GALACTIC COSMIC RAYS FROM SUPERNOVA REMNANTS. I. A COSMIC-RAY COMPOSITION CONTROLLED BY VOLATILITY AND MASS-TO-CHARGE RATIO

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ABSTRACT

We show that the Galactic cosmic-ray source (GCRS) composition is best described in terms of (1) a general enhancement of the refractory elements relative to the volatile ones, and (2) among the volatile elements, an enhancement of the heavier elements relative to the lighter ones. This mass dependence most likely reflects a mass-to-charge ($A/Q$) dependence of the acceleration efficiency; among the refractory elements, there is no such enhancement of heavier species, or only a much weaker one. We regard as coincidental the similarity between the GCRS composition and that of the solar corona, which is biased according to first ionization potential. In a companion paper, this GCRS composition is interpreted in terms of an acceleration by supernova shock waves of interstellar and/or circumstellar (e.g., $^{22}$Ne-rich Wolf-Rayet wind) gas-phase and, especially, dust material.

Subject headings: cosmic rays — ISM: abundances — stars: abundances — supernova remnants

1. INTRODUCTION

The composition of Galactic cosmic rays (GCRs) contains some of the principal clues regarding their origin. In earlier times, it was believed that GCRs originated in newly processed supernova (SN) ejecta, the SN explosion being held responsible for their general heavy-element enhancement relative to H and He as well as their acceleration. In the 1970s, however, it became clear that the detailed GCR source (GCRS) composition anomalies (§ 2.1) did not seem to be controlled by nucleosynthetic processes (§ 2.2). By contrast, they seemed controlled by atomic, rather than nuclear, parameters, such as the first ionization potential (FIP), the lower FIP elements being systematically in excess relative to the higher FIP ones (§ 2.3). This fact, together with the finding of an extremely similar FIP bias in the composition of the solar corona, solar wind, and solar energetic particles (SEPs) and with the lack of a depletion in GCRs of the refractory elements locked in grains in most of the interstellar medium (ISM), suggested a cosmic-ray origin in the coronal material of later-type stars possessing a cool, neutral-H chromosphere in which an ion-neutral separation could possibly take place. Along this line, it has been conjectured that GCRs consist of stellar energetic particles with frozen-in coronal composition (similar to SEPs), first injected at MeV energies by stellar activity and then later reaccelerated to GeV and TeV energies by passing supernova remnant (SNR) shock waves (§ 2.3). This view required an awkward two-stage acceleration process, in two separate sites. In addition, the presence of an $^{22}$Ne excess in GCRs further required the presence of a totally unrelated second GCR component, presumably originating in Wolf-Rayet wind material (§ 2.3). This scenario also had difficulty accounting for the fairly large spread of the enhancements among the high-FIP elements and, in particular, for the low abundances of H, He, and N.

It was, however, realized long ago that the FIP of the various chemical elements and the volatility of the chemical compounds they form are correlated: typically, low-FIP elements (metals) form refractory compounds, while high-FIP elements (hydrogen, nonmetals, noble gases) form volatile compounds or do not condense at all. Therefore, the apparent ordering of the GCRS composition in terms of FIP could as well reflect an actual ordering in terms of volatility (§ 3)! Such an ordering would imply an enhanced acceleration of those elements locked in dust grains in the ISM, as compared with those in the gas phase. Models for a preferential acceleration of ISM grain destruction products by SNR shock waves were actually explored in the early 1980s (§ 3).

To remove the ambiguity and choose between FIP and volatility as the relevant parameter, the behavior of those elements that are exceptions to the above general correlation must be considered: low-FIP volatile elements and high-FIP refractories. These elements, however, are not the easiest to observe in GCRs. In § 4, we find nine such appropriate clues elements whose GCRS abundances are reasonably well determined. Out of these, four are found to be clearly discrepant with FIP, suggesting volatility as the relevant parameter. The five others are consistent with both FIP and volatility.

