

Weathering in Antarctic H and CR chondrites: Quantitative analysis through Mössbauer spectroscopy

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Abstract—Mössbauer spectroscopy is a very useful tool for identifying ferric iron weathering products in meteorites because of the capability to quantify the relative amounts of ferric iron in them. Mössbauer measurements were made of 33 Antarctic H chondrites (predominately H5) and two paired Antarctic CR chondrites. The primary goals of this study are to determine if Mössbauer spectroscopy can be used to determine which phases are weathering in Antarctic meteorites and if the relative amounts of ferric iron correlate with terrestrial age. Determining which minerals are weathering in ordinary chondrites appears very difficult due to variations in composition for different ordinary chondrites of the same meteorite class and possible problems in preparing homogeneous samples. The analysis of the two paired CR chondrites appears to indicate that metallic iron is predominately weathering to produce ferric iron for this class of meteorite. No correlation is seen between the relative amounts of ferric iron and terrestrial age for ordinary chondrites. One Antarctic H5 chondrite (ALHA77294) with a short ^{14}C age of 135 ± 200 years from the dating of interior carbonate weathering products does have a relatively low amount of ferric iron, which is consistent with this meteorite being exposed on the surface for a relatively short time.

INTRODUCTION

Besides classification by meteorite type, Antarctic stony meteorites are classified according to their apparent degree of weathering and fracturing, using letters from A through C for increasing severity with the symbol "e" used for meteorites with evaporite minerals visible to the naked eye. Researchers (*e.g.*, Gooding, 1989; Koeberl and Cassidy, 1991) have generally agreed that while the weathering indices give a rough record of the condition of the meteorites, the indices are not really sufficient for scientific studies of weathering effects. One experimental technique that may be especially suited for giving a more quantitative measure of weathering is Mössbauer spectroscopy, which can distinguish easily between elemental, ferrous (Fe[II]) and ferric (Fe[III]) iron in meteorite samples (*e.g.*, Sprenkel-Segel and Hanna, 1964; Solberg and Burns, 1989; Burns, 1993; Shinonaga *et al.*, 1994). This study will use Mössbauer spectroscopy to try to understand the effects of Antarctic weathering on two types of meteorites containing metallic iron: H and CR chondrites

Ordinary chondrites should have negligible amounts of ferric iron when they fall to Earth, although potential preterrestrial ferric-bearing minerals such as smectite (Hutchison *et al.*, 1987) and scapolite (Alexander *et al.*, 1987) have been reported in ordinary chondrites. Antarctic CR chondrites (Weisberg *et al.*, 1993; Kallemeyn *et al.*, 1994) do contain ferric iron in both preterrestrial phyllosilicates (*e.g.*, chlorite and Fe-bearing serpentines) and terrestrial weathering products. The preterrestrial Fe-bearing phyllosilicates in CR chondrites are similar in composition to those of CM chondrite Nogoya (Bunch and Chang, 1980), which was found to contain at least three distinct types of Fe-bearing serpentines.

However, immediately after a meteorite lands on the surface of the Earth, ferric oxides will begin forming. A study by Gooding (1981) showed a strong resemblance of weathering features in Antarctic ordinary chondrites with a terrestrial fall (from Holbrook, Arizona), which implies that liquid water must play a major role in Antarctic weathering. The dominant alteration products were found to be hydrous ferric oxides that may comprise ~15–20 wt%

of the most intensely weathered ordinary chondrites. A study of weathering of Antarctic iron meteorites and also metallic iron in Antarctic ordinary chondrites by Buchwald and Clarke (1989) showed that the major weathering products of metallic iron were Cl-containing akaganéite and goethite. (Ferrous iron weathering products of metallic iron do not appear stable in the Antarctic environment; Buchwald and Clarke, 1989.) Examinations of ordinary chondrite thin sections (*e.g.*, Gooding, 1981; Wlotzka, 1993) have shown that metallic iron and troilite appear to weather more readily than silicates in ordinary chondrites.

However, Jull *et al.* (1988) and Velbel *et al.* (1991) have found that Antarctic weathering of silicates during the formation of evaporite minerals (*e.g.*, carbonates and sulfates) on ordinary chondrites could also produce ferric oxides such as goethite and lepidocrocite. Jull *et al.* (1988) have shown that these evaporite minerals are of terrestrial origin. Approximately 3–5% of Antarctic ordinary chondrites have visible evaporite minerals (Velbel, 1988). However, infrared diffuse reflectance spectral measurements by Miyamoto (1989, 1991) have shown that carbonates may be present in trace amounts on all Antarctic ordinary chondrites. Visible evaporite minerals are also known to occur on Antarctic ordinary chondrites (and other Antarctic stony meteorites) of all weathering grades (A through C) (Velbel, 1988).

The fall and modification history of an Antarctic meteorite (Gibson and Andrawes, 1980) is believed to be described by the following steps: (1) a meteorite falls at a random location within the Antarctic continent, (2) the formation and movement of ice results in the burial, transportation and exposure of the meteorite, (3) after the meteorite is exposed, freeze-thaw processes start degrading the surface of the meteorite and removing some of the fusion crust, which has prevented the meteorite from previous extensive alteration and (4) the interior of the meteorite begins to weather along fractures and cracks with oxidation resulting in the formation of Fe oxides and other weathering products. This scenario is supported by studies (Gooding, 1981; Nishiizumi *et al.*, 1989), which show no clear correlation between terrestrial age and weathering index.

TABLE 1. Listing of classes, weathering indices, ^{14}C and ^{36}Cl terrestrial ages and ^{14}C ages of carbonates for meteorites analyzed by Mössbauer spectroscopy in this study and the study by Shinonaga *et al.* (1994).

