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20th-Century doubling in dust archived in an Antarctic Peninsula ice core parallels climate change and desertification in South America

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Crustal dust in the atmosphere impacts Earth's radiative forcing directly by modifying the radiation budget and affecting cloud nucleation and optical properties, and indirectly through ocean fertilization, which alters carbon sequestration. Increased dust in the atmosphere has been linked to decreased global air temperature in past ice core studies of glacial to interglacial transitions. We present a continuous ice core record of aluminum deposition during recent centuries in the northern Antarctic Peninsula, the most rapidly warming region of the Southern Hemisphere; such a record has not been reported previously. This record shows that aluminosilicate dust deposition more than doubled during the 20th century, coincident with the ≈1°C Southern Hemisphere warming: a pattern in parallel with increasing air temperatures, decreasing relative humidity, and widespread desertification in Patagonia and northern Argentina. These results have far-reaching implications for understanding the forces driving dust generation and impacts of changing dust levels on climate both in the recent past and future.

aluminosilicate dust | global warming | human impacts | Patagonia | radiative transfer

Crustal dust in the atmosphere has a direct impact on climate forcing in two significant ways: modifying the radiation balance and affecting cloud nucleation and optical properties (1, 2). Atmospheric crustal dust also supplies iron, an essential nutrient for phytoplankton, to ocean surface waters and may indirectly affect climate by modulating the biological export of carbon to the deep ocean (3). Impacts of atmospheric dust on regional radiation budgets are similar in magnitude to those from sulfate and biomass burning aerosols (4) but can be either negative or positive (1). Estimates of the optical properties of dust have been revised recently as a result of improved *in situ* and remote sensing measurements (5, 6), but warming has been predicted across areas of high albedo (7, 8) such as snow- and ice-covered regions of the Antarctic Peninsula where recent warming has been pronounced (9). Although atmospheric dustiness has been linked to large-amplitude, large-scale temperature changes in past ice core studies of glacial to interglacial transitions (10, 11), it is unclear whether projected climate warming in coming decades to centuries will result in more or less atmospheric dust (12). Decadal changes in dust flux have been reported for ice cores from the Antarctic Peninsula (13, 14), but reliable, high-time-resolution records of changes in dust levels during recent decades and centuries are sparse (15).

Ice core records offer the possibility of reconstructing past changes in dust concentration (10, 11, 13–20). Most previous high-resolution ice core studies used as proxies of atmospheric dust the non-sea-salt component of soluble calcium (nssCa) or magnesium (nssMg) that are computed by using estimated elemental ratios in sea salt aerosols (11, 19). At many ice core sites, particularly coastal locations, the nssCa concentration may be uncertain because strong marine influences on glaciochemistry mean that up to 90% of total soluble calcium is derived from seawater, and varying fractionation of sea salt on sea ice surfaces and during transport means that the sea salt aerosol ratio does not always accurately reflect the seawater contribution (11, 21). In addition, the nssCa-based proxy primarily represents the chemically weathered part of atmospheric dust because the bulk of nssCa and nssMg in atmospheric dust comes from fine-grained calcium carbonate, dolomite, and gypsum deposits (17, 18, 22–25). The climate signal contained in the aluminosilicate dust record may differ significantly from that in the chemically weathered dust record because the former arises primarily from deposits of mechanically weathered sediments, such as glacial loess. Our preferred alternative to determine past changes in atmospheric dust from ice cores is to measure concentrations of elements such as aluminum that derive exclusively from crustal dust. We used a unique analytical system (26) to measure total aluminum in an ice core from James Ross Island at the northeast tip of the Antarctic Peninsula (Fig. 1) and so developed a highly resolved, continuous Antarctic ice core record of aluminosilicate dust concentration and flux during recent centuries; such a record has not been reported previously.

Results and Discussion

Our 1832–1991 high-resolution records of aluminum concentration and flux (Fig. 2) from the James Ross Island ice core indicate that atmospheric crustal dust concentrations are highly variable in time, with marked increases during the 20th century. Because aluminum in polar ice derives nearly exclusively from aluminosilicate dust, we determined the aluminosilicate dust concentration by using the mean crustal abundance of aluminum of 8.04% (27). On average, aluminosilicate dust concentration was 30 parts per billion (ppb) during the 160-year record, and flux, computed by multiplying concentration by the water equivalent accumulation rate for each year, was 17 mg m^{-2} y^{-1}. Annual average dust concentrations more than doubled during the 20th century, increasing from a mean of ≈21 ppb by mass from 1832 to 1900 to ≈44 ppb from 1960 to 1991. Changes in flux were similar, increasing from ≈12 to ≈27 mg m^{-2} y^{-1} during the same periods.