A tentative analysis of the data in terms of volatility, performed in § 5, shows that the GCRS abundances of all elements are remarkably well organized in terms of the combined effects of (1) volatility, the more refractory elements being in excess relative to the more volatile ones, and (2) mass, or more probably mass number–to–charge number ratio, $A/Q$, in specific ionization conditions, the more massive volatiles being in excess relative to the less massive ones. This description, in particular, accounts for the low GCRS abundances of H, He, and N, although the
H/He ratio may be somewhat larger than expected; this mass effect is much weaker or absent among the refractory elements. A similar combination of an ordering in terms of FIP and of mass would not account for the data as satisfactorily. Our conclusions are summarized in § 6.

This behavior interpreted in a companion paper (Ellison, Drury, & Meyer 1997, hereafter Paper II) in terms of an \( A/Q \)-dependent acceleration of interstellar and/or circumstellar volatile gas-phase elements by smoothed SNR shock waves and a preferential acceleration of entire dust grains followed by their sputtering, accounting for the roughly mass-independent excess of refractory elements. In this scenario, the acceleration takes place in a single step and at a single site; the \(^{22}\)Ne excess (Appendix) is also naturally accounted for, since higher mass stars produce SNR shocks that accelerate their own \(^{22}\)Ne-rich pre-SN Wolf-Rayet wind material.

2. GCR Composition: The Current General Picture

2.1. GCR Source Composition Determinations

The GCR source composition is derived from cosmic rays observed near Earth by correcting for the effects of solar modulation and of spallation reactions and energy loss during interstellar propagation. The deviations of the GCRS elemental composition relative to solar abundances, assumed to be typical of our local Galactic environment, are plotted versus element mass in Figure 6 below (and vs. other parameters in Figs. 1 and 5). The GCRS over-abundances relative to solar are shown normalized to H for the best-known energy range, between roughly 1 and 30 GeV nucleon\(^{-1}\), but the composition does not seem to change significantly at least up to \(~1000\) GeV nucleon\(^{-1}\) (except possibly for H; see below and, e.g., Shibata 1996). The GCRS abundances for elements up to Ni are mainly based on the \( \text{HEAO C-2 } \) abundances of Engelmann et al. (1990), the review by Ferrando (1993), and the recent \( \text{Ulysses } \) data of DuVernois & Thayer (1996), and those for the \("\text{ultraheavy }\) (UH) elements, with \( Z > 30 \), are based on Binns et al. (1989) and Binns (1995a); more specific references for particular elements are given in § 4. The reference solar elemental abundances are taken from Grevesse, Noels, & Sauval (1996; meteoric determination adopted preferentially), and the isotopic ratios from Anders & Grevesse (1989).

Two remarks apply for UH elements. First, the observations for \( Z > 60 \) have a limited charge resolution, forcing us to deal only with groups of elements: especially the \("\text{Pt group }\) elements, with \( Z = 74-80 \) (hereafter \("\text{Pt}\)\), and the \("\text{Pb group }\) elements, with \( Z = 81-83 \) (hereafter \("\text{Pb}\)\). Second, the current data suggest GCRS excesses by factors of \(~2\) for many elements with \( Z \geq 40 \) relative to Fe. This applies, in particular, for the comparatively abundant \("\text{Pt}\)\), as well as for the secondaries with \( Z \sim 61-73 \) produced by its spallation. By contrast, the rarer \("\text{Pb}\) does not seem related to the Fe. Actually, in view of the increase of the total nuclear destruction cross sections with mass, the derived source abundances of UH elements \("\text{relative to iron}\) are very sensitive to the propagation conditions (an excess of very short path lengths in the GCR path-length distribution relative to the generally assumed exponential distribution [e.g., Ptuskin & Soutoul 1990]) could yield the observed excesses of UH elements, without excess of these elements at the sources; this becomes more and more true for heavier and heavier elements). In addition, for \( Z > 60 \), where only groups of elements can be differentiated, errors on incompletely known, energy-dependent, \("\text{partial cross sections}\) also interferes with the derivation of the source abundances relative to Fe. So, the source \("\text{Pt}/\text{Fe}\) and \("\text{Pb}/\text{Fe}\) ratios cannot be precisely determined. The low source \("\text{Pb}/\text{Pt}\)\) ratio is much better established, although its value also depends upon somewhat uncertain cross sections and upon the path-length distribution (Binns et al. 1989; Clinton & Waddington 1993; Waddington 1996, 1997); this subject is discussed further in § 4. In Figures 1, 5, and 6, the error bars for those UH elements whose source abundance relative to Fe might be affected by such poorly known systematic errors are shown dashed and with a question mark.