Name	Meteorite Class [†]	Weathering Index [†]	^{14}C Terrestrial Age (years) [‡]	^{36}Cl Terrestrial Age (years) [#]	^{14}C Age of Carbonates (years B.P.) [@]
ALHA77279	H5	A			
ALHA77294	H5	Ae	10,000 ± 1000		135 ± 200
ALHA77299*	H3.7	A		80,000 ± 80,000	
ALHA78038*	L3.4	C			
ALHA81039	H5	A/B	8700 ± 1300		
ALH82114	H5	A/B			
ALH82115	H5	A/B			
ALH83003	H5	A/B			
ALH84003	H5	A/B			
ALH84069	H5	A			
ALH84074	H5	A/B	21,600 ± 1300		
ALH84139	H5	A			
ALH86603	H5	A/B			
LEW87030	H5	A/B			
TIL82415	H5	A/B			
Y-74001*	H5	C			
Y-74082*	H4	B			
Y-74155*	H4	A			
Y-74371*	H5	A			
Y-74647*	H5	A	3470 ± 70		
Y-75028*	H3				
Y-790269	H4-5			55,000 ± 55,000	
Y-790445	H6(-5)			60,000 ± 60,000	
Y-790461*	H3				
Y-790746	H6-5			55,000 ± 55,000	
Y-790986*	H3				
Y-791027	H5			30,000 ± 30,000	
Y-791047*	H4				
Y-791087*	H3				
Y-791428*	H3				
Y-791500*	H3		27,680 ± 240		
Y-791604	H5-4			30,000 ± 30,000	
Y-791820	H4-5			30,000 ± 30,000	
Y-791861	H6-5			25,000 ± 25,000	
Y-791905	H5			5000 ± 5000	
Y-791926	H5			160,000 ± 70,000	
Y-792764	H5-4			680,000 ± 80,000	
Y-792771	H5			20,000 ± 20,000	
Y-792935	H5			100,000 ± 80,000	
Y-793167	H5			320,000 ± 90,000	
Y-793222	H5			100,000 ± 70,000	
Y-793251	H5			80,000 ± 60,000	
Y-793409	H5			160,000 ± 70,000	
Y-793501	H4-5			140,000 ± 70,000	
Y-793510	H4-5			70,000 ± 70,000	
Y-793514	H5			45,000 ± 45,000	
Y-81012	H5			160,000 ± 70,000	

[†] The meteorite classes and weathering indices for the meteorites collected by the United States' Antarctic meteorite program (Allan Hills [ALH], Lewis Cliff [LEW] and Thiel Mountains [TIL]) are from Grossman (1994). The meteorite classes for the meteorites collected by the Japanese Antarctic meteorite program [Yamato Mountains (Y)] are from Yanai and Kojima (1987) except for two of the meteorites (Y-74371 and Y-791047) analyzed by Shinonaga *et al.* (1994). These two meteorites in the Shinonaga *et al.* (1994) paper were given different classifications than the ones published in Yanai and Kojima (1987). ALHA78038 was labeled as an H3.4 in Shinonaga *et al.* (1994), however Grossman (1994) labels this meteorite correctly as an L3.4.

[‡] The ^{14}C terrestrial age for ALHA77294 is from Nishiizumi *et al.* (1989). However, a slightly different ^{14}C terrestrial age of 30,000 ± 2000 years was calculated for this meteorite by Fireman and Norris (1981). The ^{14}C terrestrial age for ALHA81039 is from Jull *et al.* (1995). The ^{14}C terrestrial age for ALH84074 is from Jull (pers. comm.). The ^{14}C terrestrial ages for Y-74647 and Y-791500 are from Beukens *et al.* (1988).

[#] The ^{36}Cl terrestrial ages are from Nishiizumi *et al.* (1989) and Michlovich *et al.* (1995).

[@] The ^{14}C age of the carbonates for ALHA77294 is from Karlsson *et al.* (1991). This age is calculated for years before present (B.P.), where present is defined as 1950 A.D.

* Meteorites analyzed by Shinonaga *et al.* (1994).

To find more systematic weathering trends for Antarctic meteorites, we have done Mössbauer measurements on a number of Antarctic H5 chondrites (Table 1) and on two paired CR chondrites (EET87747 and EET87770). Table 1 lists the classes, weathering indices, ^{14}C and ^{36}Cl terrestrial ages and ^{14}C ages of carbonates for the ordinary chondrites in this study. The primary goals of this study are to determine if Mössbauer spectroscopy can be used to determine which phases are weathering in Antarctic meteorites and if the relative amounts of ferric iron correlate with terrestrial age. These results will also be directly compared to a very similar Mössbauer study of H chondrites, which are also listed in Table 1, by Shinonaga *et al.* (1994). The data in this study are also used in a companion paper by Wolf and Lipschutz (1995) to try to assess the degree to which the contents of labile trace elements in Antarctic H5 chondrites vary with weathering as reflected by ferric iron contents.

EXPERIMENTAL PROCEDURES

Samples

For each meteorite specimen analyzed, a sample mount was prepared by mixing 15 to 110 mg of powdered meteorite with 300 to 400 mg of sucrose. The range of starting samples was dependent on the amount received from the Antarctic meteorite collections of the United States and Japan. The addition of sucrose serves a two-fold purpose. First, if the density of Fe is too high in the sample, then too much of the incident radiation is absorbed and accurate statistics are not acquired. By mixing the sample with a neutral bulking material like sucrose, a more efficient Fe density is obtained, which ultimately yields a better spectrum. A density of 5–7 mg Fe/cm² is found to be optimal (Dyar, 1984). Second, mixing of the sample with sucrose reduces preferred orientation of mineral crystallites, which could adversely affect the spectrum-fitting process. The mixing of the powdered specimens and the sucrose took place under acetone using an agate mortar and pestle.

Powders for the ordinary chondrite samples were prepared for 33 specimens. Powders were also prepared for the two paired CR chondrites. All of the ordinary chondrites recovered by American expeditions measured in this study were either weathering class A or A/B (Table 1). The ordinary chondrites from the Japanese expeditions measured in this study had no

published weathering indices but were selected by Michael Lipschutz during examination at the National Institute of Polar Research in Tokyo as being relatively unweathered. The weathering classes for the two CR chondrites are B/C for EET87747 and B for EET87770.