These measurements of aluminum concentration and flux from the James Ross Island ice core closely parallel changes in

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Abbreviations: nssCA, non-sea-salt component of soluble calcium; nssMg, non-sea-salt component of soluble magnesium; ppb, parts per billion; masl, meters above sea level; HR-ICP-MS, high-resolution, inductively coupled plasma mass spectrometer; ICP-PES, inductively coupled plasma optical emission spectrometer.

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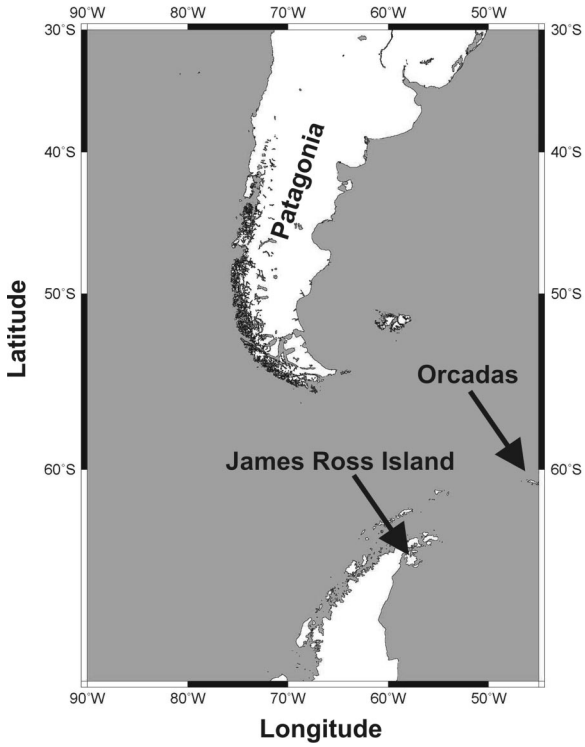

Fig. 1. Map showing the location of James Ross Island at the northeastern end of the Antarctic Peninsula and its proximity to the dust-producing regions of Patagonia and Argentina.

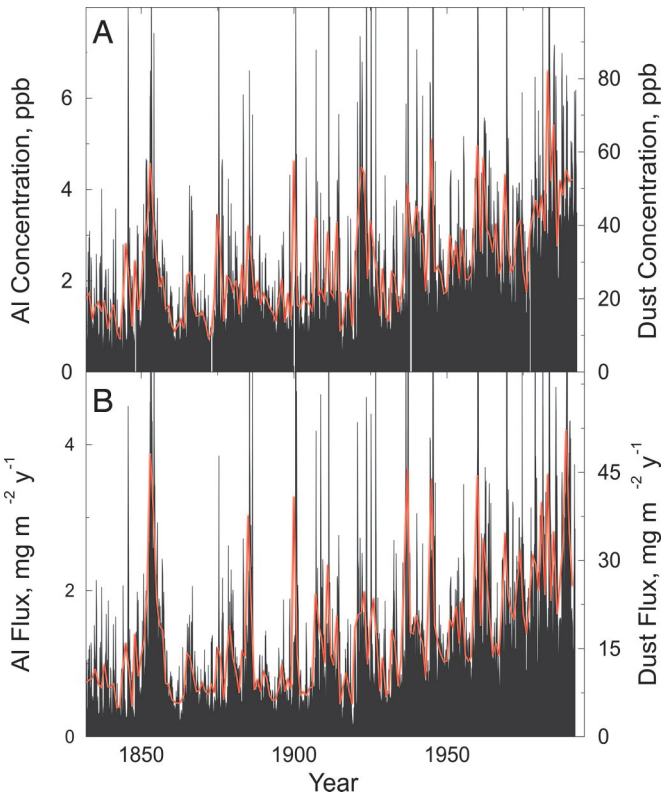

Fig. 2. Monthly averaged aluminum and aluminosilicate dust concentration (*A*) and flux (*B*) from 1832 to 1991 measured in James Ross Island ice cores. The heavy red line shows annual averages. Aluminosilicate dust was computed from the aluminum measurements by using the mean crustal abundance by mass of 8.04%.

