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Extraterrestrial accretion from the GISP2 ice core

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Abstract—The rate of extraterrestrial accretion for particles in the size range 0.45 μm to ~20 μm was determined from dust concentrates extracted from Greenland Ice Sheet Project 2 (GISP2) ice core samples. Using instrumental neutron activation analysis (INAA), we determined the iridium (Ir) content of the dust. Following a core-specific correction for terrestrial Ir and assuming a chondritic Ir abundance of 500 ppb, we measure an average accretion rate for 0.45 μm to ~20 μm particles over the entire Earth of 0.22 ($\pm$ 0.11) $\times$ 10^9 g/yr (kton/yr) for 317 years of ice through the interval 6 to 20 ka. This is consistent with the interplanetary dust accretion rate of 0.17 ($\pm$ 0.08) x 10^9 g/yr that we derive from published ^{3}He data for the GISP2 core. Accounting for particles that are larger and smaller than those detected by or experiment, our best estimate of the total accretion rate (including particle sizes up to about 4 cm in diameter) is 2.5 $\times$ 10^9 g/yr. The uncertainty in this estimate is dominated by statistical fluctuations in the number of particles expected to end up in the ice core and not by measurement error. Based on Monte Carlo simulations, we estimate the upper limit for total extraterrestrial accretion to Earth of 6.25 $\times$ 10^9 g/yr (95% confidence level). This accretion rate is consistent with some estimates from micrometeorite concentrations in polar ice, estimates from ground-based radar studies, and with accretion estimates of ^{3}He-bearing interplanetary dust particles, assuming that ^{3}He is correlated with particle surface area. It is, however, lower than estimates based on platinum group element studies of marine sediments. The conflict may indicate systematic errors with either the marine or the non-marine samples, departures from the assumed particle spectrum of Grün and coauthors, or time-variable accretion rates, with the early Holocene period being characterized by low levels of accretion. *Copyright © 2003 Elsevier Science Ltd*

1. INTRODUCTION

In this paper we present neutron activation analysis data on dust concentrates filtered from six 1 to 2 kg samples of the Greenland Ice Sheet Project 2 (GISP2) ice core, drilled at the Summit site, 72.6° N, 38.3° W. A detailed review of the GISP2 core is given in Mayewski et al. (1997). From these neutron activation analysis data we estimate the extraterrestrial accretion rate to Earth. Our samples include extraterrestrial particles in the range 0.45 μm to ~20 μm. The lower limit was set by the pore size of the filters, and the upper limit was set by the mass of Ir that we attribute to extraterrestrial sources, assuming a chondritic Ir abundance of 500 ppb. Larger particles are not expected to occur in these samples because of the low probability of capturing one with the limited area and time encompassed by our samples.

To estimate total accretion, we must correct for the particles outside of this narrow size range, and to do this requires an estimate for the shape of the particle size distribution. Despite this limitation, our measurements have sensitivities that are better than obtained from previous studies of Ir in ice cores, and yield accretion estimates that are consistent with estimates that we calculate from ^{3}He data from glacial dust obtained by Brook et al. (2000). The high sensitivity that we obtained is a consequence of the low background Ir level in our instrument, long duration irradiations, long gamma ray counting times, and considerable attention to eliminating laboratory contamination.

While it is believed that the bulk of extraterrestrial mass on the Earth comes from large impact events (Ceplecha, 1992), such events are rare. Over time periods of decades to millennia, which is a reasonable period of time for considering the role of accretion in climate change, the accretion has a higher probability of being dominated by interplanetary dust particles (IDPs). Such objects are believed to originate from comets and from collisions among asteroids. Measurements suggest that the IDP accretion rate is not constant, but can vary over time periods as short as a few thousand years (Farley and Patterson, 1995) to millions of years (Kyte and Wasson, 1986).

The rate of extraterrestrial accretion has been measured in space-based studies (Grün et al., 1985; Love and Brownlee, 1993), high altitude atmospheric aerosol collection (Helmer et al., 1998), measurements with ground-based radar (Mathews et al., 2001), and measurements from geologic and cryospheric samples based on Ir (Barker and Anders, 1968; Kyte and Wasson, 1986; Esser and Turekian, 1988; Rocchia et al., 1990; Zhou and Kyte, 1992; Rasmussen et al., 1995), Os isotopes (Esser and Turekian, 1988; Peucker-Ehrenbrink, 1996; Peucker-Ehrenbrink and Ravizza, 2000), and He isotopes (Ozima et al., 1984; Takayanagi and Ozima, 1987; Farley, 1995; Farley and Patterson, 1995; Farley et al., 1997; Patterson and Farley, 1998; Marcantonio et al., 1996, 1998, 1999; Brook et al., 2000). A book was recently published on the subject by Peucker-Ehrenbrink and Schmitz (2001). Each type of material used to study accretion has its strengths and weaknesses. For instance, condensed ocean sediment sections may provide reasonable average accretion rate estimates over tens of Myr (e.g., Peucker-Ehrenbrink, 1996), but may not provide submillennial

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Table 1. Accretion rate estimates for Earth.

Measurement	Time interval	Particle size range	Measured Flux[a] ($\times 10^9$ g yr^{-1})	Estimate of true flux[b] ($\times 10^9$ g yr^{-1})	Reference
Polar ice samples					
Ir in Greenland dust	6021–11,458 YBP	>0.45 μm dust	0.22 ± 0.11	0.17 to 6.25 (95% CL)	This paper
Ir in Antarctic dust	1904–1912 AD	>0.45 μm dust	400	—	Ganapathy (1983)
Ir in Greenland dust	14–20 ka	>0.45 μm dust	5,000 to 30,000	—	LaViolette (1985)
Ir in Antarctic dust	1900–1930 AD	>0.45 μm dust	10	—	Rocchia et al. (1990)
Ir in Greenland dust	1905–1914 AD	>0.45 μm dust	14[c]	—	Rasmussen et al. (1995)
Greenland ice cosmic spherules	0–2 ka	>50 μm dust	4	—	Maurette et al. (1987)
Antarctic ice cosmic spherules	1500–1000 AD	>53–700 μm spherules	1.6 ± 0.3	~40[e]	Taylor et al. (1998)
^{3}He in Antarctic dust	3.8, 75, 97 ka	>0.2 μm, >0.45 μm dust	0.21 ± 0.08	—	Brook et al. (2000)
^{3}He in Greenland dust	1560–1590 AD	>0.2 μm, >0.45 μm dust	0.17 ± 0.08	—	Brook et al. (2000)
Satellite and atmospheric samples					
Satellite, radio and visual meteors	present	>0.20 μm particles	4.2 to 16.1	4.2 to 16.1	Hughes (1978)
Satellite, zodiacal light, lunar samples	present	impact craters	14.60	14.60	Grün et al. (1985)
Atmospheric Ir at South Pole	present	aerosols	6 to 11	6 to 11	Tuncel and Zoller (1987)
Long Duration Exposure Facility (LDEF)	present	microimpact craters	40 ± 20	40 ± 20	Love and Brownlee (1993)
Radar micrometeors	present	1–200 μm micrometeors	1.6 to 2.3	—	Mathews et al. (2001)
LDEF re-estimate	present	microimpact craters	—	1.8	Mathews et al. (2001)
Oceanic sediments					
Ir and Os in Pacific clay	0–100 ka	bulk sediment	50 ± 25[d]	50 ± 25	Barker and Anders (1968)
Pacific clay magnetic spherules	Pleistocene	magnetic spherules	0.09	—	Murrell, et al. (1980)
Ir in pelagic clay	33–67 Ma	bulk sediment	78	78	Kyte and Wasson (1986)
Os in Pacific clay	30–270 ka	bulk sediment	49 to 56	49 to 56	Esser and Turekian (1988)
Os in Pacific clay	30–270 ka	bulk sediment	29 to 38	29 to 38	Esser and Turekian (1993)
Os in Pacific sediment	0–80 Ma	bulk sediment	37 ± 13	37 ± 13	Peucker-Ehrenbrink (1996)
Os in Pacific sediment	0–80 Ma	bulk sediment	30 ± 15	30 ± 15	Peucker-Ehrenbrink and Ravizza (2000)
^{3}He in Pacific sediments	0–40 Ma	<10μm	0.4 ± 0.2	—	Takayanagi and Ozima (1987)
^{3}He in Pacific sediments	0–1 Ma, 1–70 Ma	bulk sediment	0.3, 0.2	—	Farley (1995)
^{3}He in Atlantic sediments	250–450 ka	bulk sediment	0.29	—	Farley and Patterson (1995)
^{3}He in Pacific sediments	0–200 ka	bulk sediment	0.2 ± 0.1	~40[e]	Marcantonio et al. (1996)
^{3}He in Atlantic sediments	0–50 ka	bulk sediment	0.3 ± 0.1	~40[e]	Marcantonio et al. (1998)
^{3}He in Pacific sediments	0–700 ka	bulk sediment	0.30	—	Patterson and Farley (1998)
^{3}He in Indian Ocean sediments	0–200 ka	bulk sediment	0.3 ± 0.1	~40[e]	Marcantonio et al. (1999)

