‰ for δD and 40‰ for $\delta^{18}O$ (Dansgaard 1964). On the other hand, the ocean mean composition is, by convention, equal to 0‰ (Epstein & Mayeda, 1953). Dansgaard (1973) concluded that the most critical factor for determining δ values for the precipitation is the difference between the temperatures of the oceanic source and the sampled site. Oceanic surface temperatures are more stable than the air temperatures in high latitudes; thus, the δ values in polar snow are strongly dependent on the temperature at the site and moment of deposition.

Geographically, variations occur due to the latitude effect (lower δ values in higher latitudes) due to the precipitation's heavy components removal from clouds moving poleward. Similarly, the higher the latitude and the higher the distance from the coast (continentality), the lower the δ values.

The seasonal effect is reflected as lower δ values in winter than in summer in continental and polar regions as a consequence of colder environments when compared to the atmospheric vapor source at lower latitudes. Still, the paleoclimatic effect will also have lower δ values in colder periods when compared to warmer periods in a given site.

The main characteristics of water isotope distribution can be understood by the Rayleigh models, which consider isotopic fractionation processes occurring in an isolated air portion traveling poleward from an oceanic source (Dansgaard 1964, Masson-Delmotte et al. 2008). Such fractionation processes occur in most water phase transitions during their atmospheric cycle. These water phase transitions are derived from the difference between HDO's (or $H_2^{18}O$) saturation vapor pressure and $H_2^{16}O$ saturation vapor pressure (equilibrium effect) and between their molecular diffusivity in the air (kinetic effect) (Jouzel 2003).

The Rayleigh model proposed by Dansgaard occurs at equilibrium conditions, *i.e.*, a slow process with immediate removal of the condensed vapor after its formation. According to Jouzel (2003), these models present the combined influence of oceanic source temperature and the condensation temperature in the precipitate isotopic content. Aside from that, many other factors may influence the isotopic ratio, like other source characteristics that control the evaporation kinetics (*e.g.*, relative humidity and wind speed) (Merlivat & Jouzel 1979) or microphysics processes prevailing at the clouds (*e.g.*, saturation value during snow formation) (Fischer et al. 2007). These factors can result in seasonality changes and intermittency in radiative particles precipitation or wind erosion (Gallée et al. 2001), affecting the interglacial to glacial signal differently. Changes in the cyclonic activity and advection ratio by the mean circulation and turbulent transport also may affect the signal (Jouzel 2003).

Given that δ^2H and $\delta^{18}O$ are both modified by mass-dependent fractionation processes and are part of the same water molecule undergoing transformation, global precipitation follows a linear relationship $\delta^2H = 8\delta^{18}O + 10$, which is called the global meteoric water line (GMWL, Craig 1961). The intercept of the GMWL is referred to as d-excess (deuterium-excess factor) and helps distinguish equilibrium and nonequilibrium processes (Beria et al. 2018)

MATERIALS AND METHODS

The 22.20 m long BR-IC-4 firn core was extracted at the coordinates 83°58’59.4” S; 80°07’01.4” W, 1.295 m above sea level (measured by an altimeter with ±10 m precision) (Fig. 1). The temperature was measured at a depth of 12 m in the core borehole using a calibrated platinum probe; it is 33.0 ± 0.5°C. At this depth, the surface snowpack is no longer affected by seasonal variation in atmospheric temperature and represents the mean annual temperature at the site (Cuffey & Paterson 2010).

The sample site is located at the West Antarctic Ice Sheet, 1.694 m away from the coast. The core drilling was performed during the Latin-American traverse in December 2004. The scientists left the Parodi Chilean Station (80°18’ S, 81°21’ W) towards the geographic South Pole (90° S), covering 2.411 km. During the traverse,

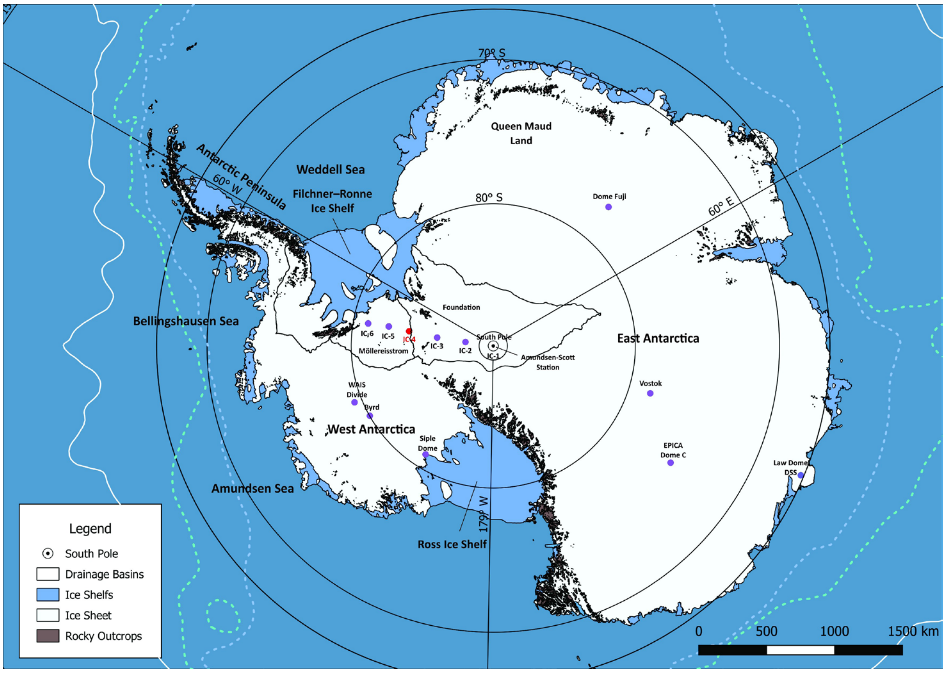

Figure 1. BR-IC-4 and other firn/ice core drilling sites drilling during the 2004–2005 Antarctic traverse, near the basin divide between the Foundation and Möller ice streams. Source: Quantararctica.

another five cores were obtained, which were analyzed by Bayer da Silva (unpublished data), Hammes et al. (2009), De Mello Marques et al. (2014), Schwanck et al. (2014), Marquetto et al. (2015), Lindau et al. (2016), Gerhard et al. (2020) and Tavares et al. (2020). The drilling site was selected based on the following conditions: (i) the area is isolated and has permanently low temperatures, avoiding superficial melting, percolation, and refreezing of the water in snow layers – this stability helps to preserve the chemical characteristics of snow at the time of deposition; (ii) the drilling site is located on drainage basin divide (separating the Foundation and the Möller ice streams basins) and, therefore, has a simple glacial dynamics, which avoids large horizontal displacements on the ice column, making corrections unnecessary.