DATA ANALYSIS

For each meteorite specimen, a Mössbauer spectrum was obtained at room temperature with a Ranger Electronics Mössbauer spectrometer and an ~ 10 mCi ^{57}Co source in Rh foil. This spectrometer resides at the Bitter National Magnet Laboratory at M.I.T. and uses a 1024-channel Canberra Series 30 multichannel analyzer, which collects two mirror image spectra of a single sample. This spectrum is then folded at mid-point to obtain a single 512 data point spectrum. Results were calibrated against the mid-point and peak positions of a metallic iron foil standard with current reference values supplied by the Mössbauer Effect Data Center. An artifact of the folding procedure was that the spectra tended to have a slight negative slope for increasing velocity. This slope was taken out of a few spectra. However, the Mössbauer results were found not to be significantly different when compared to the results for the original spectra (Fisher, pers. comm.).

Spectra were obtained at a ± 5 mm/s velocity range. This cut off the outermost peaks of any magnetic sextets present as illustrated in Fig. 1. These sextets originate from the presence of troilite and metallic iron. However, computed relative peak areas for each phase were normalized to include all six peaks of the troilite and metallic iron sextets. The advantage of this scheme is that the narrower velocity range allows the more important ferric and ferrous iron quadrupole doublets to be better resolved. A few spectra were also measured in the ± 10 mm/s velocity range (Fisher, pers. comm.). The Mössbauer results from both velocity ranges did not significantly differ. However, the larger velocity range would allow the relative peak areas to be more precisely calculated.

After data collection, the Mössbauer spectra were resolved into component Lorentzian peaks using a non-linear regression procedure (Stone *et al.*, 1984) that allows for the constraining of any set of parameters or any linear combination of parameters (peak widths, peak heights, baseline counts, *etc.*). A "best fit" is obtained when the fitting procedure converges (reaches an arbitrary predetermined minimum deviation between the calculated fit and the actual data) and the statistical parameters, χ^2 and MISFIT (the fraction of the experimental signal that remains unfitted), are minimized. The peak parameters are converted, once an acceptable fit is achieved, to mm/s units by use of the metallic iron standard. The precision of the spectrometer and the fitting procedure is, at best, $\sim \pm 0.02$ mm/s for the isomer shift (IS), ± 0.06 mm/s for the quadrupole splitting (QS) and $\pm 1.5\%$ for peak area data for spectra with well-resolved, distinct peaks (Dyar, 1984). (The IS and QS are sensitive not only to the oxidation state and electronic configuration but also to the coordination number and site distortion of the Fe atoms in the particular phase; Bancroft *et al.*, 1967.) However for spectra with overlapping peaks, as in the case here, one can expect standard deviations for these parameters to be poorer.

In all fitted spectra, component peaks for each quadrupole doublet were constrained to have equal areas. The equal area constraint is known to be reasonably justified by precautions taken during sample preparation to eliminate preferred orientation of powdered mineral constituents. Thus, one would expect doublet peaks to have equal areas and for magnetic sextets to have areas in the ratio of 3:2:1:1:2:3. Since the outermost peaks of the magnetic

peaks are not measured, the visible peaks of the magnetic sextet will have the ratio 2:1:1:2. This ratio for magnetic sextets can be used to double-check the acceptability of the fits. Half-widths of component doublets were also constrained to be equal among all peaks.

Fitting of Ordinary Chondrite Spectra

For an ordinary chondrite spectrum (Fig. 1), the four peaks constituting the inner peaks of the magnetic sextet of troilite occur at ~ -2.00 , -0.02 , $+1.61$ and $+3.66$ mm/s. They are designated as peaks T1 through T4, respectively, and in powdered samples have the area ratios 2:1:1:2. Similar peaks for metallic iron occur at $\sim \pm 3.08$ and ± 0.84 mm/s (labeled I1 through I4). Due to severe overlap of troilite's peak T2 with peaks originating from ferrous and ferric iron in coexisting silicates, an area ratio constraint of 2:1 for peaks T1 and T2 was sometimes used when peak T1 was sufficiently well defined. It was also usually necessary to constrain the position of the troilite peak T2 at -0.02 mm/s, because it almost coincides with low-velocity peaks at zero velocity belonging to quadrupole doublets of ferrous and ferric iron ions located in silicates or oxides. The other peak positions of troilite were consistent among different meteorite spectra and agree with published troilite data. A similar set of constraints were used for the metallic iron peaks (*i.e.*, the peak position of I3 was held constant at $+0.84$ mm/s due to severe overlap of peak positions).

After peaks assignable to troilite and Fe had been located, the remainder of the Mössbauer spectra were fitted to two ferrous doublets and one ferric doublet. The two ferrous doublets primarily originate from ferrous iron in olivine (M1 plus M2 sites) and orthopyroxene M2 sites. They are designated as peaks O1 and O2 (located at ~ -0.32 and $+2.62$ mm/s, respectively) and peaks P1 and P2 (located at $\sim +0.06$ and $+2.18$ mm/s, respectively) (Fig. 1). The calculated isomer shifts and quadrupole splittings (olivine: IS ≈ 1.15 mm/s, QS ≈ 2.94 mm/s; orthopyroxene: IS ≈ 1.12 mm/s, QS ≈ 2.12 mm/s) are consistent with published values (Bancroft *et al.*, 1967; Solberg and Burns, 1989). It is assumed that no ferrous iron occupies the orthopyroxene M1 site. These weak peaks

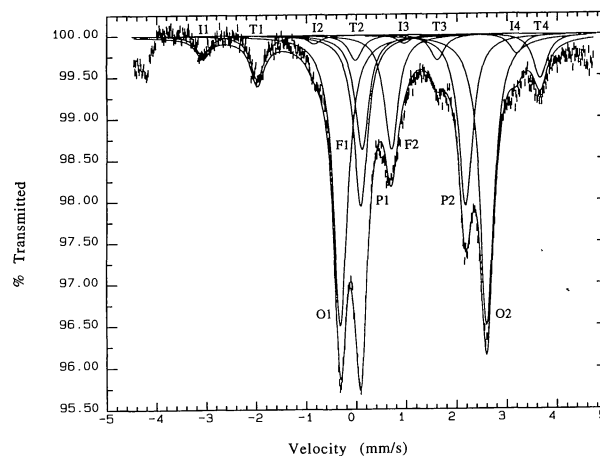


FIG. 1. Mössbauer spectrum of H5 chondrite Y-793167. The vertical lines are the points of the actual Mössbauer spectrum with error bars equal to the square root of the number of counts for each point. The smooth, continuous lines are the fitted peaks. I1, I2, I3 and I4 are peaks assigned to metallic iron. T1, T2, T3 and T4 are peaks assigned to ferrous iron in troilite. O1 and O2 are peaks assigned to ferrous iron in olivine. P1 and P2 are peaks assigned to ferrous iron in pyroxene. F1 and F2 are assigned to ferric iron.

strongly overlap the Fe peaks assigned to olivine and are very difficult to distinguish, however only ~5% of the Fe is estimated to be in this position in ordinary chondrites (Sprenkel-Segel and Perlow, 1968; Bancroft, 1973). Previous researchers (Solberg and Burns, 1989) have been unsuccessful in fitting these orthopyroxene M1 peaks in ordinary chondrites. Also, ~5% of the total pyroxene absorptions in ordinary chondrites corresponds to absorption by Fe in clinopyroxene (Sprenkel-Segel and Perlow, 1968).