[a] The actual measurement made by each reference, scaled to an entire Earth area. It does not include corrections for potential ablative loss He data is converted to mass using ^{3}He concentrations of 1.9×10^{-5} cm^3 STP g^{-1} of IDP (see text).
[b] For references that model the true accretion considering ablative loss, undersampling of large particles, or other physical parameters (see text). — Indicates that no estimate was given.
[c] Errors were improperly calculated in original publication; standard deviation of measurements shows that the error in the accretion estimate should equate to $\pm 4 \times 10^{-9}$ g yr^{-1} (1 σ). Measurement should be considered an upper limit only (see text).
[d] Recalculated here with Ir presumed to be 0.5 ppm by weight.
[e] These authors assumed the true accretion rate is that published by Love and Brownlee (1993) and/or Peucker-Ehrenbrink (1995).

time scale resolution which is necessary to investigate possible links of accretion with climate change. Conversely, ice core samples can provide annual resolution, but do not extend beyond several hundred kyr. Accretion rates reported in studies are shown in Table 1. A thorough review of earlier work is in Peucker-Ehrenbrink (1996).

As can be seen in Table 1, the results of these studies are not in obvious agreement; similar experiments yield estimates of accretion that differ by several orders of magnitude. Additionally, in many cases the measured quantity of material is interpreted to represent a minute fraction of the total accretion, and extrapolation to the whole-Earth accretion rate and/or full size spectrum of accreted particles requires a reliance on model-based assumptions. Thus, there is some difficulty relating these accretion estimates to one another.

Farley et al. (1997) and Peucker-Ehrenbrink and Ravizza (2000) present model results which suggest that much of the disagreement in estimated accretion rates from ice core, ocean core, and satellite measurement studies is due to the effective sampling area and time encompassed by each type of sample (the "area-time" product, Farley et al., 1997). There are a discrete number of accreted particles of every size, and so the sampling of shorter time segments reduces the likelihood of recording rare, large particles. Comparison of accretion data from different experiments, without consideration of the area-time product encompassed by each, could be misleading.

Farley and Patterson (1995) and Patterson and Farley (1998), used ^{3}He in ocean sediment samples to track interplanetary dust accretion. They found a quasi-100 kyr periodicity, with the accumulated ^{3}He varying by a factor of two. This supported a prediction of Muller and MacDonald (1995) that the dust would match the fundamental 100 kyr cycle of the ice ages. However, Marcantonio et al. (1996) argue that the 100 kyr variations recorded by Farley and Patterson (1995) are effects of sediment focusing rather than a true change in accretion rate, so whether accretion does vary with a quasi-100 kyr periodicity remains disputed.

The sudden terminations of glaciation periods are not adequately explained by any of the astronomical ice age theories (Muller and MacDonald, 2000). These terminations may simply be a nonlinear response to slow warming. A goal of the present experiment is to study the extraterrestrial dust accretion rate during the last glacial termination, and see if there is any indication that dust played a role in this event. Ice cores provide some of the most detailed records of the last termination. Measuring extraterrestrial accretion in ice represents a technical challenge, since the concentration of such dust is exceedingly small. Ir contamination is a serious problem, and ultra-clean-room techniques must be used. On the plus side, background Ir from terrestrial dust is much lower in ice samples than in ocean sediment.

Ablation of large extraterrestrial dust particles (>50 μm) during atmospheric entry is thought to reduce a significant fraction of the mass to nanometer-scale "smoke" particles (Hunten et al., 1980). The reduction in mass is estimated to be 70 to 90% by Love and Brownlee (1991), 80 to 85% by Herzog et al. (1999) and 78% by Cziczo et al. (2001). Ablated extraterrestrial dust particles are a primary particle source for the upper atmosphere, and they provide seed nuclei for noctilucent clouds (Fiocco and Grams, 1971; Toon and Farlow, 1981; Turco et al., 1982; Keesee, 1989; Blix et al., 1995). These particles could also provide reactive surfaces that scavenge ozone from the stratosphere. Muller (1994) suggested an effect of extraterrestrial dust on climate could be through the modulation of cloud cover.

2. MATERIAL AND METHODS

Ice core dust samples were prepared on a class 100 clean bench located in a clean room in the Physics Department at the University at Buffalo that had been constructed specifically for the purpose of analyzing ice core samples in an ultra-clean environment. A section of the GISP2 ice core, 3×3 cm in cross section, was used for dust concentration experiments as part of the original GISP2 work (Ram et al., 1995; Ram and Koenig, 1997; Ram et al., 2000). This core section has been stored at the University at Buffalo, and segments of this ice were used for the present experiment. Time resolution of the samples is based on annual layer counting (Alley et al., 1998). While the precise age of each sample may not be known, the duration encompassed by each sample is known with high accuracy.

Each sample consists of dust filtered from two- or four-meter long sections (about one to two liters) of melted ice. Before filtration, ice core samples were cut into 10-cm long sections that were individually cleaned by rinsing all surfaces in ultrapure Milli-Q water. The ice was then melted and the dust was filtered onto 13 mm diameter, 0.45 μm pore size, type HA Millipore filters, composed of mixed cellulose acetate and nitrate. The filtration apparatus was specially constructed for our experiment from Plexiglas to avoid contact of either meltwater or filter with stainless steel parts, which could introduce Ir contamination (see Rocchia et al., 1990). Filtration was carried out in a system that was pressurized with ultra-pure argon to expedite the passage of large volumes of meltwater through the small diameter filter membrane.

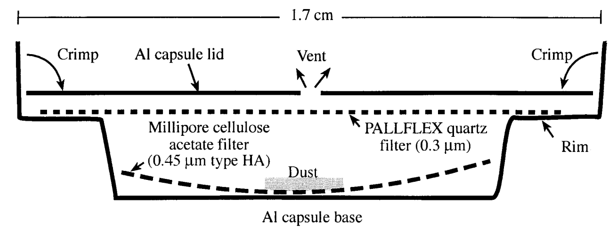

Fig. 1. Sample capsule. The glacial ice meltwater is filtered through a cellulose acetate filter. The filter and the dust retained on it are placed in a high purity (99.9999%) Al capsule and covered with high-purity quartz filter prior to sealing with an Al lid. A small hole is drilled through the lid to allow exhaust to escape during sample heating. This encapsulation procedure ensured that no dust escaped the capsule prior to analysis.

The filters with dust were then prepared for instrumental neutron activation analysis (INAA) on the class 100 clean bench. INAA differs from radiochemical neutron activation analysis in that no chemistry is performed on the sample. A diagram showing the capsule and its components is in Figure 1. Neutron irradiation produces sufficient heat to combust the cellulose acetate filters. To avoid this, the samples were heated in an oven to decompose the filters. This was done in a closed capsule to avoid loss of dust with the combustion exhaust. The cellulose acetate filters with the dust were placed in high purity (99.9999%) Al capsules, covered with high purity quartz filters (Pall-Gelman Tissuquartz filters, lot no. 53693, which retain 99.9% of particles down to 0.3 μm) and sealed with a high purity Al lid. A small hole had been drilled through the lid to allow gasses to escape as the cellulose acetate filter decomposed. The sealed capsules were baked at approximately 170°C for 12 h to decompose the cellulose acetate filters and eliminate water from the glacial dust (low water reduces the likelihood of capsule failure during irradiation).

We additionally prepared Al capsule blanks, quartz filter blanks and procedural blanks. Procedural blanks were prepared between each ice core sample, in which Milli-Q water rather than glacial meltwater was passed through cellulose acetate filters. Following filtration of the Milli-Q water, these cellulose acetate filters were packaged and analyzed identically to the filters with glacial dust.

Despite this effort, we were still concerned that some Ir might have been lost with the exhaust gas during the heating and decomposition of the filters. However, our measurements showed that little glacial dust transferred to the quartz filter, and the dust that did transfer was not enriched in Ir when compared with the dust that remained on the cellulose acetate filter. This indicates that no preferential loss of extraterrestrial material occurred during the decomposition process. Future experiments could be improved by the omission of the quartz filter, as the measurement uncertainties introduced by the presence of the quartz filter are potentially larger than the loss of Ir during sample heating. A slow burn of the cellulose acetate filter can be accomplished within a sealed Al capsule. This improvement in sample preparation will reduce the time required for gamma ray counting; it will eliminate the introduction of uncertainty due to heterogeneous Ir concentrations in the quartz filters, and from the potential loss of dust during sample handling.