The abundances of dominant H and He relative to the heavier elements deserve a special treatment. We consider them in the same energy range where the heavy-element composition is best known, i.e., mainly below \(~30\) GeV nucleon\(^{-1}\). The source abundance of He has been assessed, based on Webber & McDonald's (1994) observed low-energy He/O ratios and their renewed derivation of the source ratio, and on a comparison of the higher energy He fluxes obtained mainly by Webber, Golden, & Stephens (1987) and Soo et al. (1991, 1995, and references therein) with Engelmann et al.'s (1990) observed O fluxes, extrapolated back to the source based on Engelmann et al. (1985). Altogether, we estimate the source He/O to lie in the range \( 19 \pm 4 \), i.e., \( 0.145 \pm 0.030 \) times solar. As for H, it has been anchored to the other elements through the observed H/He ratio. We considered mainly the data by Webber et al. (1987), Soo et al. (1991, 1995, and references therein), Papini et al. (1993), and Swordy et al. (1995; see also Swordy 1994; Shibata 1996). Altogether, these data suggest a local interstellar H/He ratio around \( 23 \pm 5 \) at a given energy per nucleon in the \(~5-30\) GeV nucleon\(^{-1}\) range, which, with a rigidity-dependent escape length from the Galaxy \( \propto R^{-0.6} \) applying above \(~4\) GeV nucleon\(^{-1}\), corresponds to a \( 20^\circ \) times smaller source H/He ratio of \( 15 \pm 4 \). There may be indications of some energy dependence of the observed H/He ratio within this range (the ratio seems much more constant at a fixed rigidity) and of lower ratios at much higher energies (\(~10^2-10^4\) GeV nucleon\(^{-1}\)). All in all, with \( \text{H}/\text{He} = 10 \), H seems slightly enhanced relative to Fe at a fixed energy per nucleon in the range that we consider, by a factor of \(~1.5 \pm 0.4\).

Key determinations of GCRS isotopic ratios can be found in, e.g., Leske (1993), Leske & Wiedenbeck (1993), Ferrando (1993, 1994), Lukasiak et al. (1994, 1997), Shibata (1996), DuVernois et al. (1996a), and Connell & Simpson (1997).

2.2. Difficulties in Interpretation of GCR Composition in Terms of SN Nucleosynthesis

For a long time, it was generally accepted that GCRs originated in newly processed SN ejecta. This view was very tantalizing, indeed, since GCRs are globally enriched in

\(^{1}\) If reacceleration of GCRs while propagating in the Galaxy is important, the composition data are accounted for by a weaker rigidity dependence of the escape length \( \propto R^{-0.5} \); however, the correction factor relating the observed H/He ratio to the source ratio would not be very different from that above (Soo & Ptuskin 1994; E. S. Soo 1997, private communication). Note that the derived source spectra are softer with reacceleration than without, requiring a larger contribution of weak shocks in the primary acceleration (Paper II).
heavy elements, while supernovae synthesize heavy elements, disperse them, have ample energy available for acceleration, and are actually observed to accelerate electrons. It has, however, become clear that the detailed GCR composition is inconsistent with a predominant selection of the elements according to specific nucleosynthetic processes and, more particularly, with what could be expected from SN nucleosynthesis (Arnould 1984; Meyer 1985b, 1988).