The positions of the ferric doublet (labeled F1 and F2 and located from ~ +0.09–0.22 and +0.69–0.84 mm/s, respectively) were found to vary for different ordinary chondrite spectra. However, the calculated isomer shifts (0.39–0.53 mm/s) and quadrupole splittings (0.56–0.76 mm/s) for ferric iron in the ordinary chondrites are consistent with the parameters for ferric iron weathering products (IS ≈ 0.38–0.49 mm/s, QS ≈ 0.50–0.83 mm/s) in meteorites (Solberg and Burns, 1989).

Fitting of CR Chondrite Spectra

Figure 2 shows the resulting room temperature Mössbauer spectra of CR chondrites (a) EET87747 and (b) EET87770. For the CR chondrite spectra, the four peaks for metallic iron are also

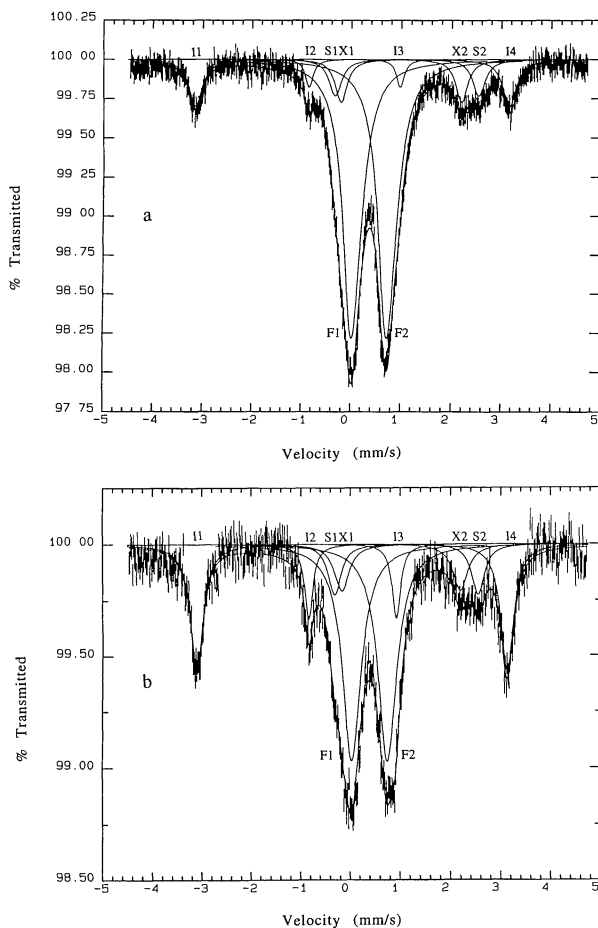


FIG. 2. Mössbauer spectra of CR chondrites (a) EET87747 and (b) EET87770. The vertical lines are the points of the actual Mössbauer spectrum with error bars equal to the square root of the number of counts for each point. The smooth, continuous lines are the fitted peaks. I1, I2, I3 and I4 are peaks assigned to metallic iron. S1 and S2 are the first set of ferrous peaks assigned primarily to ferrous iron in serpentine with a contribution from olivine. X1 and X2 are the second set of ferrous iron peaks. F1 and F2 are peaks assigned to ferric iron.

labeled I1 through I4. After peaks assigned to metallic iron had been located, the remainder of the Mössbauer spectra was fitted to two ferrous doublets and one ferric doublet. Attempts to fit a fourth set of peaks to the CR chondrite spectra were unsuccessful.

The first set of ferrous iron peaks (located at ~ -0.32 and +2.54 mm/s, respectively) is designated as S1 and S2 (IS ≈ 1.11 mm/s, QS ≈ 2.86–2.87 mm/s) and the second set (located at ~ -0.21 and +2.20 mm/s, respectively) is designated as X1 and X2 (IS ≈ 0.99–1.01 mm/s, QS ≈ 2.36–2.40 mm/s). The S1 and S2 are attributed to ferrous iron in serpentine with possible contributions from Fe in olivine offsetting the peak position. Olivine and pyroxene in CR chondrites are usually low in Fe (Fa_{1-6} , $\text{Fs}_{0.5-7}$) (Kallemeyn *et al.*, 1994). A published value, relative to an Fe standard, for low-Fe olivine (12.3% of cation sites occupied by Fe) has an IS = 1.17 mm/s and a QS = 3.02 mm/s (Bancroft *et al.*, 1967). O'Hanley and Dyar (1993) found values (average $\pm 2\sigma$) for the IS = 1.14 ± 0.02 mm/s and the QS = 2.73 ± 0.14 mm/s for ferrous iron in octahedral coordination in terrestrial lizardite. Two lizardite samples with minor olivine impurities had IS and QS values (IS = 1.13 mm/s and QS = 2.88 mm/s for one sample; IS = 1.14 mm/s and QS = 2.93 mm/s for the other) very similar to the ones calculated for peaks S1 and S2. Unresolvable olivine peaks with a larger IS and QS appear to be increasing the IS and QS of the fitted peaks (S1 and S2) that are due primarily to serpentine.