After heating, the sample capsules were then stacked vertically inside quartz tubes with geometrically arranged standards, including the DINO-1 Stevns Klint Cretaceous-Tertiary boundary clay (Alvarez et al., 1982), and standard pottery (Perlman and Asaro, 1969, 1971). Standard Pottery was used to calibrate the abundances of most elements, and the DINO-1 standard, whose Ir concentration is known to

±2.0% (Alvarez et al., 1982) was used to calibrate the Ir measurements. Capsules containing the standard pottery and DINO-1 standards were place at the top and bottom of the quartz tube. This arrangement permitted the interpolated calibration for each sample position. The quartz tubes were sealed under vacuum before irradiation. They were then exposed to a neutron flux of 8 x10^{13} n/cm^{2}sec at the University of Missouri reactor for periods of 4 or 16 d (irradiations M29 and M30, respectively). After irradiation, the samples were brought to the Lawrence Berkeley National Laboratory. The quartz tubes were opened and the Al capsules were removed. All exterior surfaces of the Al capsules were cleaned with ethyl alcohol and cotton applicators to remove loosely bound radioactive particles. Analysis of one of these cotton applicators indicated that the radioactivity removed was from trace impurities in the capsule Al.

The samples were then analyzed in the Luis W. Alvarez Iridium Coincidence Spectrometer (LWAICS). The LWAICS enabled instrumental measurements of Ir abundances below 25 ppt to be made 500 times faster than was previously possible with single detectors at the Lawrence Berkeley National Laboratory. Its operation has been briefly described (Asaro et al., 1987, 1988; Alvarez et al., 1988) and in more detail in Michel et al. (1990). The LWAICS consists of a pair of 5 cm diameter by 5 cm length intrinsic Ge detectors, each of which measures the gamma ray spectra from the samples. The LWAICS uses two Princeton amplifiers and dual 500 KHz ADAM 826 to 1 analog to digital converters. Nearly all components of the LWAICS have been modified in some fashion at LBNL.

The isotope ^{192}Ir has two near-coincident gamma rays with energies of 468.07 keV and 316.51 keV. By requiring the near-coincident detection of these gamma rays (one in each detector, with a coincidence window of 100 ns), the background is significantly reduced. Spurious counts of Ir are common and must be monitored during the measurements. For example, an irradiated nominal sample with 40000 gamma ray counts-per-second in each of the two detectors (the most efficient rate for coincidence measurements) gives 70 counts per hour of spurious Ir coincidences, due in large part to Compton interactions of high energy coincident gamma rays of both ^{46}Sc and ^{60}Co. Fortunately, such Compton interactions produce lower energy scattered radiation, which can be shielded from the detectors. For this purpose two 12 to 15 inch thick scintillator-doped mineral oil anti-Compton shields that nearly surround the Ge detectors can detect and veto approximately 90% of the scattered radiations associated with the spurious Ir coincidences. A background of remaining spurious coincidences is estimated by coincidences between the 316.5 keV region in one Ge detector and a 503 KeV region in the other detector.

Fast coincidences were measured with all fast output from the two Ge detectors and slow coincidences were measured with windows for the gamma ray peaks and background regions of interest. When useful (like for Ir), coincidence measurements were also taken between the fast and slow outputs and usually anti-coincidences were taken between the combined output of the two shields and the singles or coincidences from the Ge detectors. At a count rate of 8400 counts per second, the half width of the ^{60}Co 1332 keV peak was 2.1 keV for single gamma ray detections, which were made using the "B" detector. The corresponding half width of the "A" detector (which is also used for coincidence measurements) was 2.9 keV for the ^{60}Co 1332 keV peak. The typical count rate for the cellulose acetate filter plus dust fractions of the GISP2 samples was ~1650 counts sec^{-1}, and yielded slightly better resolution than was obtained at 8400 counts sec^{-1}. The sample with the presumed large terrestrial dust component (M30-25), however, had 33500 counts per second when it was last measured ~16 weeks after the end of the irradiation, and so had slightly worse resolution.

The detector efficiency was determined from the ratio of pulser counts that passed through the detector in the assigned energy window to the pulser input counts. The single gamma ray detection efficiency for the cellulose acetate filter plus glacial dust samples was typically ~98% and the coincidence efficiency was ~97%.

The instrument used measures 18 elements in addition to Ir, and for the six best-measured elements has a reproducibility of 0.3% (Asaro et al., 2002). Some elements are poorly measured and those data are not presented here. The measurement precision for Fe, determined by comparison with five standards of the National Institute of Standards and Technology (NIST), was previously measured at 0.3 to 0.5%. The Fe abundances determined versus Standard Pottery were uniformly 3% higher than those determined versus the NIST standards. The elements measured in addition to Ir enabled us to track each component of the capsule (Al, quartz filter, cellulose acetate filter, and glacial dust), and to isolate the GISP2 dust component.

The gamma ray counting experiment began approximately one month after the irradiation, and continued for several months thereafter. The gamma ray counting time averaged ~93 h per sample. Samples were first analyzed for several hours each by counting while they were within the irradiated Al capsules. This step ensured that all contents of the capsules were accounted for at the start of gamma ray counting. These short analyses indicated that most of the radioactivity was from the irradiated Al capsules rather than from the capsule contents. Since the rate of spurious counts of ^{192}Ir is roughly proportional to the total count rate of ^{46}Sc in the sample, the contents of the Al capsules were separated to reduce the spurious counts due to the capsule Al (which contained ^{46}Sc as an impurity). The capsules were opened and the glacial dust with cellulose acetate filter residues were transferred into unirradiated Al capsules for long duration (several days) gamma ray counting. Transfer yields of the glacial dust with cellulose acetate filter residues (measured by tracking Cs) were typically better than 90%. For each sample, the irradiated Al and quartz filter were also analyzed separately to track the glacial dust and also to quantify the Ir that was associated with these other components. The preliminary measurement in conjunction with measurements on each of the separated capsule components enabled us to determine the amount of Al, quartz filter, cellulose acetate filter and ice core dust that was in each. Procedural blanks were also divided into Al, quartz filter and cellulose acetate filter components, and each of these components was analyzed to quantify, and subtract, their individual contributions of Ir and other elements from the GISP2 dust samples.

Although the measurements on the cellulose acetate filter with dust and quartz filter components ran for many thousands of minutes, much shorter times sufficed for the other separated components. The need to evaluate the trace element content in each of the separated capsule components increased the uncertainty in the Ir contents of the glacial dust, so that the final uncertainty was ~2 to 4 femtograms of Ir for each dust sample. The abundance of Ir in the quartz filters was assumed to be constant per unit weight of filter. The cellulose acetate filter, which could have contained residual water when weighed, was assumed to have a constant mass of Ir per filter. Because they were easily separated from the other components, the quartz filters contributed very little to the Ir detected in the ice core fractions. In contrast, the cellulose acetate filter residue was mixed with the glacial dust, and contributed ~25% of the Ir mass. The average terrestrial Ir contribution deduced for the four ice core samples in the last irradiation (excluding the high Fe sample used for estimating the terrestrial background) was slightly lower than half of the total Ir measured in the GISP2 dust.

3. RESULTS

Table 2 shows the abundances of Ir plus 12 other elements measured by the INAA experiment for the six dust samples. Table 3 shows the Ir content of the dust deduced from INAA analysis and the deduced extraterrestrial accretion rates. Data for the Al, quartz filters, cellulose acetate filters, and background for the LWAICS are reported in the Appendix. Analysis of the Al, quartz and cellulose acetate filter components showed that certain elements track (primarily) one component of the sample package, whereas only a few percent occur in the other components. In particular, Sc occurs as a trace element in the Al capsule, Hf occurs in the quartz filter, Ta occurs with the cellulose acetate filter, and Cs occurs with the glacial dust. We used the abundances of these elements in the individual components relative to the original measurements made in the irradiated Al capsules to model each component during the dividing procedure, and to quantify losses during sample handling.

After subtracting the non-dust components, we distinguish the terrestrial dust from the extraterrestrial dust in our samples.