Examine the GCR elemental composition anomalies presented versus mass in Figure 6 below. First, considering heavy elements up to the Fe peak, $^{22}$Ne is found to be depleted by a factor of $\sim 8$ relative to Mg, Al, and Na, while these elements are all largely produced by C burning. Similarly, S and Ar are depleted by factors of $\sim 4$ relative to Si and Ca, while these elements are all produced by O and Si burning (see, e.g., Meyer 1988, Fig. 2). No such large anomalies are found in existing SN nucleosynthesis calculations, especially between species produced within the same burning cycle (Woosley & Weaver 1995; Timmes, Woosley, & Weaver 1995; Arnett 1995). By contrast, Mg, Al (C burning), Si and Ca (O and Si burning), and Fe and Ni ($\epsilon$-process) are found to be very close to solar proportions in GCRs (within $\sim 20\%$), while the above nucleosynthesis calculations for specific types of supernovae commonly yield deviations of these ratios by factors on the order of $\sim 2$.

Further, all GCR isotopic ratios are found to be consistent with solar, with the important exception of the $^{22}$Ne/$^{20}$Ne and possibly the $^{13}$C/$^{12}$C and $^{18}$O/$^{16}$O ratios, which are discussed below (see, e.g., Leske 1993; Leske & Wiedenbeck 1993; Ferrando 1994; Lukasjak et al. 1994, 1997; DuVernois et al. 1996a; Connell & Simpson 1997). In particular, the isotopic $^{56}$Co/$^{57}$Co ratio indicates the absence of freshly synthesized Fe-peak nuclei (Leske 1993; Lukasjak et al. 1997).

The composition of the GCR ultraheavy elements with $Z > 30$ largely confirms these views. The observations of all elements through the first and second $r$- and $s$-process peaks (i.e., up to $Z \sim 60$) show no trend for a specific enhancement or deficiency of either $r$- or $s$-nuclei. The observations are roughly consistent with a solar source composition affected by atomic selection effects, as discussed below (and possibly with the general excess of most elements with $Z \geq 40$ relative to Fe discussed above; see Fig. 6) (Binns et al. 1989; Clinton & Waddington 1993; Binns 1995a, 1995b; Waddington 1996, 1997). This observed lack of an $s$-element deficiency definitely implies that GCRs do not predominantly originate in SN-processed material, since no type of SN synthesizes $s$-nuclei (Prantzos, Cassé, & Vangioni-Flam 1993). The apparent excess of the $r$-peak "Pt," contrasting with the lack of an $s$-peak "Pb" excess, has been interpreted in terms of a specific excess of the third $r$-process peak elements in the GCRs material. In view of the total absence of an excess of $r$-elements in the first and second $r$-process peaks, this hypothesis does not seem very likely. The low GCRs "Pb"/$^\alpha$ Pt" ratio probably requires another explanation.

In brief, the current state of the art suggests that the GCRs abundances of most elements are not controlled by specific nucleosynthetic processes, and, in particular, not by SN nucleosynthesis. In fact, it seems that most of the GCR source material is more of the "solar mix" type. There is, however, an important exception to this statement: GCR sources are enriched in the isotope $^{22}$Ne, suggesting the presence of an He-burning material component most likely enriched in $^{12}$C and $^{16}$O as well as in $^{22}$Ne, presumably originating in Wolf-Rayet star wind material; this will be discussed in §§2.3, 3, 5, and 6 and in the Appendix.