Sample handling

The firn core extraction was accomplished using an electromechanical drill FELICS (Fast Electromechanical Lightweight Ice Coring System) developed and manufactured by the FS Inventor AG company in collaboration with Paul Scherrer Institute and the University of Bern (Switzerland). During the traverse, five other cores were collected – two of them previously analyzed by researchers of the Centro Polar e Climático of the Federal University of Rio Grande do Sul (Brazil) and are used for comparison in this article: BR-IC-2 (88°01’21.3” S; 82°04’21.7” W, altitude 2,621 m) and BR-IC-5 (82°30’30.8” S; 79°28’02.7” W, altitude 950 m). The ice cores were extracted every 220 km, on average. Immediately after acquiring core sections, each one was weighed with an electronic balance Bioprecisa BS3000A model with a precision of 0.1 g and had its linear dimensions (length and diameter) taken. The 10–15 m deep borehole temperatures were obtained using a probe composed of a silver strength previously calibrated and placed onto the tip of an electric cable. The probe remained in the pit for 24 hours to establish thermal equilibrium with the walls of the well (Langway 1970, Mayewski & Goodwin 1997).

Sections of approximately 1 m were stored in polyethylene packages and stowed in polystyrene boxes, transported from Patriot Hills to Punta Arenas, Chile. Afterward, the ice cores were taken under refrigeration to New York, USA, and finally driven by a refrigerated truck to Bangor, USA, where it was kept in a cold industrial chamber until preparation for analysis.

Decontamination and subsampling were performed inside a certified cold room (-20 °C) at the Climate Change Institute, University of Maine, USA. The outer layers of the sections underwent manual scraping with a ceramic knife to prevent contamination. The ends of the sections were removed as well since they are most susceptible to contamination.

The firn core melting was conducted in a cleanroom class 100, using a Continuous Ice Core Melter System developed by researchers of the CCI from the model proposed by Röthlisberger et al. (2000). The system allows the simultaneous and continuous sampling for three different glaciochemical analyses: major elements by Ion Chromatography (IC), trace elements by Inductively Coupled Plasma Sector Field Mass Spectrometry (ICP-SFMS), and stable isotopes by Isotope Ratio Mass Spectrometry (IRMS) and Cavity Ring-Down Spectroscopy (CRDS) (Osterberg et al. 2006).

Glaciochemical analyses

The glaciochemical analyses were performed at the upper 12.98 m of the BR-IC-4 core, corresponding to 599 samples (Table I). The laboratory works were developed at the Climate Change Institute (CCI, University of Maine, USA) and at the Glaciochemistry Laboratory of the

Table I. Types of analysis vs. BR-IC-4 samples.

Type of analysis	Samples	Total samples	Method	Time Slice
δD	1–200	200	GSMS	1998–2003
	202–600	399	CRDS	1990–1998
$^{18}O/^{16}O$	202–600	399	CRDS	1990–1998
MS^-, Na^+, $nsSO_4^{2}$	1–200	200	IC	1998–2003
Na, S, Sr	1–600	599	ICP-SMS	1990-2003

Centro Polar e Climático (CPC, Federal University of Rio Grande do Sul, Brazil).

The $^2H/^1H$ (δD) ratios of the first 200 samples were analyzed using Gas Source Mass Spectrometry (GSMS) at the CCI, following the analytical procedures described by Osterberg et al. (2006) – as part of Lindau and contributors' work entitled "Ionic variability in snow and firn along an Antarctic transect" (2016). The isotopic ratios were measured via Cr reduction with a Eurovector elemental analyzer coupled to a Micromass Isoprime mass spectrometer (0.5‰ precision based on long-term standard analysis statistics). The δD parameter was determined according to the reference standard VSMOW2 – Vienna Standard Mean Ocean Water.

The other samples (202 –600 to $^2H/^1H$ and 01–600 to $^{18}O/^{16}O$) had their stable isotopic ratios (δD and $\delta^{18}O$) measured at the CPC using the PICARRO® L2130i Cavity Ring-Down Spectroscopy (CRDS) system. The near-infrared laser technology measures the absorption rate of an electromagnetic pulse confined in a closed optical cavity with a high-quality factor. Before beginning the ice core's samples analyses, two rounds of analyses of internal standards of the laboratory – which are regularly calibrated by certified standards (SLAP – Standard Light Antarctic Precipitation and GISP – Greenland Ice Sheet Precipitation) – are performed. The internal standard values are adjusted in calibration curves through international certified standards in the Excel software. Subsequently, each internal standard has its mean calculated, according to the values obtained in the two rounds of analyses. Finally, it's verified if the deviation between the means of the internal standards does not differ more than 0.03‰ for $\delta^{18}O$ and 0.2‰ for δD. The resulting delta values are expressed per mille using the VSMOW2 standard.

Trace element analyses were performed with the CCI Thermo Electron Element2 ICP-SMS, with the isotopes measured in low (Sr) and medium resolution (S and Na). An ESI Apex high-sensitivity inlet system is used to increase instrument sensitivity and reduce oxide formation in the plasma, lowering detection limits and allowing less abundant isotopes to be measured. The Cetac (Omaha, NE) Model ASX-260 autosampler is located within a class-100 HEPA clean bench adjacent to the instrument to reduce contamination further. The ICP-SMS is calibrated daily with five standards that bracket the expected sample concentration range. Certified water reference material, SLRS-4 (Environment Canada), is used to verify the calibration. No internal standard is used to minimize sample handling and possible contamination (Osterberg et al. 2006). Major ions were analyzed in the first 200 samples using a Dionex ion chromatograph with chemical suppression and conductivity detectors. Anions were measured using an AS-11 column, a 400 µL sample loop, and a Dionex reagent free controller producing a KOH eluent gradient of 1–8 µM. Cations were measured using a CS-12A column, 500 µL loop, with 25 mM methanesulfonic acid eluent (Osterberg et al. 2006). The systems were calibrated weekly using five standard solutions prepared from concentrated stock solutions (Ultra Scientific, USA). Those results were checked with the reference solution ION-92 (National Water Research Institute, Canada).