Southern Hemisphere (28) and Patagonian (29) air temperatures during recent decades to centuries (Fig. 3). For monthly averaged aluminum concentration and flux, correlations to Southern Hemisphere air temperatures during the past 150 years are (Pearson's r) 0.398 ($P < 0.0001$) and 0.362 ($P < 0.0001$), respectively. For annual averages, correlations are 0.649 ($P < 0.0001$) and 0.615 ($P < 0.0001$), respectively. Crustal dust mobilization depends on a number of factors including wind velocity and surface conditions (e.g., type and density of vegetation cover), soil moisture content, and soil composition (22–25), so both land-use and climate changes impact dust mobility and export. A plausible explanation for this close agreement between air temperatures and dust levels is that observed warming during the 20th century, possibly amplified by land-use changes (30), has resulted in decreased soil moisture and altered vegetation cover, enhancing dust mobility in the likely source regions for dust arriving at James Ross Island.

Located at the northeastern tip of the Antarctic Peninsula, James Ross Island (Fig. 1) is an ideal site from which to develop a long-term record of atmospheric dust concentration. It is situated relatively near major dust-producing regions of southern South America, which have experienced significant desertification during the 20th century (31, 32), and also is surrounded by ocean water and thus is distant from any significant local sources of dust.

The loess region of Argentina, the most extensive in the Southern Hemisphere, covering $>1.0 \times 10^6$ km^2, is thought to be the source of crustal dust found in East Antarctica during the last glacial maximum (11, 16), whereas atmospheric aerosol studies identify Patagonia as the modern source region for crustal dust in the northern Antarctic Peninsula (23, 32). Rock outcrops that may act as local sources of crustal dust are relatively close to the Dalinger Dome ice coring site on James Ross Island, although they are also much farther away than on alpine glaciers where rock outcrops can be a few kilometers or less from coring sites. Atmospheric transport results in particle-size sorting because smaller particles (0.1 to $\approx$6.0 μm) stay suspended in the air for days to weeks and can be transported long distances. Conversely, larger particles ($>$6.0 μm) have atmospheric lifetimes of hours to days. Although episodic long-range transport of very large particles has been reported (33), larger particles are not generally transported long distances (23).

To determine whether dust found in the James Ross Island ice core is primarily from local sources or more distant sources as the result of long-range transport, we evaluated continuous microparticle count and size distribution measurements (34) made on the same ice core samples throughout the size range of 0.8–10.0 μm. Although these continuous measurements were not calibrated with Coulter counter measurements on discrete samples (34), they are in agreement with earlier studies of particles in ice cores (17) and generally consistent with long-range transport of crustal dust to the James Ross Island ice cap (23). Particle concentrations are approximately log-normally distributed with a volume-weighted mean particle size of $\approx$1.7 μm and a geometric standard deviation of $\approx$4.6 μm (34). Comparison with other ice core measurements suggests that the relative concentration of larger particles is greater at James Ross Island than at sites in West and East Antarctica, which are much more distal from potential lower latitude dust sources, but that the distribution is similar to those measured in ice cores from the central Greenland ice sheet where long-range transport is implicit.

To further evaluate possible local dust sources, we compared the James Ross Island aluminum concentration record with air temperatures in the northern Antarctic Peninsula to see whether local warming may have resulted in decreased snow cover, thus