The designation of the second set of peaks (X1 and X2) is more difficult. (This set of peaks is also apparent in the fitted Renazzo spectrum; Burbine, unpublished data.) Melilite, which is found in CR chondrites, is listed by Burns and Solberg (1990) as having an IS (≈ 0.95 mm/s) and QS (≈ 2.40 mm/s) similar to the values for X1 and X2. However, no significant feature due to melilite should be apparent in the spectra since the abundance of refractory inclusions, where melilite is predominately found, is only ~1 vol% in CR chondrites (Weisberg *et al.*, 1993), plus the Fe content of melilite is very small. Staurolite, which has not been identified in any CR chondrite, also has an IS (≈ 0.98 mm/s) and QS (≈ 2.45 mm/s) (Dyar *et al.*, 1991) similar to those of X1 and X2. Published values, relative to an Fe standard, for low-Fe pyroxene (14.5% of cation sites occupied by Fe) (IS = 1.16 mm/s, QS = 2.11 mm/s) are significantly different than the values for X1 and X2. No other minerals (*e.g.*, spinel, chlorite) that have been identified in CR chondrites have IS and QS values similar to X1 and X2. Possible explanations are that the X1 and X2 peaks are due to ferrous iron in some unidentified type of phyllosilicate (or phyllosilicates) or a combination of a few unresolvable peaks.

The one resolved ferric doublet (located at ~ +0.03 and +0.73 mm/s, respectively) (IS ≈ 0.38 mm/s, QS ≈ 0.70–0.71 mm/s) was also fitted to the Mössbauer spectra. The values for the IS and QS for the ferric iron in CR chondrites are also comparable to published values for ferric iron weathering products. No peaks attributable to magnetite or sulfides (troilite or pyrrhotite) were discernible in the Mössbauer spectra, which is expected because the abundance of these minerals in CR chondrites is estimated to be <1 vol% (Kallemeyn *et al.*, 1994). The calculated IS and QS values for the silicates in the two paired CR chondrites are similar to each other and to those of Renazzo (Fisher and Burns, unpublished data), which supports the pairing of these Antarctic samples and their assumed genetic relationship with Renazzo.

Calculation of Relative Areas

The total area of all the Fe peaks was computed by adding the areas of each of the peaks in the spectrum plus the area of the two

metallic iron and two troilite peaks (not present in the CR chondrite spectra) that are present outside the ± 5 mm/s velocity range, whose peak areas are calculated from the 3:2:1:1:2:3 area ratio for the peaks in magnetic sextets. The relative area for iron in a particular phase is calculated by dividing the area of the peaks corresponding to that phase by the total area of all Fe peaks.

TABLE 2. Listing of weathering indices and relative areas of Mössbauer absorption peaks assigned to ferrous iron in olivine, pyroxene and troilite; metallic iron and ferric iron for each of the ordinary chondrite samples.†

Name	Weathering Index	Ferrous Iron in Olivine (%)	Ferrous Iron in Pyroxene (%)	Ferrous Iron in Troilite (%)	Metallic Iron (%)	Ferric Iron (%)
ALHA77279	A	33	19	18	15	15
ALHA77294	Ae	39	22	20	10	8
ALHA77299*	A	38	12	8	12	31
ALHA78038*	C	36	21	10	9	24
ALHA81039	A/B	34	23	10	12	21
ALH82114	A/B	43	24	16	12	6
ALH82115	A/B	36	21	14	20	8
ALH83003	A/B	33	22	12	19	14
ALH84003	A/B	32	21	9	31	7
ALH84069	A	36	20	15	19	10
ALH84074	A/B	24	22	12	34	8
ALH84139	A	28	21	9	24	19
ALH86603	A/B	31	20	5	23	20
LEW87030	A/B	38	21	17	16	8
TIL82415	A/B	42	24	14	13	7
Y-74001*	C	39	22	13	5	21
Y-74082*‡	B	?	?	10	9	36
Y-74155*	A	48	27	15	6	4
Y-74371*	A	48	24	14	7	7
Y-74647*	A	48	23	14	6	8
Y-75028*		38	16	10	7	29
Y-790269		26	13	12	14	35
Y-790445		15	17	4	33	31
Y-790461*		35	18	12	7	29
Y-790746		28	16	11	39	6
Y-790986*		37	24	15	7	18
Y-791027		16	13	13	29	29
Y-791047*		31	18	10	4	38
Y-791087*		31	20	10	6	32
Y-791428*		34	13	15	9	30
Y-791500*		39	22	16	9	15
Y-791604		31	16	14	10	29
Y-791820		28	21	14	24	12
Y-791861		31	25	12	17	15
Y-791905		33	20	5	18	24
Y-791926		36	22	13	14	15
Y-792764		35	17	7	19	22
Y-792771		33	20	18	12	16
Y-792935		21	20	0	29	30
Y-793167		38	22	18	7	15
Y-793222		34	20	14	7	25
Y-793251		27	16	12	13	33
Y-793409		34	19	9	22	16
Y-793501		35	19	15	10	21
Y-793510		35	19	13	16	17
Y-793514		30	16	6	21	27
Y-81012		15	12	13	39	22

† Weathering indices, when available, are included for each meteorite. The total percentage for each meteorite equals 100% (or 99% or 101% due to round-off errors).

* Meteorites analyzed by Shinonaga *et al.* (1994).

‡ In Shinonaga *et al.* (1994), the published (not rounded to nearest percent) relative areas for the peaks assigned to ferrous iron in olivine and pyroxene were 35.9% and 20.8%, respectively, for Y-74082. The published sum of these two areas was 45.0% (not 56.7%). The sum of 45% appears correct and also the other relative areas for Y-74082, but one or both of the relative areas for the Fe peaks of olivine and pyroxene appear to have been published with incorrect values.