Reanalysis data and back trajectories

The reanalysis data used in this article were obtained in the Climate Reanalyzer of the Climate Change Institute in association with the National Science Foundation, using the ECMWF – ERA5 data set. ERA5 can approximate the Antarctic near-surface temperature measured by weather stations and effectively represent Antarctica's temperature changes and its three subregions (Zhu et al. 2021). The backward trajectories model was made using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) software (Windows version) from the National Oceanic and Atmospheric Administration (NOAA) Air Research Laboratory (available online at http://ready.arl.noaa.gov/HYSPLIT.php). All trajectories were run backward for 120 h using NCEP/NCAR Reanalysis, which can determine long-term monthly means. Five-day back trajectories are appropriate for this work since shorter trajectories tend to be truncated close to the ice core site, while longer trajectories extend into the Southern Ocean and show little spatial coherency north of ~60° S (Sinclair et al. 2013). Initial starting heights of 500, 1000, 1500, 2000 and 2500 m were chosen for showing consistent flow paths.

RESULTS

Core stratigraphy and climate history

The ice core's annual layers in Antarctica provide a detailed record of the accumulation rate, which can be used to examine the climatic variability through time and change in mass balance in the ice sheet. The mean accumulation rate in a given site is partially related to the topography of the ice sheet and the distance to humidity sources. At the same time, the temporal variability in these records reflects changes in atmospheric transport of humidity, cyclonic positioning and intensity, precipitation seasonality, and temperature (Zwally et al. 1989, Vaughan et al. 1999, Kaspari et al. 2004).

Firn/ice cores can record the amount of snow accumulation at a specific location over a range of timescales. Barring post-depositional processes, the recorded snow accumulation results from solid precipitation, sublimation, wind erosion/deposition, and meltwater runoff. Integrated over the Antarctic Ice Sheet, the contributions made by sublimation/deposition, wind redistribution, rainfall, and meltwater runoff are relatively small, and therefore the dominant component of Antarctic surface mass balance is solid precipitation. However, post-depositional processes can also play a role in shaping the final state of the ice sheet, especially in the West Antarctic Ice Sheet, since it has a lower average elevation and is characterized by numerous ice streams and glaciers that flow towards the sea. Furthermore, the coastal parts of the WAIS are subject to both higher temperatures and greater precipitation, making them more susceptible to post-depositional changes.

The BR-IC-4 drilling site is located in the ice divide between Foundation and Möller ice streams. The mean annual accumulation rate increases as it distances from Patriot Hills (80°20' S, 81°25' W) because near the mountains, ablation through sublimation occurs – resulting in blue ice areas. However, moving towards the South Pole, the annual accumulation mean rate diminishes due to higher continentality and altitude. By the BR-IC-4 drilling site, the ice flows to the Filchner-Ronne Ice Shelf (Casassa et al. 1998). The peaks in this region influence the wind patterns, causing them to flow to the NW. The highest spot elevations are the Thiel (2.810 m; 85°3' S, 90°51' W) and Withmore (3.020 m; 82°35' S, 104°30' W) mountains, which distance respectively 175 km and 165 km from BR-IC-4 site. The site's borehole temperature is -33 °C.

The accumulation rate was determined at the BR-IC-4 core site based on the dating acquired by the seasonal chemical variability, using the density to calculate in m eq H_2O. The mean annual accumulation corresponds to 0.48 ± 0.09 m eq H_2O a^{-1} with maximum values detected in 1999, 0.70 m eq H_2O a^{-1}, and minimum values in 2002, 0.37 m eq H_2O a^{-1}. Calculated mean annual precipitation (ECMWF – ERA5) at the drilling site and mean accumulation rate values from 1990 to 2003 are found in Figure 2.

The BR-IC-4 relationship between snow accumulation and calculated precipitation presents a correlation coefficient (R^2) of 0.0826 ($\rho = 0.822$), possibly due to post-depositional changes as snowdrift is common on an ice sheet.

Air mass trajectory models indicate that the primary wind sources are the Weddell Sea and the Queen Maud Land during the austral summer months. During the winter months, wind originates mostly in the interior of Queen Maud Land and its coastal zones according to reanalysis models (Figure 3). HYSPLIT simulations were run for each day of summer and winter peak months for every year of the ice core's time slice (1990–2003). Cluster trajectories were elaborated for demonstration purposes.

BR-IC-4 firn core stratigraphic analysis (Fig.4) was made during fieldwork using a transmitted light table to identify and classify visible polar snow layers originated by deposition due to the precipitation variation and fluctuations in surface meteorological conditions (Langway 1970).

The upper 4.7 m consists of snow (ice crystals that have not suffered significant modifications since its deposition) and presents four interleaved layers (varying between 2 and 6 cm thick) of compaction zones, *i.e.*, layers whose density was increased due to wind action (Fig. 4).

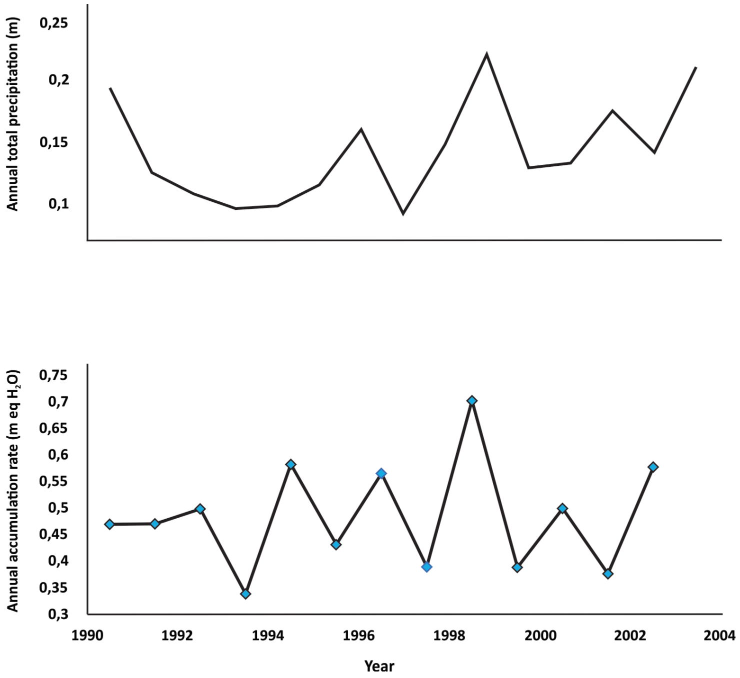

Figure 2. Graphic showing the annual total precipitation in the drilling site during the recording period (Monthly Reanalysis Timeseries (ECMWF – ERA5) – Climate Reanalizer) and BR-IC-4 core annual accumulation rate.