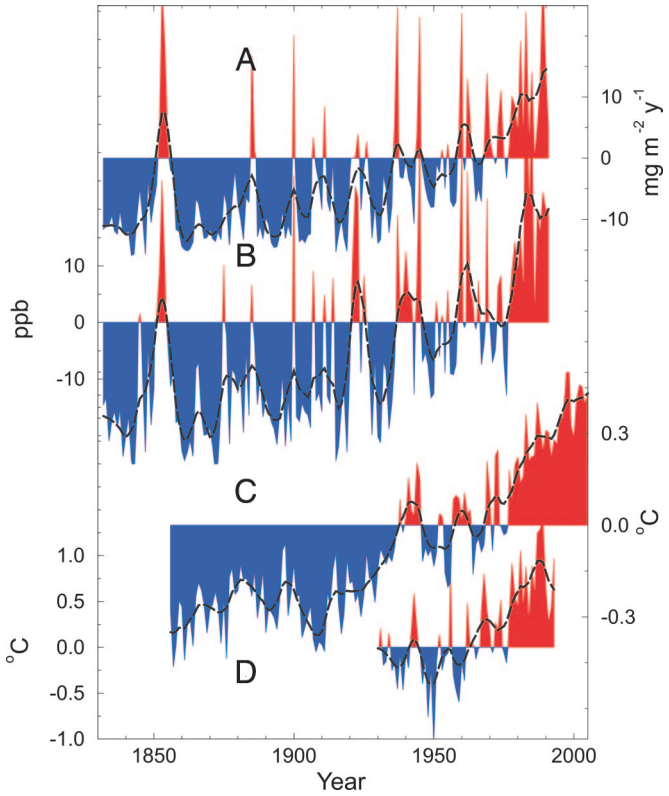

Fig. 3. Comparison of aluminosilicate dust levels at James Ross Island with air temperatures during the last two centuries. Shown are departures in annual averages from the 1930–1980 mean for aluminosilicate dust flux (*A*) and concentration (*B*) at James Ross Island, and Southern Hemisphere (28) (*C*) and southern Patagonian (29) (*D*) air temperatures. The heavy black line is based on smoothing with a nine-point triangular filter.

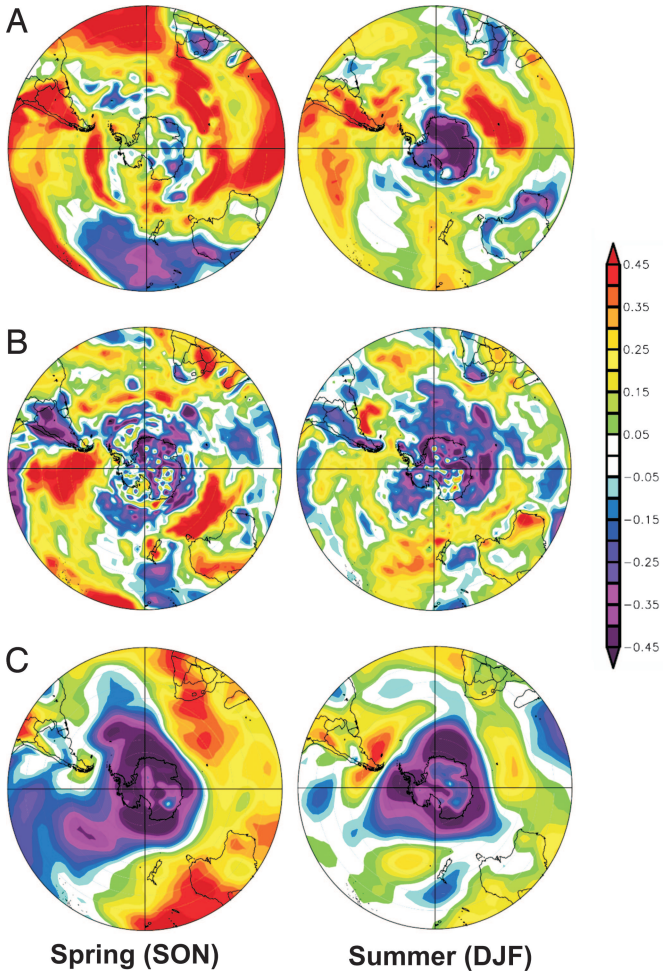

Fig. 4. Comparison of 1948–1991 Southern Hemisphere meteorology with James Ross Island aluminosilicate dust concentrations. Shown are correlations of monthly averaged aluminosilicate dust concentration with monthly air temperature (*A*), relative humidity (*B*), and sea level pressure (*C*) from the NCEP/NCAR Reanalysis. Correlations with magnitude >0.43 are highly significant ($P > 0.001$). High dust concentrations correspond to warm, low-humidity spring (*Left*) and summer (*Right*) conditions in Patagonia and northern Argentina and low sea level pressures south of 60°S.