2.3. The "FIP" Plus He-burning, Later-Type Stars Plus Wolf-Rayet Picture

Early in the 1970s, it was noted that the detailed GCR heavy-element composition, while not easily ordered in terms of nuclear physics parameters, could be rather well organized in terms of atomic physics parameters. The FIP, or related parameters that control the tendency of an element to be neutral or ionized in a gas at $\sim 10^4$ K (or subjected to radiation of comparable energy), seemed to roughly order the deviations of the GCR elemental composition relative to solar abundances, at least for elements up to Ni (Cassé & Goret 1978 and references therein; Meyer 1985b; Arnaud & Cassé 1985; Silberberg & Tsao 1990). As shown in Figure 1, heavy elements with FIP $\lesssim 8.5$ eV ("low FIP") are typically enhanced by a factor of $\sim 5$ relative to those with FIP $\gtrsim 11$ eV ("high FIP").

The low temperature required for the parent gas ($\sim 10^4$ K), together with the lack of a depletion in GCRs of the refractory elements locked in grains in virtually all but the hottest interstellar medium (Cassé, Goret, & Cesarsky 1975; Cassé & Goret 1978; Dwek 1979), first suggested that the GCR nuclei did not originate in the ISM, but in stellar surfaces (Meyer, Cassé, & Reeves 1979; Meyer 1985b). This conclusion, of course, assumed implicitly that only gas-phase atoms of the ISM could be accelerated.

A concomitant advance in a totally different context, our own solar environment, then strongly influenced our views about the GCR source material. Hints were first found in the observed gradual-event SEP compositions, suggesting FIP-related anomalies, much like those found in GCRs. The difficulty here was that the composition of SEPs is changing all the time. Meyer (1985a) and Breneman & Stone (1985) managed to separate out the permanent FIP-bias imprint on the data, clearly related to the composition of the heliospheric source material, from the rigidity-dependent variations of the composition that resulting from variable conditions of particle acceleration. The solar wind and the spectroscopic studies of the solar coronal gas (EUV, X-rays, nuclear $\gamma$-rays) have progressively confirmed the presence of FIP-biased gas in the entire corona and outer heliosphere outside coronal holes (e.g., Meyer 1985b, 1993, 1996; Feldman 1992; Geiss, Gloeckler, & von Steiger 1995; Garrard & Stone 1993; Reames 1995). The similarity between the GCRS and the solar coronal composition, therefore, strongly supported the earlier arguments suggesting that the GCR nuclei had been first extracted from stellar atmospheres.

The parent gas of the GCR particles could now be specified more precisely: probably the coronae of F–M later-type stars possessing, like the Sun, a cool, predominantly neutral-H chromosphere at around $\sim 7000$ K in which, in some yet debated way, ionized heavy elements may be separated from neutral ones and rise preferentially into the corona (Meyer 1985b; see also the review by Héoux 1995). Efforts have been devoted recently to try to observe the FIP effect in the coronae of later-type stars, thanks to the instruments on board the Extreme Ultraviolet Explorer and ASCA spacecraft, with variable outcomes (Laming, Drake, & Widing 1996; Drake, Laming, & Widing 1997; Singh, White, & Drake 1996; and references therein).
Stellar flare activity is, however, certainly energetically unable to accelerate the bulk of the (GeV) GCRs in the Galaxy. It may, however, accelerate some of the “FIP biased” coronal material to low (MeV) energies, just as the Sun accelerates SEPs, thus providing a suprathermal component with “frozen” coronal composition. These “injected” MeV particles must then be later preferentially accelerated by more powerful SNR shock waves, which boost them to the GeV and TeV energies of GCRs (Meyer 1985b). So, this scenario requires two separate acceleration stages in two separate sites, clearly an undesirable feature.

Further, the coronal gas of later-type stars cannot be the source for the $^{22}$Ne-rich material required to account for the observed GCRS $^{22}$Ne excess. This excess, the only significant source isotopic anomaly found besides possibly low $^{13}$C/$^{12}$C and $^{18}$O/$^{16}$O ratios, together with the high GCRS elemental C/O ratio, suggests the presence of a pure He-burning material component in GCRs, whose most likely origin is Wolf-Rayet wind material (Appendix). So another weak point of the FIP/later-type star scenario is that an additional, entirely unrelated source is required for the $^{22}$Ne-C-O–rich components.