The ice core comprises a thick layer of firn in the following meters, which occurs until the core's base. Firn represents crystals in an intermediate phase, indicating a continuous transformation from snow properties without sudden changes (Cuffey & Paterson 2010) and still maintaining the snowpack permeability. The BR-IC-4 core has a mean density of 0.53 g cm^{-3} and no evidence of melting, percolation, or refreezing water during fieldwork. The density was determined for each core piece, which varied between 20 and 50 cm with a diameter of 8.25 cm.

Dating

The BR-IC-4 core was dated by multiparametric counting of annual layers (Fig.5). Isotopic ratios (δD, $\delta^{18}O$), major ions (MS^-, Na^+, $nssSO_4^{2-}$), and trace elements (Na, S, Sr) data were utilized for identifying seasonal variations. Peaks corresponding to the austral summer signal were used to indicate annual sequences. The isotopic ratios, ionic concentrations, and trace elements profiles were elaborated with the moving average from two adjacent elements, so any noises can be filtered, giving better intelligibility in identifying the years of precipitation.

Peaks of δD and $\delta^{18}O$ indicate high-summer precipitation when isotopic fractionation is lower than in the middle of winter. Near air masses, sources that carry moisture (due to sea ice retraction) result in precipitation with a more positive isotopic signal. The ionic parameters such as MS^- (methylsulfonate) and $nssSO_4^{2-}$ (non-sea-salt sulfate) have their peaks in summer. The MS originated from marine biologic activity, and the $nssSO_4^{2-}$ is predominantly derived from methysulfonate oxidation and volcanic activity (Benassai et al. 2005, Koffman et al. 2013). On the other hand, the Na^+ has its maxima in the winter, being an important proxy for sea spray generated by frost flowers ablation (Rankin et al. 2000, 2002, Benassai et al. 2005). The Sr, mainly derived from sea salt, peaks in the winter (Schwanck et al. 2016). Sulfur is more variable and may be originated in many sources such as volcanoes, crustal particles, sea salt, marine biologic activity, and anthropogenic (Herron 1982, Koffman et al. 2013). It was possible to identify a sulfur peak in 1991 CE between September and November when plumes from Mount Pinatubo (Philippines) and Cerro Hudson (Chile) eruptions were recorded above the geographic South Pole.

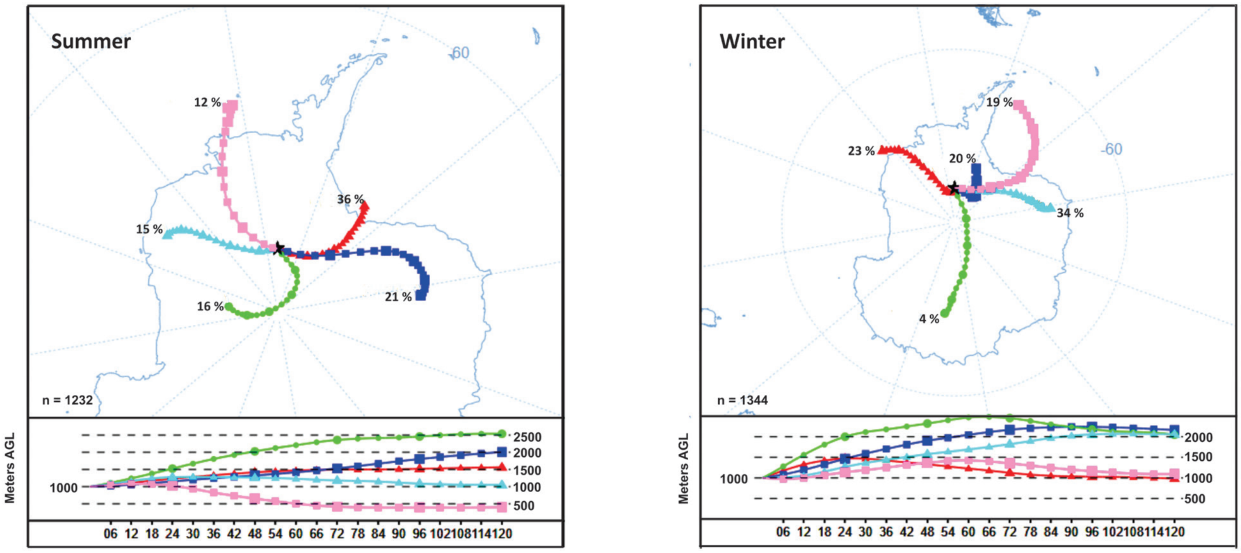

Figure 3. HYSPLIT model (NOAA) of air masses five-day backward trajectories during the summer and winter months from 1990 to 2003.

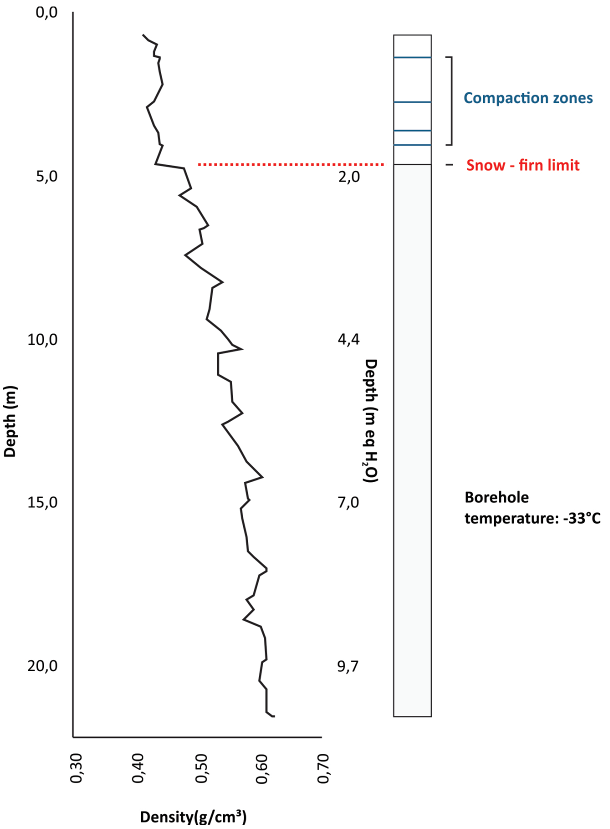

Figure 4. Density profile (g cm^{-3}) and stratigraphy of the BR-IC-4 core (real depth scale on the left and meter water equivalent on the right side).