Table 2. Instrumental neutron activation analysis data.[a]

Sample Number	Depth (m)	Core length (cm)	Age (Years B.P.)	Dust Weight ($\times 10^{-3}$ g)[b]	Ir (Coincidence) ($\times 10^{-15}$g)	Ta (68 keV) ($\times 10^{-9}$g)	Co (1332 keV) ($\times 10^{-9}$ g)	Fe (1099 keV) ($\times 10^{-6}$ g)	Sc (889 keV) ($\times 10^{-9}$ g)	Sb (1691 keV) ($\times 10^{-9}$ g)
M30-19	1131–1135	400	6021–6052	0.171	15.9 ± 3.9	1.91 ± 0.36	26.1 ± 3.3	8.55 ± 0.28	32.5 ± 1.1	4.15 ± 0.52
M30-21	1360–1364	400	7922–7959	0.053	8.9 ± 2.2	0.56 ± 0.32	4.5 ± 3.2	2.64 ± 0.16	10.1 ± 0.6	2.09 ± 0.28
M30-23	1561–1565	400	9957–10003	0.078	4.2 ± 2.8	1.46 ± 0.42	9.3 ± 2.7	3.88 ± 0.43	15.4 ± 1.2	3.13 ± 0.34
M30-27	1663–1667	400	11389–11458	0.080	8.2 ± 2.4	1.63 ± 0.21	9.1 ± 2.4	4.00 ± 0.22	14.5 ± 0.6	2.68 ± 0.24
M29-09	1364–1368	400	7959–7993	0.046	6.0 ± 3.6	0.79 ± 0.30	5.8 ± 3.1	2.31 ± 0.16	7.9 ± 0.6	1.40 ± 0.29
M30-25	1934–1936	200	20583–20683	2.446	146.2 ± 14.2	27.40 ± 0.80	444.0 ± 8.0	122.30 ± 0.80	466.0 ± 7.0	37.40 ± 0.80

Sample Number	Cs (796keV) ($\times 10^{-9}$ g)	Eu (Coincidence) ($\times 10^{-9}$ g)	Hf (Coincidence) ($\times 10^{-9}$ g)	Th (312 keV) ($\times 10^{-9}$ g)	Cr (320 keV) ($\times 10^{-9}$ g)	Ni (811 keV) ($\times 10^{-9}$ g)	Rb (1077keV) ($\times 10^{-9}$ g)
M30-19	19.50 ± 0.60	2.22 ± 0.24	7.7 ± 3.1	28.1 ± 2.8	446 ± 110	149 ± 26	207 ± 10
M30-21	8.39 ± 0.37	0.79 ± 0.12	2.3 ± 1.6	11.4 ± 1.5	191 ± 58	78 ± 22	76 ± 5
M30-23	12.30 ± 0.80	1.31 ± 0.13	4.4 ± 0.7	16.8 ± 1.4	139 ± 90	65 ± 36	128 ± 12
M30-27	10.80 ± 0.40	1.16 ± 0.10	2.6 ± 0.9	11.9 ± 1.0	169 ± 81	91 ± 15	108 ± 7
M29-09	8.60 ± 0.50	0.66 ± 0.15	1.2 ± 1.0	9.4 ± 1.1	174 ± 87	38 ± 23	76 ± 6
M30-25	230.80 ± 1.80	36.56 ± 0.26	111.7 ± 0.8	374.0 ± 1.0	2844 ± 127	1630 ± 133	2950 ± 45

[a] Elemental abundances corrected to 100% yield, after corrections for instrumental background, quartz and cellulose acetate filters, and A1 capsule.
[b] Dust weights estimated from Fe by assuming dust is 5% Fe by weight.

To set an upper limit for extraterrestrial dust content, we could assume that all the Ir is extraterrestrial. By doing so we calculate accretion rates that vary from 0.2 to 3.0 $\times$ 10^9 g/yr for our samples. However, we believe that a significant terrestrial Ir correction must be applied to these data. The terrestrial correction is commonly made using the average crustal Ir/Fe ratio measured by Fenner and Presley (1984), who analyzed Mississippi Delta sediment by INAA and their data correspond to an Ir/Fe ratio of 1.4 $\times$ 10^{-9}. Had we used this ratio, there would have been insufficient Ir to account for the terrestrial component, and no Ir attributable to extraterrestrial sources in our samples. Therefore we conclude that the Mississippi Delta sediment estimate is not appropriate for the GISP2 terrestrial dust source.

An alternative correction for terrestrial Ir could be made using the Ir content of loess, which is a primary eolian dust component during glacial periods. Loess that had been collected at many different locations around the world was determined recently by Peucker-Ehrenbrink and Jahn (2001) to have an average Ir/Fe ratio of 6.3 $\times$ 10^{-9}, approximately half that of Mississippi Delta sediment. However, had we used this loess Ir/Fe ratio, we would have derived an extraterrestrial Ir signal from the GISP2 dust samples that was positively correlated with total dust content. Rather than interpreting the data to show a correlation with total dust and total accretion, we prefer the simpler explanation that such a correlation is caused by the underestimation of the terrestrial Ir/Fe ratio.

To estimate an Ir/Fe ratio that is perhaps more appropriate for the GISP2 terrestrial dust, we plot Ir versus Fe (Fig. 2), and fit a line to these data by χ^2 minimization. By doing so we make the assumption that all six samples can be represented by a single accretion rate and a single terrestrial Ir/Fe ratio. We interpret the slope of this line (Ir/Fe=1.10 ($\pm$ 0.18) x 10^{-9}) to be the Ir/Fe ratio of the terrestrial dust. The fact that this slope is between those for Mississippi Delta sediment and loess suggests that the correction factor we derive is reasonable, and may reflect the mixing of loess and some other terrestrial dust source (a 3:1 mixture of loess and Mississippi Delta sediment could account for the Ir/Fe ratio we derive). We interpret the positive Ir intercept of 2.2 ($\pm$ 1.1) x 10^{-17} g/cm^2 yr in Figure

Table 3. Estimated chondritic-equivalent accretion rate of 0.45–20 μm dust from the GISP2 core.

Sample	Depth (m)	Core length (cm)	Age (years before present)	Duration (years)	Ice mass (g)	Sample area (cm^2)[a]	Ir content ($\times 10^{-15}$ g)	±	Fe content ($\times 10^{-6}$ g)	±	Terrestrial Ir ($\times 10^{-15}$g)[b]	±	Cosmic Ir ($\times 10^{-15}$g)	±	Accretion rate ($\times 10^9$g/yr)[c]	±
M30-27	1131–1135	400	6021–6052	31	1402.5	3.853	8.2	2.4	4.00	0.22	4.4	0.8	3.8	2.5	0.32	0.21
M30-23	1360–1364	400	7922–7959	37	1711.4	4.702	4.2	2.8	3.88	0.43	4.3	0.8	−0.1	2.9	0.00	0.17
M30-21	1561–1565	400	9957–10003	46	1647.3	4.526	8.9	2.2	2.64	0.16	2.9	0.5	6.0	2.3	0.29	0.11
M30-19	1663–1667	400	11389–11458	69	1579.1	4.338	15.9	3.9	8.55	0.24	9.4	1.6	6.5	4.2	0.22	0.14
M29-09	1364–1368	400	7959–7993	34	1720.0	4.725	6.0	3.6	2.45	0.14	2.7	0.5	3.3	3.6	0.21	0.23
M30-25	1934–1936	200	20583–20683	100	899.7	4.943	146.2	14.2	122.30	0.80	137.0	22.4	9.2	26.6	0.19	0.55
													Best Value[d]		0.22	0.11

[a] Assumes an ice density of 0.91 g/cm^3.
[b] Terrestrial Ir correction is Fe $\times$ 1.10 ($\pm$ 0.18) $\times$ 10^{-9}.
[c] Assumes Ir = 0.5 ppm, area Earth = 5.1 $\times$ 10^{18}cm^2.
[d] Based on χ^2 minimization, error is 1σ.

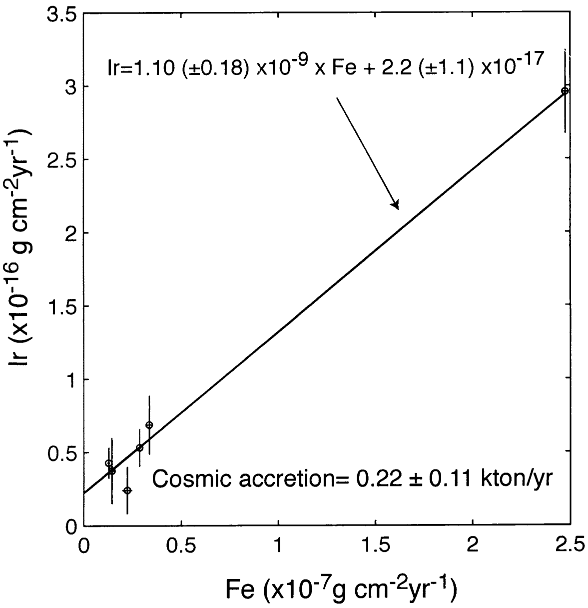

Fig. 2. Plot of Fe versus Ir in the glacial dust for the six ice samples. The regression line is based on the χ^2 minimization of the six Ir values. The straight-line fit of the data suggests a constant Ir/Fe ratio in the terrestrial dust component, and the positive Ir intercept is taken to be the level of extraterrestrial Ir. The extraterrestrial component yields a chondritic accretion rate for particles in the size range 0.45 to ~20 μm of 0.22 ($\pm$0.11)$\times 10^9$ g/yr. The slope of the line indicates an Ir/Fe ratio of 1.1×10^{-9}, significantly lower than the value 1.4×10^{-9} calculated for Mississippi Delta sediment (Fenner and Presley, 1984), a commonly used estimate for terrestrial Ir.