exposing more local soil surfaces to wind erosion, especially during summer and autumn. Note that aluminum concentrations in the ice core record are highest in late winter when local dust mobilization is likely to be at a minimum because the seasonal snow cover on the Peninsula is at a maximum. The longest monthly air temperature record in the region is the 1903 to present record from the Argentine meteorological station at Orcadas (60.45°S, 44.43°W) (Fig. 1). Previous work has shown that the Orcadas temperature record is representative of the northern part of the Antarctic Peninsula (35). Correlations between the Orcadas air temperature record and the aluminosilicate dust record were not significant ($P < 0.01$), providing further evidence that variations in dust concentration observed in the Dalinger Dome ice core record are not simply reflecting local changes in dust sources.

To investigate spatial relationships between Southern Hemisphere meteorology and the James Ross Island aluminosilicate dust record, we compared seasonal averages computed from monthly dust concentrations from the ice core against monthly National Centers for Environmental Predication (NCEP)/National Center for Atmospheric Research (NCAR) reanalysis fields for the Southern Hemisphere (www.cdc.noaa.gov). Comparisons were made for each season (winter, JJA; spring, SON; summer, DJF; autumn, MAM) for the available 43-year overlap period of 1948–1991 (Fig. 4). Correlations with magnitudes greater than ≈0.35 are significant ($P < 0.01$), whereas those greater than ≈0.43 are highly significant ($P < 0.001$). Results show that aluminosilicate dust concentrations at James Ross Island are highly positively correlated ($P < 0.001$) with surface air temperatures over land areas in the Southern Hemisphere only during spring in northern Argentina and Chile and central Patagonia in summer. Similarly, we found that aluminum concentrations are negatively correlated with surface relative humidity in southern South America during winter, spring, and summer, but especially in southern and central Patagonia in winter and summer, and in Patagonia, northern Argentina, and Chile in spring.

Ice core chemical records reflect changes in source strength and also in atmospheric transport processes. Correlations between aluminosilicate dust concentrations at James Ross Island and air temperature and relative humidity suggest a change in source strength, and correlations with sea level pressure show that periods of high dust concentration also correspond to unusually low sea level pressures in spring and summer, particularly in the South Atlantic and Waddell Sea regions in spring and the Bellingshausen Sea in summer (Fig. 4). These high correlations suggest that dust transport to James Ross Island is related to increased cyclonic storm activity and vigorous atmospheric circulation in the region (9).

Although the only significant correlations over land areas in the Southern Hemisphere between the reanalysis air tempera-

ture field and aluminum concentration and flux were found in southern South America (Fig. 4), much of the agreement between Southern Hemisphere air temperature and the aluminosilicate dust record occurs in the very long period increasing trend throughout the 20th century. To evaluate this, we compared the air temperature and dust records over the period of greatest long-term change from 1930 to 1990 with and without the linear trend removed. Correlations between the detrended Southern Hemisphere air temperature and detrended aluminosilicate dust concentration and flux were highly significant at 0.498 ($P < 0.0001$) and 0.353 ($P < 0.01$), respectively. These compare with correlations of 0.652 ($P < 0.0001$) and 0.547 ($P < 0.0001$) before trend removal.

We also examined linkages at shorter timescales in the spectral domain. Multiannual signals, significant at 95% or greater above the red noise background (36), were found in the aluminosilicate dust record with periods of 1.52, 1.80, 3.55–3.71, 4.73, and 10.7–13.2 years, as well as a broad-band signal with a maximum at 21.3 years. The 3.55- to 3.71-year band corresponds to the dominant signal found in the Southern Oscillation Index and Southern Hemisphere air temperature records. Studies based on analyses of tree ring widths and instrumental meteorology records have demonstrated the importance of El Niño/Southern Oscillation (ENSO) on climate in southern South America (37), providing a physical explanation for the influence of ENSO on the dust record at James Ross Island. The 10.7–13.2 and broad 21.3-year bands are similar to 11- and 22-year sunspot cycles that are also known to have a significant impact on climate (38) and have been observed in Greenland ice core records of dust (39).