It is estimated that the precipitation of volcanic material continued until 1994 (Cole-Dai & Mosley Thompson 1999).

The dating of the BR-IC-4 core begins in the year 2003 CE, due to the removal of the first 32 cm for other analysis, and it extends back to 1990, covering 13 years of record.

Water stable isotope ratios

The summary of the isotopic content (δD, $\delta^{18}O$ and d) and statistical analysis of the BR-IC-4 firn core samples are presented in Table II. Due to mismatched and incoherent values, three samples were discarded from 599 analyzed samples (215, 216, and 217). The probable cause for this is post-sampling alteration.

The isotopic series (δD and $\delta^{18}O$) and the deuterium excess (d = δD - 8$\delta^{18}O$) in-depth scale

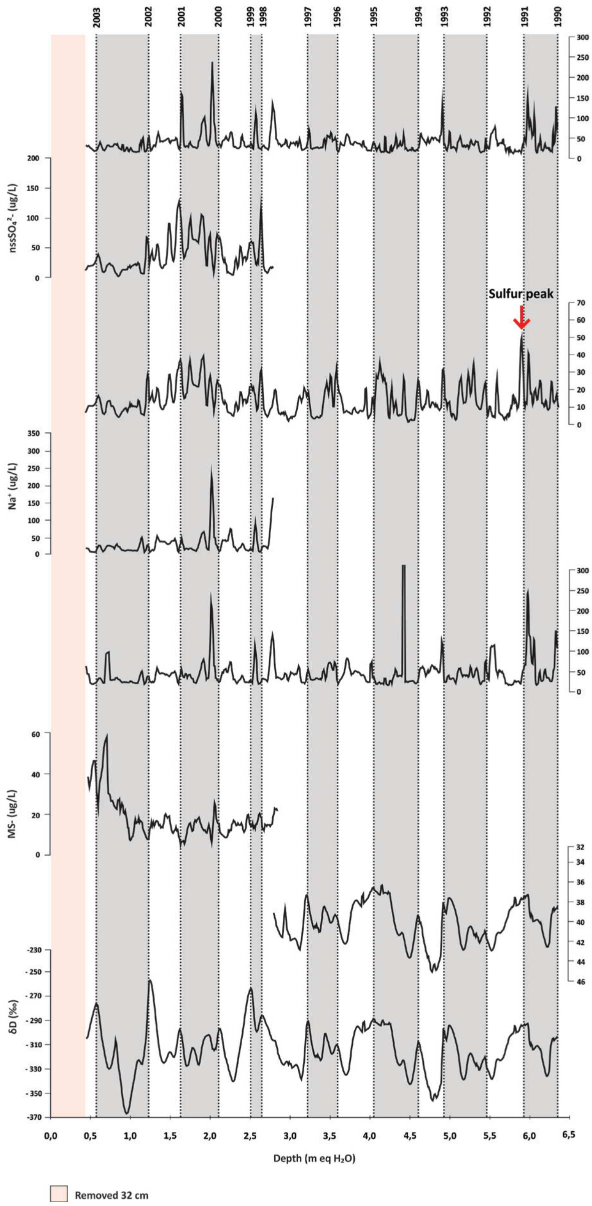

Figure 5. BR-IC-4 core dating determined by the isotopic ratios (δD, $\delta^{18}O$), major ions (MS^-, Na^+, $nssSO_4^{2-}$), and trace element concentrations (Na, S, Sr) seasonal variations. The arrow indicates the sulfur peak in 1991.

(meter water equivalent) of all the samples are presented in Figure 6. The obtained isotopic values ratify the compilation of results from the studied area, shown in a stable isotope ratio map of Antarctica (Masson-Delmotte et al. 2008), in which the δD varies between -240 and -400‰ and the $\delta^{18}O$ between -30 and -50‰. The excess deuterium values are similar to those found in this sector by Dahe et al. (1994).

The results follow the global meteoric water line (Craig 1961), pointing out the intimate relationship between hydrogen and oxygen isotopes in water molecules. This relationship, shown in Figure 7, also states no primary post-depositional isotopic fractionation (by melting or refreezing).

DISCUSSIONS

Geographic factors and precipitation origin

BR-IC-4 core was collected 1.694 kilometers away from the coast at an altitude of 1.295 m, with a borehole temperature of -33 °C. Despite the study area being in the continent's interior, the precipitation originates on the coast and ocean (Bellingshausen Sea, Amundsen Sea, Weddell Sea, and Queen Maud's Land). We estimate the potential origin of precipitation only using the backward trajectories model resulting from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT). However, as we have not sampled precipitation episodes of precipitation at the BR-IC-4 site in conjunction with air mass trajectories and/or wind directions, we are not able to say which source area contributes most to annual precipitation and whether there is

Table II. Isotopic ratios summary.

Isotopic ratios (δD and $\delta^{18}O$) and deuterium excess (d)			
Isotopic parameters	**δD**	**$\delta^{18}O$**	**d**
Sample number	596	396	396
Mean	-314.42	-39.95	3.70
Maximum	-256.30	-35.08	6.45
Minimum	-367.90	-44.96	-3.23
Median	-314.95	-39.91	3.77
Deviation standard (±)	19.01	2.05	1.55

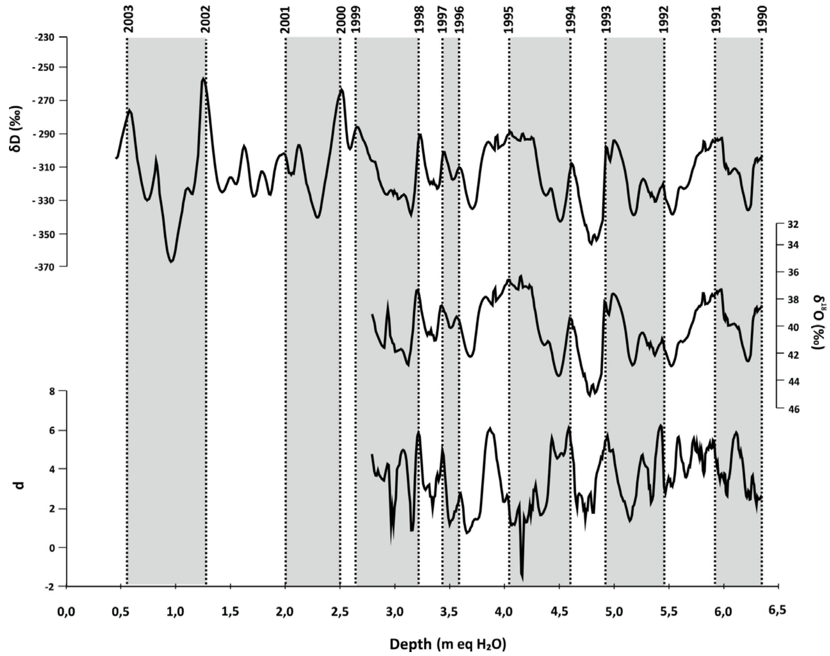

Figure 6. Isotopic profiles (δD, $\delta^{18}O$) and deuterium excess (depth in m eq H_2O).