RESULTS

Relative Areas

Table 2 has the relative areas of the peaks assigned to ferrous iron in olivine, pyroxene and troilite; metallic iron and ferric iron for the 33 samples analyzed in this study and 14 samples analyzed by Shinonaga *et al.* (1994). The Mössbauer analyses of Shinonaga *et al.* (1994) were done using similar procedures and have results that should be directly comparable to the results of this study. Table 3 has the relative areas of the peaks assigned to ferrous (S1 and S2, X1 and X2), metallic and ferric iron for the two CR chondrites. Each calculated relative area was rounded to the nearest percentage point. The sum of the relative areas for each meteorite will equal 100% (or 99% or 101% due to round-off errors). The relative area for each phase is proportional to the amount of Fe associated with each phase and the value of its parameter f (recoil-free fraction), which is defined as the probability of a recoilless emission and absorption occurring. Approximate values for f at room temperature tend to range from 0.65–0.90 for silicates and oxides with minerals containing ferric iron tending to have slightly higher values than those containing ferrous iron (De Grave and Van Alboom, 1991). The f at room temperature for metallic iron is estimated to be greater than the values for silicates and oxides (Sprenkel-Segel and Hanna, 1964). Because f is not precisely known for all phases in this study, relative areas will be given. However, f for the same phase in different samples of the same type of meteorite should be approximately the same since the mineralogic composition of each phase should be similar. The relative areas of the peaks for each phase should be directly proportional to the percentage of Fe in that particular phase for each sample analyzed and will be interpreted as such.

Weathering of Ordinary Chondrites

To try to determine if the Mössbauer results can be used in determining which phases are weathering in ordinary chondrites, the relative areas of the peaks assigned to metallic iron (Fig. 3a) and ferrous iron in troilite (Fig. 3b), olivine (Fig. 3c) and pyroxene (Fig. 3d) were all plotted against the ferric iron relative areas for both the samples in this study (open squares) and the samples in the study of Shinonaga *et al.* (1994) (filled squares). If the samples have the same initial (before weathering) relative amounts of Fe in different phases and no Fe is lost nor added to the sample during weathering, then the creation of

TABLE 3. Listing of weathering indices and relative areas of peaks assigned to ferrous iron (S1 and S2, X1 and X2), metallic iron and ferric iron for CR chondrites EET87747 and EET87770.†

Name	Weathering Index	S1 and S2 (%)	X1 and X2 (%)	Metallic Iron (%)	Ferric Iron (%)
EET87747	B/C	6	6	23	65
EET87770	B	6	7	46	40

† The total percentage for each meteorite equals 100% (or 99% due to round-off errors).

ferric iron during weathering should decrease the relative areas of Fe in the phases that are weathering. Phases that are not weathering should show no decrease in their relative areas with

increasing amounts of ferric iron. However, as can be seen in Figs. 3a–d, there is a wide scatter in relative areas. The relative areas of metallic iron show no real decrease with increasing ferric iron. The relative areas of Fe in troilite, olivine and pyroxene do show a slight decrease with increasing ferric iron. The large amount of scatter makes any interpretation difficult. However, the apparent decrease with increasing ferric iron content for troilite, olivine and pyroxene is consistent with the weathering of these minerals.

The reason for this lack of any apparent decrease in the relative areas of metallic iron with increasing ferric iron content may be due to two factors. One factor may be the variation in normative mineralogies for many H chondrite samples. For example, McSween *et al.* (1991) found that the abundance of metallic iron varied from