2 to be the average extraterrestrial Ir accretion rate. This positive Ir intercept occurs when the Ir abundance is plotted against the Sc, Cs, Co, and Th abundances as well, showing that the presence of extraterrestrial Ir is not an artifact of the choice of Fe as our comparison element. From the Ir intercept, and assuming uniform deposition of extraterrestrial material over the Earth, we obtain an average global accretion rate for 0.45 μm to ~20 μm chondritic particles of 0.22 ($\pm$ 0.11) x 10^9 g/yr for the 317 yr recorded by our samples (Table 3).

We note that the assumption of a constant Ir/Fe ratio in the terrestrial dust and a constant extraterrestrial accretion rate for the six samples could be in error, particularly for sample M30-25. Sample M30-25 is from the last glacial period and contains 10 to 40 times more dust (and Ir) than do the Holocene age samples (Table 4), a consequence of high terrestrial dust loading of the atmosphere during that time. Sample M30-25 has a strong influence on the slope of the regression line used to estimate the terrestrial Ir/Fe ratio. If the slope of this line were less steep, then more Ir in the samples could be attributed to extraterrestrial dust. We cannot rule out this possibility with the limited data presented here. Had we fit a line to only the five Holocene age samples to predict the terrestrial Ir component in sample M30-25, the terrestrial Ir/Fe ratio would be 0.88 ($\pm$ 1.12) x 10^{-9}, which is poorly constrained but consistent with the ratio that included sample M30-25. Therefore, the assumption of a constant Ir/Fe ratio in the terrestrial dust may be valid, but requires further testing.

Our measured total dust content in the core by INAA and by Coulter counter tests conducted at the University at Buffalo (Table 4) are consistent with each other and with other measurements of the ice core dust content (Zielinski et al., 1997). This increases our confidence that our samples accurately represent the >0.45 μm fraction of dust in the GISP2 core.

3.1. Estimation of Total Accretion Rate

To estimate the total accretion rate, we must correct for extraterrestrial particles with diameters that are outside the range covered by our samples (0.45 to ~20 μm). We do this by using the particle size distribution of Grün et al. (1985), which was based on early spacecraft studies, analysis of zodiacal light scattering, and microcrater measurements on returned lunar samples. The correction is large, since large particles with diameter greater than 200 μm are expected to dominate the mass flux, and yet are so rare that we found no such particles in our samples. The six samples, when combined, equal 317 yr of accumulation over an area of 4.59 cm^2, or equivalently, an area-time product of 0.146 m^2yr. Based on our extensive Monte Carlo simulations (discussed below), we estimate that this area-time product has only a 5% probability of capturing a particle with diameter 100 μm or greater; in fact, we found no particles of this size. This undersampling bias has been discussed previously by Farley et al. (1997), and by Peucker-Ehrenbrink and Ravizza (2000).

We have performed extensive Monte Carlo simulations to determine from our measurements the best estimate for the total extraterrestrial flux and the 95% confidence limits. We assume no ablative loss of particles in the range we observe (0.45 to

Table 4. Comparison of Coulter counter and INAA dust estimates.

Sample	Depth (m)	Age (Years before present)	Ice wt. (g)	Duration (years)	Sample area (cm^2)[a]	Fe wt. from INAA ($\times10^{-3}$g)	Dust wt. assuming Fe = 5% of INAA ($\times10^{-3}$g)	Dust wt. from Coulter counter ($\times10^{-3}$g)	INAA/Coulter counter (%)[b]
M30-27	1131–1135	6021–6052	1402.5	31	3.853	0.0030	0.0604	0.0599	101
M30-23	1360–1364	7922–7959	1711.4	37	4.702	0.0022	0.0440	0.1036	42
M30-21	1561–1565	9957–10003	1647.3	46	4.526	0.0028	0.0564	0.0971	58
M30-19	1663–1667	11389–11458	1579.1	69	4.338	0.0073	0.1454	0.2028	72
M30-25	1934–1938	20583–20683	899.7	100	4.943	0.1340	2.6800	1.7140	156

[a] Sample area is based on an estimated ice density of 0.91 g cm^{-3}.
[b] Differences of 50% could reflect the size range included in Coulter counter estimates, or the presumed 5% Fe crustal abundance.

~20 μm); this assumption is consistent with the ablation study by Farley et al. (1997). To find accretion rates that give a reasonable probability of our observed fluxes, we begin by picking a value for the total flux; we let this vary over 168 different values between 0.01 and 30 $\times$ 10^9 g/yr. The mass distribution is divided into 100 bins per decade for a total of 2000 mass bins. An inverse Poisson distribution around the mean is used to generate a random number for each bin in a given simulation. We performed 42000 simulations for each of the 168 possible accretion rates, for a total of over 7 million simulations. In the vast majority of the Monte Carlo simulations, the detected accretion rate (i.e., the sum of the masses in all the bins), is less than the true accretion rate. These low accretion rates are compensated for by infrequent, large particles, which account for most of the accreted mass. As a check on the procedure, we averaged all our simulations together, and obtained (as expected) the correct (assumed) rate.

Treating the Monte Carlo-generated data as we did our real measurements, we combined sets of six successive samples into a composite measurement of an (assumed constant) accretion rate, and compared these results with our measured value of 0.22 ($\pm$ 0.11) x 10^9 g/yr. Repeating our Monte Carlo simulations with different values of the accretion rate, we ask what fraction of the time an average of six samples with area-time products equal to that of our GISP2 samples yields an accreted mass that is equal to or less than our measurement. The highest modeled value that yielded an accretion rate less than or equal to our measurement 5% of the time is 6.25 $\times$ 10^9 g/yr. Thus, we conclude that 6.25 $\times$ 10^9 g/yr is the 95% confidence upper limit on the true accretion rate derived from our measurements. Similarly, we ask what fraction of the time the modeled accretion rate is equal to or greater than our measured value. The 95% confidence lower limit of the true accretion rate determined in this way is 0.17 $\times$ 10^9 g/yr. The confidence interval is illustrated graphically in Figure 3. The modeled accretion rate that is most likely to yield a measurement of 0.22 ($\pm$ 0.11) x 10^9 g/yr is 2.5 $\times$ 10^9 g/yr. Compared with our measurement of 0.22 ($\pm$ 0.11) x 10^9 g/yr, these results imply that we are missing approximately 92% of the extraterrestrial material because of undersampling.

We note that our Monte Carlo tests using the Grün et al. (1985) distribution were not subject to the ~20 μm upper limit of the extraterrestrial particles that we estimated for our samples. Rather, the Monte Carlo test only had the lower limit set at 0.45 μm; the upper limit was unconstrained. Thus, the accretion rate that was most similar to our measurements was based on the whole particle distribution of Grün and coauthors above 0.45 μm. Additionally, the effects of variable Ir contents in individual interplanetary dust particles had little effect on our Monte Carlo results. The Monte Carlo tests that were run included tens of particles in each, and for each mass bin we ran 42000 Monte Carlo tests. Because of this large sampling, the effects of variable Ir contents in individual particles, as was reported by Kurat et al. (1994) are minimized. Instead, rare particles with anomalously high or low Ir content have little influence on varying the Ir concentration from that of the chondritic value.