greater or lesser precipitation at a certain period of the year.

Isotopic ratios depend on fractionation processes during water phase changes in its atmospheric cycle and due to different saturation vapor pressures and diffusivity of water molecules (which is inversely proportional to its mass) (Cuffey & Paterson 2010). Heavier isotopes are preferably extracted from water vapor during condensation while transported, making the rest and subsequent precipitations isotopically lighter.

The different origins of moisture and their paths also interfere with this fractionation. Regions in the interior of the WAIS, especially at altitudes higher than 2,000 m, present more negative isotopic values than areas closer to the Austral Ocean (Masson-Delmotte et al. 2008, Hou et al. 2013). Different moisture origins and transportation paths may account for regional peculiarities, regional impacts of moisture history and origin on isotopic values, and BR-IC-4 isotopic depletion, compared to coastal ice cores, may be linked to an orographic shadow effect on air masses that cross the ice divide between the Foundation and Möller ice steams.

When comparing the BR-IC-4 to other ice cores, such as BR-IC-2 and BR-IC-5 retrieved in the same traverse, it is possible to observe the mentioned relations (Table III). Because the BR-IC-4 predominantly receives influences from the coast and due to its lower altitude (1,295 m), it shows lower isotopic fractionation when compared to the BR-IC-2 (located closer to the geographic South Pole at an altitude of 2,621 m) and greater when compared to the BR-IC-5 (closer to the coast at the height of 950 m). Therefore, it’s possible to verify that the farther from the shore (*i.e.,* higher latitudes), the more intense the isotopic fractionation of the air masses, mainly due to lower temperatures associated with the increase in elevation and distance from the source of humidity. Mean isotopic ratios values can be seen in Table III.

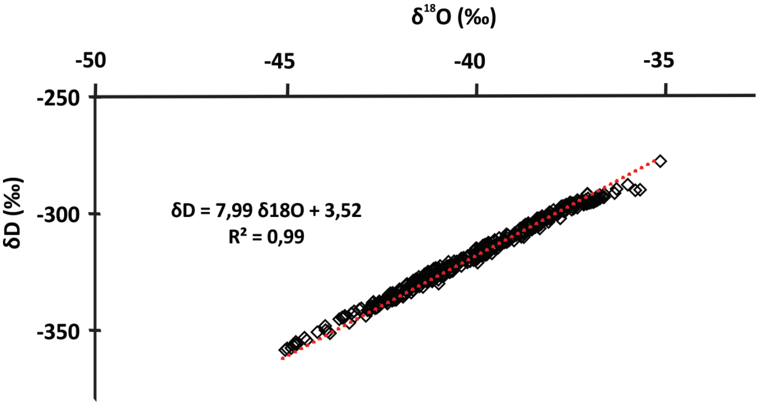

Figure 7. Graphic showing the linear relation between the isotopic ratios (δD, $\delta^{18}O$) of the BR-IC-4 core.

Table III. BR-IC-2, BR-IC-4, and BR-IC-5 cores details. BR-IC-2 and BR-IC-5 data from Ilha, unpublished data.

Core	BR-IC-2	BR-IC-4	BR-IC-5
Latitude	88° 01’21.3”	83° 58’59.4”	82° 30’30.8”
Longitude	82° 04’21.7”	80° 07’01.4”	79° 28’02.7”
δD	-373.18	-314.42	-285.59
$\delta^{18}O$	-46.66	-39.95	-35.95
d	3.17	3.70	3.25
Time slice	1918–2003	1990–2003	1978–2003

Topography and accumulation rate

The mean accumulation rate of snow is negatively related to the distance of the coast and elevation; nevertheless, these factors are not the only determining ones. During the traverse, it was observed that the sampling area is relatively near to a surface of ice glaze, located between the BR-IC-2 and the BR-IC-3 cores (Figure 8). Localities with ice glaze surfaces have the annual snow precipitation volume removed by wind action and sublimation. It occurs in relatively steep areas that generate katabatic winds. This characteristic may contribute to higher accumulation at the site of the BR-IC-4 (0.48 m eq H_2O a^{-1}), since the removed snow is transported from those sites to the lower areas. Such factors can be observed when comparing the accumulation rate of the BR-IC-4 with the accumulation rate of BR-IC-5 and BR-IC-2 (Table III). The annual mean accumulation rate at the BR-IC-2 site is 0.25 m eq H_2O a^{-1} and 0.35 m eq H_2O a^{-1} at the BR-IC-5 site – the data was calculated based on their stable isotopic ratios.

According to Thomas et al. (2017), in Ellsworth Land (where the BR-IC-4 is located), there has been an increment in precipitation volume from the 20th century due to reducing regional atmospheric pressure at sea level. It results in stronger circumpolar westerly winds and amplified north flux. The current positive phase of the Southern Annular Mode and the surface temperature increase in the West Pacific may also be linked to increased precipitation. Furthermore, sea ice conditions in the Bellingshausen Sea play an essential role in the climatic system, acting as a barrier to the humidity and heat transport between the ocean and the atmosphere. Reconstructions of sea ice show a marked decline in the area of the Bellingshausen Sea during the 20th century (Abram et al. 2010). It causes greater availability of moisture at surface level and an increase in atmospheric humidity transport poleward.

Variations in δD and $\delta^{18}O$ isotopic ratios

Stable isotopic ratios in snow and ice are crucial for temperature reconstruction, mainly because there is a linear relationship between oxygen and hydrogen isotopes and the mean annual temperature of the deposition site (Dansgaard 1964, Schlosser et al. 2008).