Although 20th-century increases in dust concentration and flux at James Ross Island are coincident with changes in meteorology in southern South America, they also coincide with marked changes in land use in the same region that have resulted in widespread deforestation and desertification. During the past 75 years, Argentina has lost ≈66% of its forested lands. In Patagonia, extensive sheep farming has been a primary industry for more than a century, with sheep populations increasing dramatically in the early 1930s and peaking in ≈1952 (40). Most of Argentina's cattle stock and nearly all of the goat stock are located in northern Patagonia as well. Combined with a warmer climate, overgrazing and poor land-use practices have resulted in severe land degradation and desertification throughout the region, including vegetation loss and shifts in plant species that have increased water runoff, lowered infiltration, and increased evaporation. Desertification affects as much as 93.6% of Patagonia's 78.5-million ha land surface, with 31.8% in severe to very severe stages (31).

Conclusions

Our high-resolution ice core record of aluminum from the northern Antarctic Peninsula suggests that aluminosilicate dust in the local atmosphere doubled during the 20th century. These results are in agreement with previously reported decadal records of insoluble dust particle flux from two ice cores from the central and southern Antarctic Peninsula (13, 14), suggesting that the region of increased 20th-century dust flux may be extensive. Although it is not possible to quantify how much of the recent doubling in dust levels at James Ross Island might be associated only with land-use changes, it is likely that overgrazing and deforestation together with global warming have led to desertification and thus to increased dust mobility and export from southern South America (7, 30). Atmospheric crustal dust also supplies iron, an essential nutrient for phytoplankton, to ocean surface waters and may indirectly affect climate by modulating the biological export of carbon to the deep ocean (3, 41). Relatively high surface water chlorophyll to the north of James Ross Island has been found to be highly correlated with present day Patagonian dust deposition (42). We hypothesize that the biological activity of this region and waters surrounding James Ross Island have been impacted by increased dust deposition during the 20th century. Increased dust concentrations in the atmosphere also may have altered the region's radiation balance.

Methods

Ice Cores. Two ice cores (D98 and DD98) were collected in February 1998 near Dalinger Dome (Fig. 1) at the summit of the James Ross Island ice cap [64.2°S, 57.7°W, 1,600 meters above sea level (masl)] (35). Separated by only a few meters, the cores were drilled by using an electromechanical, 10-cm-diameter drill to depths of 105.11 and 121.91 m for D98 and DD98, respectively. Following standard procedures, both cores were measured and weighed in the field and packed in plastic layflat tubing and insulated boxes for shipment to the Laboratorio de Estratigrafía Glaciar y Geoquímica de la Nieve in Mendoza, Argentina, where they were placed in frozen storage (below −10°C).

Continuous, longitudinal samples of the entire D98 and deepest 20 m of the DD98 ice core were taken in November 2005, and the samples were transported in insulated boxes to the trace element analysis laboratory at the Desert Research Institute in Reno, NV. Longitudinal samples had cross-sections of ≈3.1 × ≈3.1 cm. The D98 and DD98 glaciochemical measurements were combined to form the single JRI98 continuous record from 1832 to 1991.

Glaciochemical Analyses. Continuous, longitudinal samples of the James Ross Island ice cores were analyzed for a broad range of elements and chemical species at very high depth resolution (≈1 cm) by using a unique analytical system called Continuous Flow Analysis with Trace Elements-Dual (CFA-TED). In this system (adapted from ref. 26), a continuous series of longitudinal samples (cross-section ≈3.1 × ≈3.1 cm) were analyzed by using an ice core melter coupled to two high-resolution, inductively coupled plasma mass spectrometers (HR-ICP-MS), an inductively coupled plasma optical emission spectrometer (ICP-OES), and a traditional continuous flow analysis system.

Although the ice core samples were analyzed for a broad range of elements and chemical species, we used aluminum as a measure of aluminosilicate dust concentration and flux. Unlike other dust components, such as calcium, magnesium, and strontium that also are found in seawater or heavy metals that also are found in industrial pollution, volcanic fallout, and biomass burning emissions, aluminum is found nearly exclusively in atmospheric dust. The analytical system was configured in this experiment to include replicated measurements of aluminum concentration over the entire depth range. One measurement was made by using an HR-ICP-MS and a second measurement using an ICP-OES. With the instrument settings and sample introduction systems used, detection limits for monthly averaged aluminum concentrations were ≈0.08 and 0.8 ppb for the HR-ICP-MS and ICP-OES instruments, respectively.