Figure 1 shows an up-to-date version of the correlation of the GCRS abundance enhancements with FIP. Obviously, FIP does roughly order the data, but there is much scatter around the correlation. Among the high-FIP elements, H and He are deficient relative to all heavy elements, as was known early on. Regarding N, accurate GCR isotopic observations and spallation cross sections have now well established its low source abundance. We have plotted the points for C and O as upper limits, because we expect specific $^{12}$C and $^{16}$O contributions associated with the $^{22}$Ne-rich component from Wolf-Rayet stars, discussed in the Appendix; for $^{12}$C, we propose a tentative estimate of its non–Wolf-Rayet source abundance (Appendix). Other elements deviate from the correlation. Among lower Z elements, the source abundance of Na seems low, and that of P high, at least based on the currently best available spallation cross sections for these largely secondary species (see discussion in § 4). The UH-element data by and large confirm the FIP correlation, but with a larger scatter and a general trend toward greater enhancements for $Z \geq 40$. This trend may be real, or it may be due to an improper account of the interstellar propagation (§ 2.1). An important
exception is Ge, which is reliably determined to be low compared with Fe, with exactly the same FIP value (§ 4).

Finally, while the "Pt" and "Pb" abundances relative to Fe are poorly determined (§ 2.1), the low "Pb"/"Pt" ratio also conflicts with an FIP ordering, as also illustrated in Figure 1.

3. THE FIP VERSUS VOLATILITY ISSUE AND GRAIN ACCELERATION MODELS

There exists for most elements a general correlation between FIP and the volatility of the chemical compounds that the element forms. This is illustrated in Figure 2, which shows a cross plot of each element's condensation temperature $T_c$ versus its FIP (updated from Meyer 1981b). This temperature $T_c$ is the calculated 50% condensation temperature in a $10^{-4}$ atm gas with solar composition, taken from Wasson (1985). The lower the $T_c$, the higher the volatility of the element. Figure 2 shows that FIP and $T_c$ are, indeed, anticorrelated for the majority of the elements.

Therefore, the apparent correlation of the GCRS abundances with FIP might just mimic an actual correlation with the element volatility. With this viewpoint, the refractory (low FIP!) elements, those generally locked in grains in the ISM, would be overabundant in GCRs. This would imply a preferential acceleration of grain destruction products, presumably in SNR shock waves. One nice point with such a scenario is that the same SNR shock waves could destroy the grains and fully accelerate the particles to their final GeV and TeV energies. We are back to a one-step acceleration process, in a single site.

Now, it is believed that SNR shocks accelerate mainly external interstellar or circumstellar material, not the SN ejecta themselves. As discussed in Paper II, the role of the outer, forward shock should indeed be dominant, the reverse shock being less energetic and short-lived, so that any particles it may accelerate later suffer severe adiabatic losses (Drury & Keane 1995). Further, while instabilities in the flow may allow some of the ejecta material to speed ahead of the external shock, this effect is believed to be comparatively minor (see, e.g., Jun & Norman 1996; Drury & Keane 1995). It is therefore no surprise that we find little trace of SN nucleosynthesis in the GCRS composition (§ 2.2), despite the key role played by SNR shocks in accelerating the particles. This applies to all types of supernovae, Type I as well as Type II.

The next important question is, of course, what does this external material consist of? Around Type I supernovae and lower mass core-collapse supernovae, i.e., Type II's with comparatively weak winds prior to explosion, this material ought to be ordinary interstellar material, with

![Figure 2](image-url)