According to Dansgaard (1964), the isotopic variations are substantially controlled by the condensation temperature at the precipitation site. However, many other aspects modulate the isotopic composition in the ice core archives, especially geographic factors (*e.g.*, continentality, latitude, and elevation) and different moisture sources and transport forms (Cuffey & Paterson 2010). As stated by Masson-Delmotte et al.

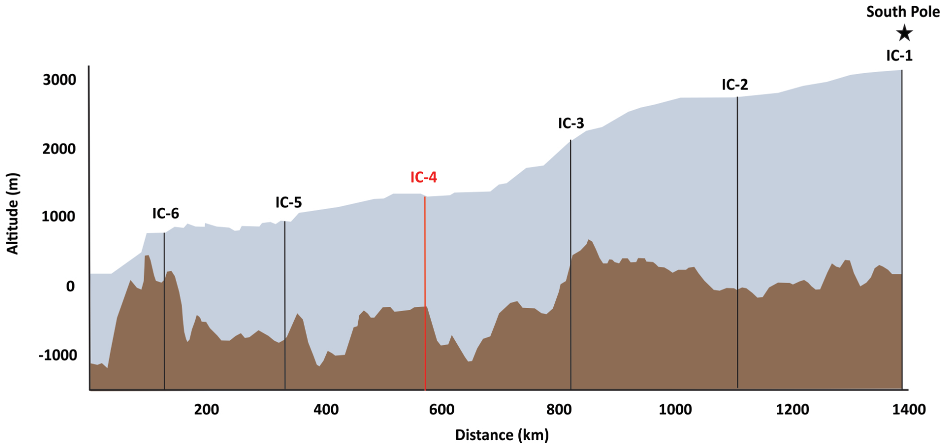

Figure 8. Cross-section of the Antarctic ice sheet over the rocky basement (IC-6: Schwanck et al. 2014; IC-5: Bayer da Silva, unpublished data, Ilha, unpublished data; IC-3: De Mello Marques et al. 2014, Gerhard et al. 2020, Tavares et al. 2020; IC-2: Ilha, unpublished data). Source: Quantarctica.

(2008), the site elevation appears to be the primary control for Antarctic δD and $\delta^{18}O$ spatial variations. The observations show isotopic values that are less depleted than modeled mean isotopic ratios in their Antarctic surface snow isotopic composition (positive anomalies) on the flanks of the ice sheet (at elevations from 1,000 to 2,000 m) and inland West Antarctica. In contrast, their isotopic values are more depleted than the calculated ones (negative anomalies) in the central Antarctic Peninsula and the central East Antarctic plateau.

The temperature anomaly variation analysis shows that the core site has shown warming trends in the last years (Fig. 9). Although the isotopic fractionation is inversely proportional to the temperature, this tendency is not perceptible in the core isotopic record, which is relatively uniform throughout the analyzed period (the correlation between stable isotope ratios and temperature is not significant). However, when examining the mean annual temperature at the sampling site on a broader time scale, it is possible to observe the increase in temperature during the last decades (Fig. 9). If the progressive increase in the temperature persists, the alteration in the isotopic fractionation likely will become even more evident, resulting in less negative isotopic ratios.

According to Bromwich et al. (2013), the center of West Antarctica is one of the fastest-warming regions globally. Although only one site in the WAIS has direct temperature measurements comprising the last century (Byrd Station, 80° S, 120° W), satellite observations since 1982 provided the first evidence of the warming tendency of WAIS, especially in the winter and spring. Steig and Orsi (2013) pointed out that this scenario may be associated with factors such as higher oceanic surface temperatures in low latitudes, generating anomalous convections, variation in the local geopotential heights, and alterations in atmospheric circulation, leading to changes in the oceanic circulation pattern and, consequently, to an increase in the contribution of warmer waters in the West Antarctica margins.

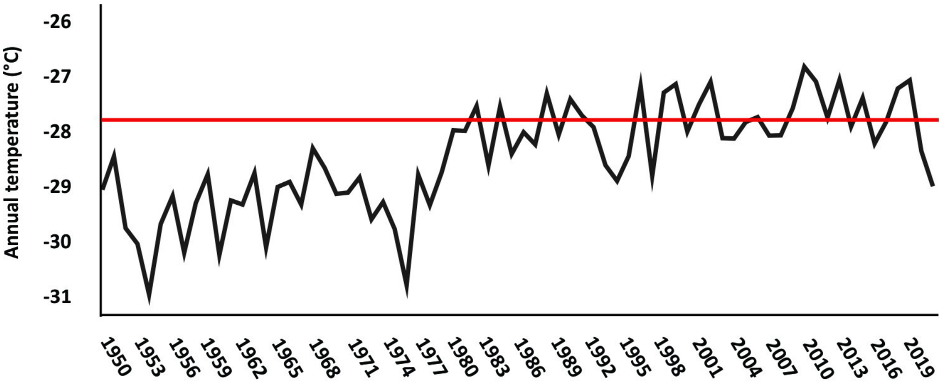

Figure 9. Annual mean surface temperature (2 m). The annual temperature anomaly in BR-IC-4 core drilling site is represented above the red line, calculated by the Monthly Reanalysis Timeseries (ECMWF – ERA5) – Climate Reanalizer, using the reference period of 1940 – present.

Table IV. **BR-IC-2, BR-IC-4 and BR-IC-5 geographic information, and accumulation rate. BR-IC-2 and BR-IC-5 from Ilha, unpublished data.**

Core	Altitude (m)	Distance from the coast (km)	Accumulation rate (m eq H_2O a^{-1})
BR-IC-2	2,621	1,967	0.25
BR-IC-4	1,265	1,694	0.48
BR-IC-5	950	1,613	0.35

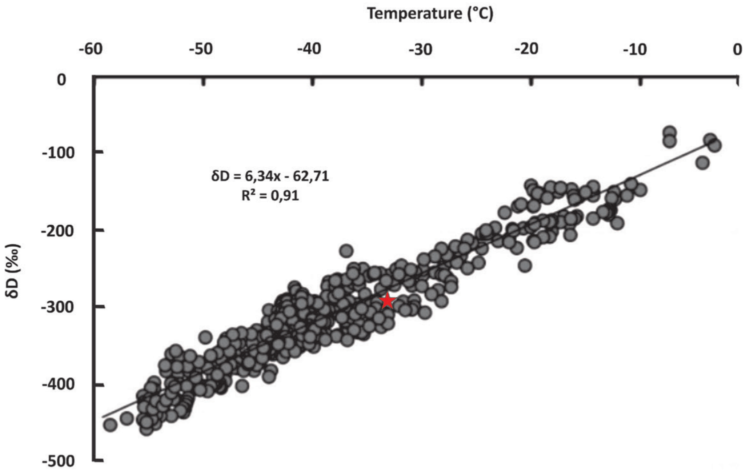

Figure 10. **BR-IC-4 (red star) results compared to a database of surface Antarctic snow isotopic composition elaborated using available measurements (more than 1000 documented locations) compiled by Masson-Delmotte et al. (2008).**