Our Monte Carlo simulations are sensitive to the shape of the size spectrum of extraterrestrial dust particles. Love and Brownlee (1993) derive a different size spectrum than do Grün

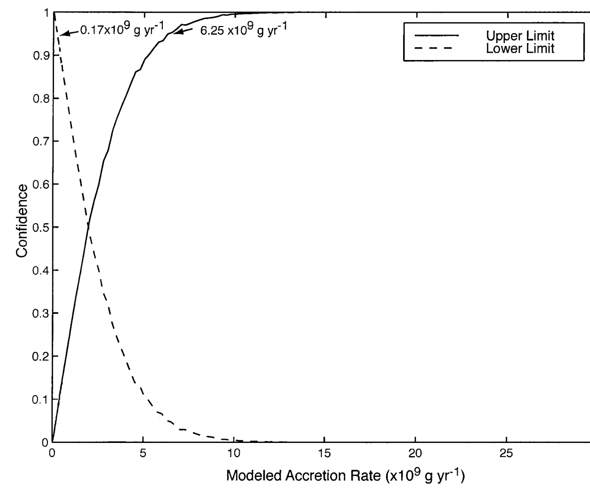

Fig. 3. Monte Carlo simulation results showing the upper and lower confidence limits for accretion from the GISP2 measurements. The Monte Carlo simulation utilized the particle distribution of Grün et al. (1985) to account for particles outside the size range 0.45 to ~20 μm. The 95% upper limit of accretion is 6.25 $\times$ 10^9 g/yr, and the 95% lower limit is 0.17 $\times$ 10^9 g/yr. The best estimate given our measurement is 2.5 $\times$ 10^9 g/yr, indicating that our samples contain approximately 8% of the accreted mass.

et al. (1985), based on their analysis of microcraters on the LDEF satellite. In particular, Love and Brownlee (1993) calculate twice as many 100 to 200 μm sized particles as do Grün and coauthors, even after their two curves are scaled to agree for the particle sizes measured in our experiment. If the Love and Brownlee (1993) curve were correct, our confidence limits and best estimate of the accretion rate would change by approximately a factor of two. Based on this approximation, our upper 95% confidence limit is increased from 6.25 $\times$ 10^9 g/yr to 12.5 $\times$ 10^9 g/yr, and our lower 95% confidence limit is increased from 0.17 $\times$ 10^9 g/yr to 0.34 $\times$ 10^9 g/yr. Using the Love and Brownlee curve, the modeled accretion rate that is most likely to yield our measurement of 0.22 ($\pm$ 0.11) x 10^9 g/yr is 5.00 $\times$ 10^9 g/yr. The Love and Brownlee results imply that we are missing 96% of the extraterrestrial material because of undersampling. However, as we discuss below, the Love and Brownlee (1993) distribution function may represent a systematic overestimate of particle masses because of an underestimation of impact velocity (Mathews et al., 2001), although the results from Mathews and coauthors are still under debate.

4. DISCUSSION

In this section we compare our results with previously published accretion estimates. Seven of these are based on ice core samples, five from atmospheric aerosol and satellite measurements, and 14 from deep-sea cores (Table 1). While there are other studies in the literature, we believe that comparison of our work with these 26 studies is sufficient to place our results into context; see Peucker-Ehrenbrink (1996) for a more complete survey of earlier work. We caution that these comparisons are not straightforward because each experiment has its own area-time product, has its own bias due to the methodologies for collection and analysis, and samples different time intervals.

The time interval studied may be particularly important if there is a time-varying accretion rate.

4.1. Comparison with Other Ice Core Samples

The first nine entries in Table 1 show estimates of accretion from polar ice samples. In addition to our work, four other studies have been made in the past two decades (Ganapathy, 1983; LaViolette, 1985; Rocchia et al., 1990; Rasmussen et al., 1995) in which the Ir content in ice cores was measured by filtering the meltwater to study dust particles with diameters >0.45 μm. These studies calculate the accretion to Earth based only on the particles found in their samples, and they made no attempt to correct for undersampling large particles. They report accretion rates of 400×10^9 g/yr, 5000 to $30\,000 \times 10^9$ g/yr, 10×10^9 g/yr, and 14×10^9 g/yr, respectively. We believe that each of these studies has a flaw that led to an overestimation of the accretion rate. Ganapathy (1983) and LaViolette (1985) calculated accretion rates that are one to two orders of magnitude higher than those in the more recent studies. Rasmussen et al. (1995) suspected that the high values reported by Ganapathy (1983) were due to contamination of the samples before analysis by Ganapathy. We concur in this suspicion because in our own work we have found that it is extremely difficult to eliminate such contamination. We suspect that the LaViolette data also suffered from contamination, although much of this may have been natural contamination of the ice from bedrock that was incorporated into the glacier. The data from Rocchia and coauthors and Rasmussen and coauthors are closer to our results, but we suspect that they too overestimate the accretion rate, as we discuss below.

Rocchia et al. (1990) analyzed Ir in 45 samples of Antarctic ice deposited in the years 1895 to 1940 to search for evidence of the Tunguska event. They concluded that there was no significant Tunguska Ir pulse in their data, and stated an accretion rate of 10×10^9 g/yr based on the average of their data. However, Rocchia and coauthors did not calculate an accretion rate that takes into account the undersampling of large particles. As we have shown above, our best estimate of the total accretion is a factor of 12.5 (based on Grün et al., 1985) to 25 (based on Love and Brownlee, 1993) more than we actually measure, and because of undersampling the Rocchia and coauthors data would need an even larger correction than we use. The area-time product for the entire Rocchia and coauthors experiment was approximately 0.05 m^2yr, a factor of three less than covered by our experiment. Therefore, the Rocchia and coauthors measurement of 10×10^9 g/yr implies an accretion rate for all particles of at least 125×10^9 g/yr, a significantly higher accretion rate than we obtain.

There are several reasons why our results could differ from those of Rocchia and coauthors First, they do not make a correction for terrestrial Ir, which accounts for 60% on average of the total Ir in the GISP2 Holocene age samples. While there is a lower terrestrial dust flux to Antarctica than to Greenland (a factor of four is suggested by the ^{4}He data in Brook et al., 2000), we do not know the Ir content of the Antarctic terrestrial dust, or whether there are large particles of terrestrial dust in the Rocchia and coauthors samples (a 300 to 400 μm diameter particle with average crustal Ir abundance could account for the highest Ir peak in the Rocchia and coauthors data). A second possibility is contamination in the Antarctic samples. The large Ir peaks in the Rocchia and coauthors data (at roughly 11.3 and 12.3 m in their core), would equate to 50 μm diameter chondritic particles. Our samples of Greenland ice represent a significantly larger area-time product than do the samples studied by Rocchia and coauthors, yet the largest extraterrestrial particle we find is less than ~20 μm. The probability that two 50 μm chondritic particles would occur in the Rocchia and coauthors sample whereas only a 20 μm particle would occur in ours is less than 1%. Given this low probability, we conclude that these high Ir data in the work by Rocchia and coauthors are not micrometeorites, and if not due to large terrestrial dust particles, they are probably contaminants. A third possible explanation for our disagreement is underrepresentation of extraterrestrial dust at the GISP2 site relative to Antarctica. However, preferential deposition of micrometeorites at Antarctica, or stronger corrosion of particles at Greenland relative to Antarctica (Olinger et al., 1990; Robin et al., 1990) is unsupported by ^{3}He data in Brook et al. (2000) (see below), which shows the same rate of deposition of extraterrestrial material in this size range at Antarctica and Greenland. Based on these considerations, we conclude that the data in Rocchia and coauthors are probably influenced strongly by terrestrial Ir or contamination, and so when corrected for undersampling, overestimate the true accretion rate. Ironically, the numbers that Rocchia presents, which did not have an undersampling correction, are in rough agreement with our results for the total accretion. This agreement, however, is accidental.

Rasmussen et al. (1995) searched for evidence of the Tunguska event by measuring Ir in dust from the Crête (Greenland) ice core through the years 1905 to 1914. That study was designed to find a large pulse of Ir associated with the Tunguska event and not to assess the relatively low background accretion rate, and according to a co-author of that work, G. Kallemeyn (personal communication, 2002), these measurements should properly be considered only an upper limit. Rasmussen and coauthors measure an average Ir content of the dust of 0.367 ($\pm$ 0.147) x 10^{-9} g Ir/g dust. Using their published Fe data, we estimate that at least 25% of their Ir is from terrestrial dust, and assuming an Ir content for chondritic material (500 ppb) calculate an upper limit of uniform accretion to Earth of particles in the approximate size range 0.45 to ~20 μm to be 14×10^9 g/yr (95% confidence). The area-time product for the entire Rasmussen and coauthors experiment was approximately 0.18 m^2yr, slightly more than covered by our experiment. Like our data, the Rasmussen and coauthors estimate needs to be increased by a factor of 12.5 to 25 to account for undersampling, which yields an accretion rate of at least 175×10^9 g/yr.

It is possible that the samples used by Rasmussen and coauthors were affected by laboratory contamination. Rasmussen and coauthors used a steel microtome knife to shave the surfaces of the ice samples before filtration. Rocchia et al. (1990) noted that stainless steel screws in their filter holders were sufficient to contaminate their samples with Ir. Some of our initial measurements (unpublished) were made using samples that had been scraped with a steel microtome knife, and we found fragments of the knife in the dust concentrates. While the Rasmussen and coauthors data do not indicate the presence of large pieces of steel, as little as 0.3 μg of steel with 100 ppb Ir in each of their samples could account for the Ir excess relative to our measurements. Given the relative ease of contaminating an ice sample with this much steel, we consider the Rasmussen and coauthors data an upper limit on accretion.