Fig. 2.—Cross plot of the condensation temperature $T_c$ of each chemical element vs. the element's FIP. Each $T_c$ value is the 50% condensation temperature of the dominant solid compound formed by the element, for a solar initial gas composition at $10^{-4}$ atm (from Wasson 1985, with additional information from Anders 1977 and Anders & Grevesse 1989). Along with the grouping of the elements into three groups according to their FIP (Fig. 1), we define four classes of volatility: "refractories," with $T_c > 1250$ K, "semivolatiles," with $1250$ K $> T_c > 875$ K, "volatiles," with $875$ K $> T_c > 400$ K, and "highly volatiles," with $400$ K $> T_c$. (see also Figs. 3 and 4). This figure shows the general anticorrelation between FIP and $T_c$, most lower FIP elements being refractory, and higher FIP ones volatile. Those elements for which we currently have reasonably accurate estimates of their GCRS abundance are denoted by large filled dots. Among them, the elements lying outside the main FIP-$T_c$ correlation, which can serve as clues to distinguish between FIP and $T_c$ as the parameter governing the GCRS composition, are enclosed by boxes (filled squares in Figs. 1, 3, and 5). "REE" stands for "rare earth elements."
roughly solar composition (gas plus grain). Its grains consist of old ISM grains. As one proceeds to more and more massive SN progenitors, earlier wind ejections may become more and more important, so that the shocks may accelerate the progenitor’s own wind material. Its grains will then consist of newly formed grains, presumably with different properties. This is almost certainly the case for the highest mass, Wolf-Rayet (W-R) star progenitors, which have been stripped off by huge winds to the point where their He-burning layers have been tapped and their winds are highly enriched in He-burning products (see, e.g., van der Hucht & Hidayat 1991; van der Hucht & Williams 1995). So, another extremely nice feature of this type of scenario is that it may account in a natural, continuous way for the observed excess of He-burning material ($^{22}$Ne, $^{12}$C, possibly $^{16}$O excesses) in GCR sources.

Note that, in this context, the logic of the “grain constraint” earlier put forward by Meyer et al. (1979) to exclude the ordinary ISM as a possible source of the GCR ions is completely reversed: the ISM was excluded as a possible source of the GCRs based on the implicit assumption that ions had to be accelerated out of the ISM gas phase, which is depleted of its low-FIP refractory elements locked in grains. Here we turn the argument around, considering the opposite possibility of a preferential acceleration of this very material locked in grains!

Such a preferential acceleration of grain material in SNR shock waves was considered in the early 1980s. Epstein (1980) first introduced the concept considered in the present work: an acceleration of the entire grain, followed by grain sputtering and by a reacceleration of the suprathermal grain sputtering products to GeV and TeV energies. Cesarsky & Bibring (1981) and Bibring & Cesarsky (1981), on the other hand, suggested that grains freely cross the shock, so that they acquire a bulk speed relative to the ambient shocked gas equal to the shock speed. When these grains undergo strong sputtering in the downstream region, the sputtered ions are thus injected with a speed equal to the shock speed, relative to the ambient gas, and preferentially stochastically accelerated to cosmic-ray energies. First-order Fermi acceleration in the shock itself is, indeed, unlikely for these particles in this scenario, because most ions are sputtered off too far downstream of the shock to diffuse back to it. Of these two ideas, we believe Epstein’s is most likely because of the well-known problems with stochastic acceleration of cosmic rays. First, for stochastic acceleration to be efficient requires Alfvén Mach numbers considerably lower than those expected for most remnants (see, e.g., Reynolds & Ellison 1993). Second, unlike first-order Fermi acceleration, different ionic species (as well as the same species in different environments) can acquire quite different spectral shapes (see, e.g., Forman, Ramaty, & Zweibel 1986), leaving one of the basic observations, i.e., that all cosmic-ray ions have similar power laws, unexplained at a fundamental level.

Further, Meyer (1981b), Taranudar & Apparao (1981), Soutoul et al. (1991), and Sakurai (1995 and references therein) have provided analyses of the GCRs composition data in light of the condensation of elements into grains.