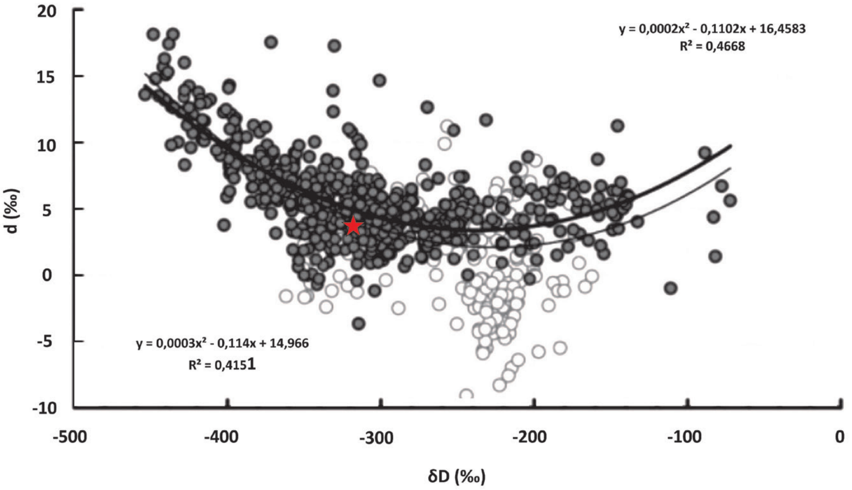

Figure 11. **BR-IC-4 (red star – d = 3.70 and y = 7,99 $\delta^{18}O$ + 3,52; R^2 = 0,99) results compared to 789 data points on the Antarctic ice sheet compiled by Masson-Delmotte et al. (2008). The white dots refer to Dry Valleys' anomalous results.**

Deuterium excess

During kinetic water fractionation, the deuterium excess reflects the slight difference in hydrogen and oxygen isotopic behavior. The kinetics depends on the oceanic source properties such as ocean surface temperature (which influences the saturation vapor pressure), the relative humidity (which controls the vapor diffusion), and wind speed (which contain the turbulent vapor transport in higher levels) as well as the kinetics of crystal formation in clouds (Jouzel & Merlivat 1984). Consequently, the deuterium excess yields integrated information about precipitation origin (Schlosser et al. 2008). Deuterium excess tracks changes in the ocean surface conditions during evaporation, primarily sea surface temperature and wind speed and humidity to a lesser extent. Pfahl and Sodemann (2014) point out that moisture source relative humidity, not sea surface temperature, is the primary driver of *d* variability at seasonal timescales.

Coastal regions show lower isotopic ratios and deuterium excess values when compared to regions in the interior of the continent. Statistical studies show that *d* is correlated positively to altitude and distance to the coast and negatively to temperature (Hou et al. 2013). The BR-IC-4 core shows low deuterium excess values (mean of 3.70 ± 1.54‰), coherent with the provenience of moisture and previous isotopic ratios distribution analysis in the Antarctic ice sheet (Dahe et al. 1994, Masson-Delmotte et al. 2008). Also, BR-IC-4 deuterium excess mean values are higher during winter months since the precipitation is originated in more distant areas (the Weddell Sea and the Pacific Ocean) – resulting in higher isotopic fractionation – when compared to summer months, in which the precipitation originates in the interior of Antarctica and the coastal zones, especially Queen Maud's Land.

BR-IC-4 low deuterium excess overall values can be attributed to the episodic nature of intense oceanic evaporation and high relative humidity in the moisture sources (evaporation is more extensive in periods with low relative humidity, corresponding to a higher *d*) (Pfahl & Sodemann 2014). Also, low *d* can be linked to a shorter transportation path of the air masses and have not undergone many condensation processes. It indicates that the air masses from oceanic sources suffered a relative direct transport, and those from the continent's interior did not go through significant orographic lifting on the way.

CONCLUSIONS

The glaciochemical analysis of the BR-IC-4 firn core confirms the linear relation between the isotopic ratios of δD and $\delta^{18}O$. The firn core represents 13 ± 1 years of snow accumulation, showing a marked seasonal variability, supported by major ions (MS^-, Na^+, $nssSO_4^{2-}$) and trace elements (Na, S, Sr) concentrations. The backward trajectories investigation and comparison with the BR-IC-2 and BR-IC-5 cores also collected during the 2004-2005 austral summer Antarctic traverse, enabled the correlation with geographic factors and the identification of moisture sources. The firn core showed isotopic values coherent with its coast distance (1,694 km) and altitude (1,295 m) when compared to the nearby snow and ice cores obtained during the ITASE's 2004 Chilean-Brazilian expedition and to those compiled by Masson-Delmotte et al. (2008). The low deuterium values likely occur due to the conditions of the precipitation origin's ocean surface (potentially mainly from the Bellingshausen, Weddell and Amundsen seas and the coast of Queen Maud Land), with episodic intense oceanic evaporation and high relative humidity. The accumulation rate of 0.48 ± 0.09m eq H_2O a^{-1} is high compared to the BR-IC-5 rate – a drilling site closer to the coast, possibly due to the snowdrift coming from near areas, as attested ice glaze surfaces in the area between the BR-IC-2 and the BR-IC-3 cores.

Although the reanalysis data obtained through the Climate Reanalizer (ECMWF – ERA5) show a clear warming trend at the BR-IC-4 drilling site, this is not clear in the stable isotopic record, and it is not possible to establish correlations of the isotopic ratios with the mean annual temperature over the analyzed period. The area's susceptibility to snowdrift might be a determining factor for this lack of correlation, since it could affect the isotopic record. Further studies with spatial coverage, including more recent records, and focusing on snowdrifiting events may produce data capable of identifying changes in the atmospheric temperature in West Antarctica and contribute to models for future climate projections.

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