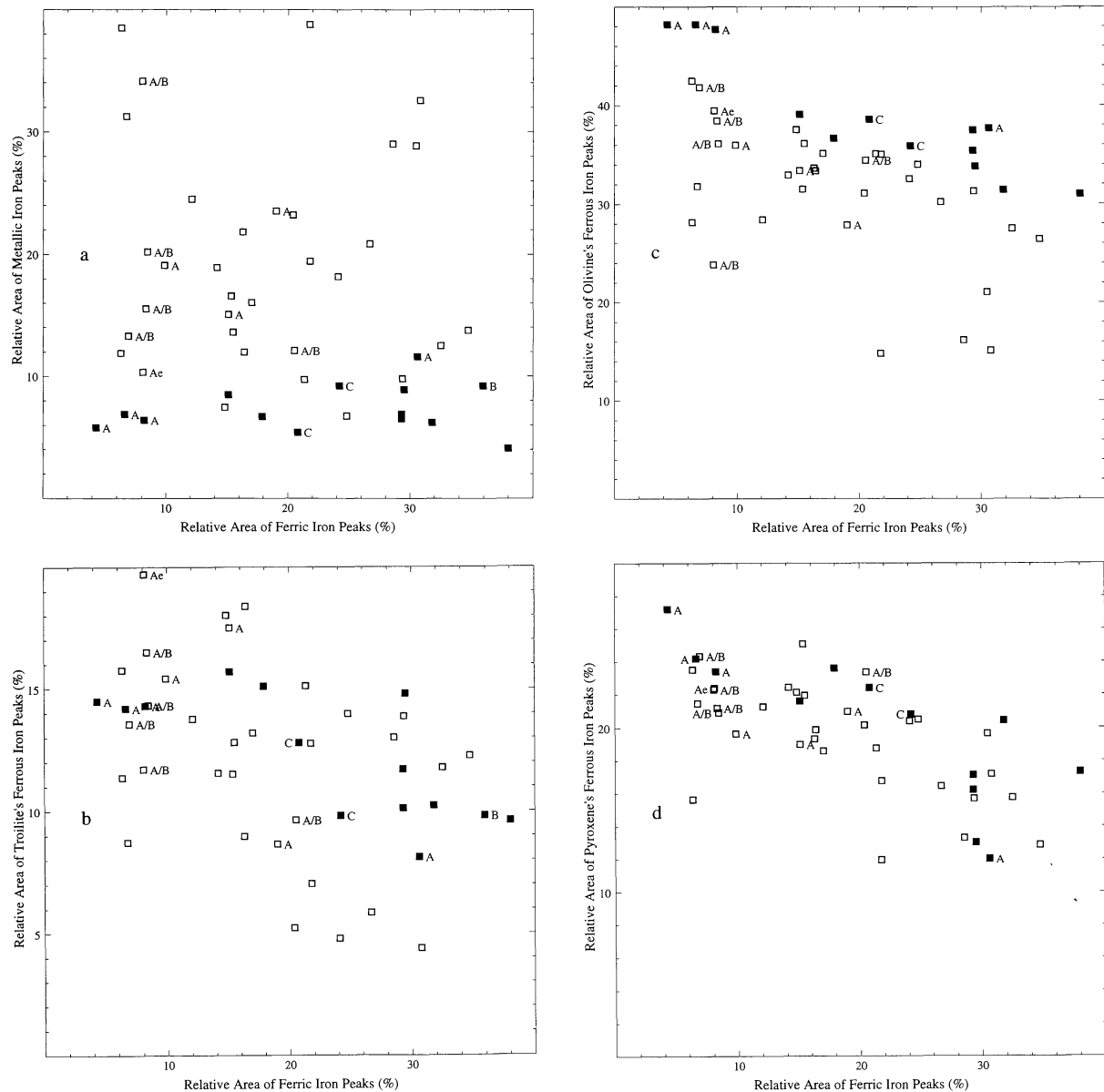


FIG. 3. Relative areas of peaks assigned to (a) metallic iron and ferrous iron in (b) troilite, (c) olivine and (d) pyroxene vs. relative areas of peaks assigned to ferric iron for 33 ordinary chondrite samples analyzed in this study (open squares) and 14 ordinary chondrite samples analyzed by Shinonaga *et al.* (1994) (filled squares). Data for Y-74082 are not plotted in (c) and (d) since the published relative areas for the Fe peaks assigned to olivine and pyroxene for this meteorite are incorrect (see Table 2 for further explanation). Weathering index labels for meteorites from American (Grossman, 1994) and Japanese expeditions (Yanai and Kojima, 1987) are placed to the right of the corresponding square except for a few cases where the weathering index labels overlap other data points and the labels are placed to the left. Meteorites without published weathering indices are from the Yamato Mountains of Queen Maud Land. Relative areas in the figure are from Table 2.

15 to 21% for H5 chondrites. Another factor may be the problem of preparing meteorite powders (15–110 mg) with similar relative amounts of metallic iron, troilite, olivine and pyroxene (Kallemeyn *et al.*, 1989). Since the mechanical crushing properties of metal and silicate phases are so different, it is very difficult to produce well-homogenized splits in the 100-mg-size range since the metal grains often deform rather than break and therefore are not uniformly dispersed into the powder (M. Gaffey, pers. comm.). If these two factors are important, the measured relative areas of each phase would then be related to the initial composition of the ordinary chondrite, the sample preparation process and the amount of weathering. The relative amounts of metallic iron would probably show the most variation, which would explain the large amount of scatter in Fig. 3a.

This interpretation is different from the conclusions of Shinonaga *et al.* (1994) from the analysis of their data (filled squares). They also concluded that there was no correlation between the relative abundances of metallic and ferric iron, but that the decrease in relative amounts of troilite and the silicate minerals (olivine and pyroxene) with increasing relative amounts of ferric iron was indicative of weathering of these minerals to produce ferric oxide products.

The reason for this different interpretation is that the relative areas of the metallic iron peaks in the study of Shinonaga *et al.* (1994) (filled squares in Fig. 3a) were relatively constant and varied only from 4 to 12% while the relative areas of the ferric iron peaks vary from 4 to 38%. In contrast, the relative areas of the metallic iron peaks in the samples analyzed in this study (open squares in Fig. 3a) tended to be higher and varied from 7 to 39% for approximately the same range of relative areas of the ferric iron peaks. The samples of Shinonaga *et al.* (1994) had relative amounts of metallic iron that tended to be constant for different amounts of weathering while the samples in this study had relative amounts of metallic iron that varied considerably for specimens even with the same amount of weathering. For the other phases, the range of relative areas of the Fe peaks of troilite and pyroxene are comparable in both studies while the relative areas of the Fe peaks of olivine tended to be slightly higher in the study of Shinonaga *et al.* (1994) than in this study. The reason for this difference in relative amounts of metallic iron in H chondrites for these two studies is unknown.

However even though the points (squares) in Figs. 3a–d show a considerable amount of scatter, the relative areas of ferric iron of a particular sample should be giving an indication of the amount of weathering for that meteorite. When they fall to the Earth, ordinary chondrites should contain essentially no ferric iron. The amount of ferric iron will then increase as the meteorite weathers. Meteorites with low relative areas of ferric iron will have experienced much less weathering than those meteorites with high relative areas of ferric iron.

Weathering of CR Chondrites

In comparing the relative areas (Table 3) for the two Antarctic paired CR chondrites (assumed to have almost exactly the same preterrestrial composition), EET87747 can be seen to have higher relative amounts of ferric iron and lower relative amounts of metallic iron than EET87770. (The ferric iron will be found in both preterrestrial phyllosilicates and weathering products.) This higher amount of ferric iron indicates that EET87747 is more heavily weathered than EET87770. The relative amounts of ferrous iron

(peaks S1 and S2 and X1 and X2) are approximately the same. The sum of the relative areas for the Fe peaks assigned to metallic and ferric iron for both samples are both ~87%. Assuming that the *f* for ferric and metallic iron are roughly comparable, this constant sum appears to indicate that the ferric iron is being predominately produced from the weathering of metallic iron, which keeps the sum of these two relative areas constant. For these two paired CR chondrites, the metallic iron appears to be substantially weathered while the olivine and pyroxene appear relatively unaffected.

Terrestrial Ages

One of the goals of this study was to see if there was any correlation between the relative amounts of ferric iron and the terrestrial ages (Table 1) of the samples. Five of the meteorites had terrestrial ages calculated for them using ^{14}C activities (Beukens *et al.*, 1988; Nishiizumi *et al.*, 1989; Jull *et al.*, 1995; Jull, pers. comm.), and 21 of the meteorites had terrestrial ages calculated using ^{36}Cl activities (Nishiizumi *et al.*, 1989; Michlovich *et al.*, 1995). Ages up to 30,000 years can be measured by ^{14}C due to its half-life of 5730 years. In many older meteorites, the ^{14}C terrestrial age is only a lower limit due to contamination by atmospheric ^{14}C and production of ^{14}C in the sample (Nishiizumi *et al.*, 1989). The nuclide ^{36}Cl can be used to measure ages up to 1.5 Ma due to its half-life of 301,000 years. The large uncertainties of the ^{36}Cl ages is primarily due to the uncertainty in the assumed saturation value for ^{36}Cl (22.8 ± 3.1 dpm/kg metal) (Michlovich *et al.*, 1995).