Extraterrestrial particles have been separated from polar ice, including magnetic spherules (El Goresy, 1968), and other micrometeorites (Maurette et al., 1986, 1987, 1991; Taylor et al., 1998). Maurette et al. (1987) estimate the accretion rate of 0 to 300 μm micrometeorites at Greenland to be 4×10^9 g/yr. Their estimate for particles in the size range 0 to 50 μm is 0.9 $\times 10^9$ g/yr. These values are in agreement with our estimates extrapolated from our measurements of particles in the size range 0.45 to $\sim$20 μm. Taylor et al. (1998) measured the extraterrestrial spherule component of accreted material (unmelted material was not included) which were collected from an Antarctic water well. They estimate an accretion rate of 1.6 ($\pm$ 0.3) x 10^9 g/yr for spherules in the size range 50 to 700 μm, and conclude that their estimate of accretion is about half that calculated by Maurette and coauthors Despite the factor of two disagreement, both of these results are within the accretion limits that we set.

As noted in Table 1, however, Taylor and coauthors do not give the accretion as equal to their measured value. Rather, they assume that the Love and Brownlee (1993) estimate of 40×10^9 g/yr is the true accretion rate. By comparison of their measurement with the estimated accretion rate of 40×10^9 g/yr by Love and Brownlee (1993), Taylor and coauthors estimate that 96% of the mass would have ablated during atmospheric entry. Therefore, they multiplied their measured mass by 25 to estimate the total accretion rate.

Noble gas isotope ratios have also been used to estimate the extraterrestrial dust content in ice core studies. Brook et al. (2000) measure He in 0.2 to 0.45 μm and >0.45 μm dust concentrates from both the GISP2 and Vostok cores. Their data show that only $\sim$2% of the extraterrestrial ^{3}He was found in the fraction 0.2 to 0.45 μm, supporting the ablation model of Farley et al. (1997) which indicates that most extraterrestrial ^{3}He is in particle sizes ranging from 3 to 35 μm. Brook and coauthors also noted a larger terrestrial He component in the 0.2 to 0.45 μm fraction (measured using ^{4}He) than in the >0.45 μm fraction. This result suggests that there is more terrestrial dust in the < 0.45 μm fraction than in the particles with larger diameter. Thus, accurate measurement of extraterrestrial dust in the < 0.45 μm fraction, particularly by He analysis, may be hindered by the large terrestrial signal.

Brook and coauthors calculate an extraterrestrial ^{3}He flux in the GISP2 core of 0.62 ($\pm$ 0.27) x 10^{-15} cm^{3}STP/cm^{2}yr, and in Vostok 0.77 ($\pm$ 0.25) x 10^{-15} cm^3 STP/cm^{2}yr. To translate these results into estimates of the mass of accreted dust, we use He data from IDPs determined by Nier and Schlutter (1992). Analyzing individual IDPs with diameters ranging from 10 to 20 μm, they published the ^{4}He content and the ^{3}He/^{4}He ratio for 13 IDPs, 12 of which we use to calculate the volume of He per gram of dust (Nier and Schlutter eliminated one of their data because of low gas yield). For the Nier and Schlutter IDPs we calculate the ^{4}He average and standard deviation from the mean to be 0.69 ($\pm$ 0.14) x 10^{-3} cm^{3}STP/g, and an error-weighted average ^{3}He/^{4}He ratio of 2.7 ($\pm$ 0.1) x 10^{-4}. From these data we calculate an average IDP ^{3}He concentration of 1.9 ($\pm$ 0.3) x 10^{-5} cm^{3}STP/g. We note that these numbers differ slightly from those reported by Nier and Schlutter (1992); it is possible that the average ^{4}He value published in their data table contains a calculation error. Given our estimate of the ^{3}He content of IDPs, the ^{3}He measured by Brook and coauthors equates to an IDP accretion rate for >0.45 μm particles of 0.17 ($\pm$ 0.08) x 10^9 g/yr for the GISP2 core, and 0.21 ($\pm$ 0.08) x 10^9 g/yr for the Vostok core. These statistically-indistinguishable measurements of accretion suggest that there is no deposition bias between Antarctica and Greenland.

As with our Ir measurements, the Brook and coauthors ^{3}He measurements undersample the total accretion, both by missing large rare particles, and by He loss due to ablation. Converting ^{3}He measurements to total extraterrestrial accretion requires knowledge of where ^{3}He resides in extraterrestrial particles; perhaps because of this uncertainty many papers reporting extraterrestrial ^{3}He do not attempt to estimate the total extraterrestrial accretion rate. Farley et al. (1997) modeled the fraction of total surface area and mass that survives atmospheric entry below the degassing temperature of He. These model results suggest that 4% of the total surface area and 0.5% of the total mass retains its preentry He. If He is surface correlated, as would be the case for He implantation by the solar wind, then the estimated IDP mass made from ^{3}He measurements should be multiplied by 25 to yield the total extraterrestrial accretion rate. Alternatively, if He is volume correlated, as would be expected if He is a primordial component of IDPs, then the IDP mass should be multiplied by 200 to give the total accretion rate. Applying these correction factors to the Brook and coauthors data gives accretion rates for these two alternatives of 4.25 ($\pm$ 2.00) x 10^9 g/yr or 34.00 ($\pm$ 16.00) x 10^9 g/yr for GISP2 and 5.25 ($\pm$ 2.00) x 10^9 g/yr or 42.00 ($\pm$ 16.00) x 10^9 g/yr for Vostok. We note that only the accretion rate calculated from surface-correlated He would make the Brook and coauthors accretion rate consistent with ours. Based on these comparisons, we consider our Ir results to be consistent with the data from Brook and coauthors, and conclude that He is a surface area correlated feature of IDPs.

We recognize that our use of the Farley and coauthors model results above (which uses an entry velocity of 17 km/s) to estimate the mass of IDPs from the Brook and coauthors He data is not entirely appropriate. Our preference is for the higher velocity of 50 km/s suggested by Mathews et al. (2001) which brings the LDEF accretion data into agreement with our results. In a qualitative sense, we can say that higher entry velocities will shift the distribution of particles that retain He after atmospheric entry to smaller diameters. While we have not calculated the percent of He that would survive atmospheric entry based on an average velocity of 50 km/s, we can state that there would be a greater delivery of particle surface area than particle mass below this new threshold. Based on this qualitative argument, we would expect even greater disagreement between our data and revised IDP mass estimates based on mass-correlated He. Therefore, regardless of the exact correction factor to translate He measurements into IDP mass, the surface area-correlated He will be more consistent with our model estimates.

4.2. Comparison with Ocean Sediment Studies

Ir and Os extracted from mid-Pacific Ocean sediment samples have been used to estimate accretion through the last 80

Myr (e.g., Barker and Anders, 1968; Murrell et al., 1980; Kyte and Wasson, 1986; Esser and Turekian, 1988, 1993; Peucker-Ehrenbrink, 1996; Peucker-Ehrenbrink and Ravizza, 2000). Accretion estimates based on these measurements range from a high value of 78×10^9 g/yr based on Ir in the interval 33 to 66 Ma (Kyte and Wasson, 1986) to a low value of 30 ($\pm$ 15) x 10^9 g/yr for the interval 0 to 80 Ma based on Os isotopic studies (Peucker-Ehrenbrink and Ravizza, 2000). The large uncertainties in the more recent measurements make those values compatible with ours at the two standard deviation level. We note that the accretion rate reported by Kyte and Wasson (1986) has been revised downwards by Peucker-Ehrenbrink (1996), who suggests that most of the Ir measured by Kyte and Wasson was not extraterrestrial in origin. Peucker-Ehrenbrink (1996) calculates an accretion rate of approximately 30×10^9 g/yr from the Kyte and Wasson data.

Extraterrestrial accretion has also been estimated from excess ^{3}He in ocean sediment. Studies by Takayanagi and Ozima (1987), Farley (1995), Farley and Patterson (1995), Patterson and Farley (1998) and Marcantonio et al. (1996, 1998, 1999) have yielded surprisingly consistent measurements of ^{3}He in deep sea sediment, averaging around 1×10^{-15} cm^3 STP/cm^2 yr. Using the estimated IDP ^{3}He content of 1.9×10^{-5} cm^3 STP/g that we calculated above from data in Nier and Schlutter (1992), and assuming that the He is correlated with IDP surface area, the estimated mass flux of extraterrestrial material to the deep ocean is approximately 7.5×10^9 g/yr. When measurement errors are considered for the He data, we consider this value to be consistent with our 95% confidence limits, and with the accretion estimates based on ^{3}He in polar ice core samples (Brook et al., 2000). The consistent He data from oceanic and polar ice samples supports the notion that relatively small polar ice samples can be used to adequately measure the long-term ^{3}He flux to Earth.