Recovery Efficiency. For highly heterogeneous sample mixtures, such as ice core samples that include both dissolved and particulate components, measurements by HR-ICP-MS may not recover 100% of the sample concentration. Simultaneous ICP-OES and HR-ICP-MS measurements provide a means to evaluate recovery efficiency. Comparison of the HR-ICP-MS and ICP-OES measurements of aluminum in the James Ross Island ice cores shows that HR-ICP-MS average recovery was 82% and that recovery was essentially constant throughout the full depth range of the measurements. Both HR-ICP-MS and ICP-OES measurements were resampled to 5-cm-depth resolution for comparison. The correlation coefficient between the ≈2,250 samples in the overlapping record was 0.81 ($P < 0.0001$) with an average HR-ICP-MS to ICP-OES concentration of 0.82.

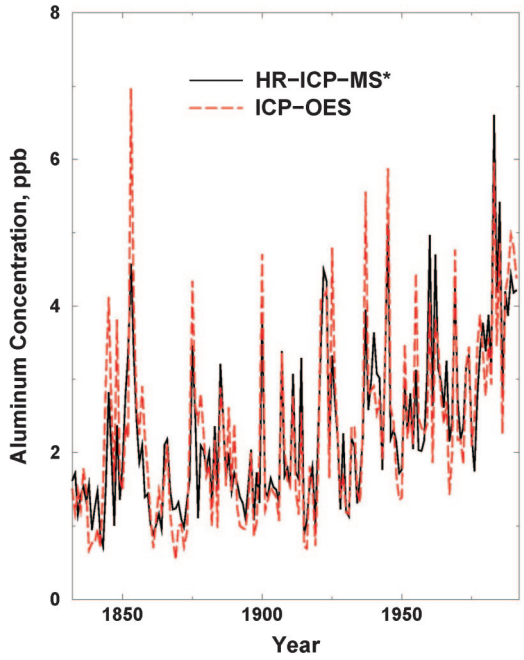

Fig. 5. Comparison of annually averaged aluminum concentrations at James Ross Island measured by HR-ICP-MS and ICP-OES. The HR-ICP-MS* concentrations were scaled by 1.22 to account for an estimated 82% recovery efficiency (see text). Similarly computed recoveries for more soluble species measured by both HR-ICP-MS and ICP-OES were 96%, 100%, 99%, and 96% for Na, Mg, Ca, and Sr, respectively.

Note that the summer to autumn minimums in aluminum concentration in the James Ross Island ice core approach the detection limit of aluminum on the ICP-OES, resulting in a lower correlation coefficient. To account for the 82% recovery efficiency, all of the HR-ICP-MS measurements of aluminum were scaled by 1.22. After mapping from core depth to year, the mean annual aluminum concentration during the 160-year record was ≈2.4 ppb with a mean difference in the annual average measured by ICP-OES and scaled HR-ICP-MS of 0.02 ppb and a standard deviation of 0.51 ppb (Fig. 5). Similarly computed recoveries for more soluble species measured by both HR-ICP-MS and ICP-OES were 96%, 100%, 99%, and 96% for Na, Mg, Ca, and Sr, respectively.

Dating. Although nearly all of the elements and chemical species measured on the James Ross Island ice cores show annual cycles in concentration that can be used to assist in dating the ice core record, here we primarily used measurements of sea salt tracers including chloride and sodium, together with non-sea-salt sulfur (nssS) and hydrogen peroxide. Percolation of surface melt water, particularly during summer, results in vertical mixing of the glaciochemical record. Little evidence for interannual mixing of the glaciochemical record at the Dalinger Dome core site was found, however, most likely because of the relatively low mean annual temperature (−14°C) and high snowfall rates (570 kg m^{-2} y^{-1}). Dating validation came from comparisons of nssS, water isotope, and other chemical concentration profiles with related records from the well-dated 1981 core (64.22°S, 57.68°W, 1,640 masl) from James Ross Island (35) and the 2001 ITASE01–5 (77.06°S, 89.13°W, 1246 masl) ice core from the base of the Antarctic Peninsula (43, 44). Uncertainty in the dating is estimated at ±1 year, although sulfur fallout from the well known 1883 Krakatoa and 1835 Cosinguina volcanic eruptions provided nearly unambiguous confirmation of the dating at 1884 and 1836, respectively (43, 45).

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