Figure 4 is a plot of the calculated terrestrial ages vs. the relative areas of the ferric iron peaks. As can be seen in the figure, there appears to be no correlation between terrestrial age and relative amount of ferric iron. The meteorites with the longest terrestrial ages all have ferric iron relative areas between ~15% and 25%, while the meteorites with the shortest terrestrial ages appear to have relative areas of ferric iron that are distributed almost randomly. The absence of any apparent correlation between terrestrial age and relative amount of ferric iron in the meteorite firmly supports the scenario that the amount of weathering of an Antarctic meteorite is indicative of the amount of time the meteorite has spent on the surface and not the total time that it has resided on the Earth.

Carbon-14 Age of Carbonates

One of the meteorites (ALHA77294) had ^{14}C dating done on extracted CO_2 from carbonates from the interior of the meteorite (Karlsson *et al.*, 1991). The CO_2 was released by reacting powdered bulk meteorite samples with 100% phosphoric acid and heating to a maximum temperature of 50 °C. The reaction with the acid and heating only to low temperatures should result in the released CO_2 only being from carbonates in the meteorite. An accelerator mass spectrometer was used to measure the $^{14}\text{C}/^{13}\text{C}$ ratio in the sample (Donahue *et al.*, 1990). The percent of modern carbon (% modern) of the sample can be determined by dividing the $^{14}\text{C}/^{13}\text{C}$ ratio of the sample to the $^{14}\text{C}/^{13}\text{C}$ ratio of a known standard that corresponds to a 1950 A.D. standard value.

The percent of modern carbon for the carbonate sample from ALHA77294 was 98.3 ± 2.5 , which corresponds to an age of 135 ± 200 years before present (B.P.) (Table 1) where present is 1950 A.D. Ages for carbonates from the interiors of other ordinary chondrites ranged from post 1950 A.D. to $15,000 \pm 3400$ years B.P. with most of the ages between 1000 to 4000 years B.P. (Karlsson *et al.*, 1991). There are two interpretations for these ^{14}C ages. One is that the carbonates formed as the meteorite was on the surface of the ice and

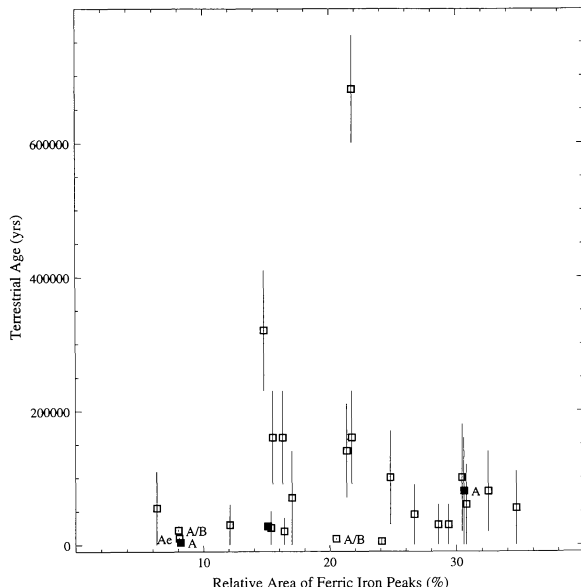


FIG. 4. Terrestrial ages with error bars from ^{14}C (Nishiizumi *et al.*, 1989; Beukens *et al.*, 1988; Jull *et al.*, 1995; Jull, pers. comm.) and ^{36}Cl (Michlovich *et al.*, 1995) radioactive dating (Table 1) vs. relative areas of peaks assigned to ferric iron (Table 2) that were calculated for ordinary chondrites in this study (open squares) and the study of Shinonaga *et al.* (1994) (filled squares). Weathering index labels for meteorites from American (Grossman, 1994) and Japanese expeditions (Yanai and Kojima, 1987) are placed to the right of the corresponding square except for one case where the weathering index label overlaps another data point and the label is placed to the left. Meteorites without published weathering indices are from the Yamato Mountains of Queen Maud Land.

the ^{14}C ages are giving an indication of time the meteorite was exposed on the surface. Another is that either the extracted CO_2 represents several generations of carbonate of different age or that the carbonates have equilibrated with atmospheric CO_2 . This second scenario, deemed more plausible by Karlsson *et al.* (1991), would result in ^{14}C ages that give a lower limit to the age of the carbonates. However, the ^{14}C age for carbonates from the surface of a meteorite will be the actual "formation" age since these carbonates will be unlikely to survive "long" due to the carbonate's vulnerability to aeolian erosion (Jull *et al.*, 1988). The ^{14}C age for the carbonates measured by Jull *et al.* (1988) was ~40 years.

For either of the scenarios of Karlsson *et al.* (1991), the short ^{14}C age of the carbonates from ALHA77294 implies a relatively short time that this meteorite has been exposed on the surface. This proposed short residence time on the surface is supported by the Mössbauer measurements of ALHA77294, which determined that this meteorite had one of the lowest relative areas of ferric iron (8%) of all meteorites in this study.

CONCLUSIONS

This study of Mössbauer spectroscopy of Antarctic meteorites shows the usefulness of this technique in identifying ferric iron weathering products and comparing the relative amounts of ferric iron in Antarctic (and also non-Antarctic) samples. However, determining which minerals are weathering in ordinary chondrites appears very difficult due to variations in composition for different ordinary chondrites and possible problems in preparing homogeneous samples: No correlation is evident between relative amounts of ferric iron and terrestrial age for Antarctic ordinary

chondrites, which corroborates with the work of many other researchers (*e.g.*, Gooding, 1981; Nishiizumi *et al.*, 1989) who have found no correlation between weathering index and terrestrial age. One Antarctic meteorite (ALHA77294) with a short ^{14}C age of 135 ± 200 years (from the dating of interior carbonate weathering products) does have a relatively low amount of ferric iron. The short ^{14}C age and low amount of ferric iron for ALHA77294 is consistent with this meteorite being exposed on the surface for a relatively short amount of time.

For the two paired CR chondrites, the sum of the relative areas of the peaks assigned to metallic and ferric iron is approximately constant even though these two samples have significantly different relative ferric and metallic iron contents. This constant sum appears to indicate that the metallic iron is predominately weathering to produce ferric iron in these CR chondrites and implies that analyzing paired meteorites may be one of the best ways to understand weathering effects.

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