4.3. Comparison with Satellite, Stratosphere and Radar Estimates

Love and Brownlee (1993) estimate extraterrestrial accretion based on the study of micrometeorite impact craters on the LDEF satellite. Their estimate of 40 ($\pm$ 20) $\times 10^9$ g/yr agrees well with the long-term accretion rate estimates made from pelagic ocean sediment samples (Kyte and Wasson, 1986; Peucker-Ehrenbrink and Ravizza, 2000). Love and Brownlee (1993) estimated the kinetic energy necessary to produce the impact craters on the LDEF panels to derive their accretion estimate. The mass estimate was contingent on the assumed impact velocity of 17 km/s for the micrometeorites. Their assumed value of 17 km/s has been recently called into question by Mathews et al. (2001). Mathews and coauthors studied the speed and deceleration of micrometeorites through the atmosphere (effective mass range 10^{-11} to 10^{-4} g) using ground-based radar at the Arecibo observatory. Based on their study, they calculate an average near-Earth velocity of 50 km/s for near-vertically incident micrometeorites, and an accretion rate of 1.6 to 2.3×10^9 g/yr for particles in the approximate size range 1 to 200 μm. The near Earth velocity of 50 km/s is significantly higher than that used by Love and Brownlee, and results from the fact that most micrometeorites observed at Arecibo occurred at apex (sunrise), when velocities are at a maximum. The assumption of an isotropic IDP flux, made by Love and Brownlee disagrees with the Mathews and coauthors data. We note, however, that the particle velocity estimate by Mathews and coauthors does not agree with estimates in other studies (see Grün et al., 1985 and references therein), which suggest velocities closer to 20 km/s.

Mathews and coauthors re-calculated the LDEF accretion rate. Using the Love and Brownlee impact velocity of 17 km/s, they found the accretion rate was 27×10^9 g/yr, somewhat smaller than the value of 40×10^9 g/yr reported by Love and Brownlee. Next, they applied their new velocity estimate of 50 km/s, which reduced by a factor of 14 the mass component of the impact crater kinetic energy. Thus, Mathews and coauthors estimate the LDEF dust accretion rate to be 1.8×10^9 g/yr to Earth, well within the 95% confidence interval for accretion that we derived from the GISP2 Ir data. If this high average velocity is supported by future work, it may resolve some of the disagreement found in the literature regarding the mass of extraterrestrial material delivered to Earth.

4.4. Accretion at the Last Glacial Termination

Our initial motivation for this experiment was to determine whether there was a pulse of extraterrestrial accretion that affected climate at the termination of the last ice age (Karner et al., 1998, 2001). The extraterrestrial Ir contents of the six ice core samples, which cover 317 yr of ice accumulation during the interval 20 to 6 ka, are all consistent at the 1σ level, and therefore we cannot detect an accretion pulse. We note, however, that this may be due to a faulty assumption of a constant Ir/Fe ratio in the terrestrial dust. We can only address this issue with additional sample analyses. Moreover, as we discussed above, the Ir data from 0.45 to $\sim$20 μm particles yielded an estimate of the accretion rate that is substantially lower than previously deduced from other Ir and Os measurements from polar ice and ocean sediments.

While these data do not favor an accretion-related cause for the last glacial termination, several possible relationships of climate and dust accretion are not addressed by our study. First, pulses of accretion could have occurred between the time intervals covered by our samples. Second, pulses of accretion could be missed because measurements were made only on particles in the 0.45 to $\sim$20 μm range. For instance, the recent break-up of an asteroid might produce large particles that, if ablated to $<$0.45 μm, would have been undetected by the present experiment. To improve upon our estimate of the total accretion rate, larger samples of ice, and/or ocean sediment samples, need to be used. Third, low accretion may coincide with interglacial periods; such a relationship is suggested by the ^{3}He data in Farley and Patterson (1995). Our low accretion rate estimates for the Holocene, when compared with the long-term Ir and Os studies (Kyte and Wasson, 1986; Peucker-Ehrenbrink 1996; Peucker-Ehrenbrink and Ravizza, 2000) support this relationship.

5. CONCLUSIONS

We have succeeded in measuring femtogram levels of Ir in small samples ($\sim$1 kg) of polar ice. Terrestrial dust was estimated from measurements of Fe, and accounts on average for

60% of the Ir in the Holocene age samples. With small samples such as these, the terrestrial Ir correction is particularly important and so it is necessary to determine the core-specific terrestrial dust Ir content. Had we used the average crustal Ir abundance found in Mississippi Delta sediment, we would not have detected an extraterrestrial Ir component in our samples. The limited size of the samples means that we find only particles in the small (<20 μm) diameter range. While these small samples do not enable us to measure the total accretion, they do allow us to estimate accurately the component of accretion of particles with diameters 0.45 to $\sim$20 μm, which are largely unaffected by ablation.

Our measured extraterrestrial mass is similar to the extraterrestrial mass determined from ^{3}He measurements made on ice core and ocean sediment samples. From our measurements, we place upper limits on the total accretion of 6.25×10^9 g/yr and 12.5×10^9 g/yr using the particle size spectra of Grün et al. (1985) and Love and Brownlee (1993), respectively. These limits are several times lower than the best estimates obtained by other studies of Ir or Os in ice and sediment cores; however, because of the large uncertainties with these Ir or Os ocean sediment measurements ($\pm$ 50%) their results are consistent with our new data.

For the 317 yr of ice included in our experiment, our measurements of Ir yield a consistently lower accretion rate estimates than previous Ir analyses on ice cores. While our Monte Carlo estimates of the true accretion rate vary by a factor of 30 (as set by our upper and lower 95% confidence limits), the upper limit of 6.25×10^9 g/yr is near the 95% confidence limits for the long-term accretion rate averages obtained from Ir and Os measurements from deep sea sediment. These potentially inconsistent accretion rate estimates may indicate that systematic errors are not being accounted for, or they may indicate that the accretion rate varies by nearly an order of magnitude over millions of years.

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Appendix A1. Data summary for components of INAA experiment in non-dust samples.

				Cellulose acetate filter			Quartz filter (per 14.8 mg)			A1 capsule (per 230.0 mg)			Instrument background	
Number of measurements				5			2			2			1	
Total count time (minutes)				43,430			6,723			2,099			3,849	
Element	Isotope measured	Energy measured (keV)	Reported weights	Mean	RMSD[a]	Average counting error	Mean	RMSD[a]	Average counting error	Mean	RMSD[a]	Average counting error	Value	Counting error
Ir	^{192}Ir	316.5–468.1[b]	($\times 10^{-16}$ g)	48[c]	7[c]		187[c]	19[c]		71[c]	45[c]		−2	+6/−0
Co	^{60}Co	1332	($\times 10^{-9}$ g)	2.5	3.2	0.06	2.23	0.18	0.014	0.86	0.36	0.07	0.1497	0.0023
Cr	^{51}Cr	320.1	($\times 10^{-9}$ g)	424	103	10	316	89	0.7	74	21	0.6	0.29	0.07
Cs	^{134}Cs	795.9	($\times 10^{-9}$ g)	0.34	0.26	0.007	0.093	0.028	0.011	0.12[c]	0.05[c]		0.0083	0.0011
Eu	^{152}Eu	344.3–778.9[b]	($\times 10^{-9}$ g)	0.09	0.08	0.033	2.620[c]	0.016[c]		0.095[c]	0.007[c]		0.0095	0.0010
Fe	^{59}Fe	1099.3	($\times 10^{-6}$ g)	0.23	0.14	0.008	0.5123[c]	0.0009[c]		0.238	0.013	0.0037	0.0092	0.0003
Hf	^{181}Hf	132.9–482.0[b]	($\times 10^{-9}$ g)	0.6	0.5	0.5	33.5	0.8	0.11	3.430[c]	0.028[c]		0.0009	0.0006
Ni	^{58}Co	810.8	($\times 10^{-9}$ g)	48	16	5	213	53	2.6	40[c]	8[c]		−0.16	0.27
Rb	^{86}Rb	1076.7	($\times 10^{-9}$ g)	8	5	1.3	5.2[c]	0.4[c]		1.5[c]	2.1[c]		0.12	0.37
Sb	^{124}Sb	1691.0	($\times 10^{-9}$ g)	0.54	0.27	0.08	5.0	0.8	0.07	1.37[c]	0.13[c]		0.001	0.005
Sc	^{46}Sc	889.2	($\times 10^{-9}$ g)	0.8	0.5	0.035	2.00	0.13	0.0022	148.8	2.9	0.037	0.0044	0.0002
Ta	^{182}Ta	67.7	($\times 10^{-9}$ g)	5.65	0.19	0.10	1.296	0.020	0.008	0.104[c]	0.015[c]		0.0022	0.0023
Th	^{233}Pa	312.0	($\times 10^{-9}$ g)	0.9	0.8	0.5	28.8	0.7	0.04	2.08	0.28	0.08	−0.001	0.005

[a] Root mean square deviation.
[b] Coincidence measurement.
[c] Error-weighted mean value.