Before we proceed to analyze the GCR data, we review the significance of the condensation temperature $T_c$ and its limitations. We examine to what point the actual, observed composition of two types of astrophysical condensed material seems organized in terms of $T_c$. We first consider
Fig. 3.—Depletion of the more volatile elements among the various types of carbonaceous chondrites, illustrated by the Vigarano-type C3/C1 abundance ratio, vs. condensation temperature $T_c$ (Wasson & Kallemeyn 1988). In C3's the more volatile elements are incompletely condensed, while in C1's most elements are entirely condensed, with relative abundances equal to those in the protosolar nebula. All abundances are normalized to the group of the most refractory elements. "REE" stands for "rare earth elements." The key elements for our analysis of GCRs have been singled out by filled squares. This figure shows that the correlation between the C3/C1 abundance ratio and $T_c$ is quite good, thus confirming the relevance of the parameter at least in this context.

It shows a few distinct groups of elements: (1) very refractory, lithophile elements with $T_c \geq 1400$ K and a group of elements condensing as silicates or together with metallic Fe (siderophiles) around 1350 K, here together denoted "refractories," with $T_c \approx 1250$ K; (2) a group of "semivolatile" elements, with $1250 < T_c < 1150$ K, whose depletion in C3's varies rapidly with $T_c$; (3) a group of "volatile" elements, with $875 < T_c < 400$ K, with C3/C1 ratios less than 0.30; and (4) the "highly volatile" elements, not significantly condensed even in C1's. Not plotted in the figure are H, C, N, O, and the noble gases.

To investigate this point, let us look more closely into the correlation between FIP and volatility for the various elements, shown in Figure 2. By distinguishing between the two types of scenario, in terms of FIP or of volatility, we have to look at the GCRS abundances of those few elements that do not entirely condense in C1's.

The depletions for a given $T_c$ might result from a slow chemical reprocessing governing grain destruction and growth in the ISM, largely independently of $T_c$, subsequent to the primary grain condensation phase in cooling stellar ejecta and winds, which could be more closely controlled by $T_c$; a major grain regrowth in the ISM seems, indeed, required, in view of the short lifetime of each individual grain in the ISM (see, e.g., Joseph 1988; Draine 1990; Cardelli 1994; Savage & Sembach 1996 and references therein). In that sense, newly formed grains in stellar wind envelopes might have a composition different from old ISM grains (Fig. 4) and more closely controlled by $T_c$. All this may, in particular, apply for the comparatively refractory elements P and As ($T_c \sim 1150$ K), which are found to be only slightly depleted in the ISM (Fig. 4); they might be more locked in grains in circumstellar material than in the ISM.

4. CLUES SUGGESTING THE RELEVANCE OF VOLATILITY TO GCRS COMPOSITION

We now ask whether there are observational clues that will allow us to choose between FIP and volatility as the key factor governing the GCRS abundances. This question was addressed early on by Meyer (1981b), and we now update that analysis.

To investigate this point, let us look more closely into the correlation between FIP and volatility for the various elements, shown in Figure 2. To distinguish between the two types of scenario, in terms of FIP or of volatility, we have to look at the GCRS abundances of those few elements that do
Our adopted four classes of volatility have been singled out, as in Fig. 3. The general trend is clearly a larger depletion of the more refractory elements in the ISM gas phase, but the spread in the depletions between elements with similar values of $T_c$ is much larger than in Fig. 3, presumably as a result of grain destruction and reprocessing in the ISM. Note in particular the apparent small depletion of comparatively refractory P (and As, which has similar chemical properties). The behavior of the more volatile elements suggests that some of the spread may be observational and that there exist some problems with the solar normalization.

Sodium ($Z = 11$).—Despite its very low FIP of 5.1 eV, Na is a rather volatile element ($T_c = 970$ K; Fig. 3). Na seems deficient by a factor of $2.0 \pm 0.8$ relative to Si (Ferrando 1993; DuVernois & Thayer 1996). This points toward volatility as the relevant parameter controlling the GCRS composition. The measured Na abundance in arriving GCRs is absolutely foolproof (Engelmann et al. 1990; DuVernois, Simpson, & Thayer 1996b). However, Na is a predominantly secondary element in these observed GCRs, so the determination of its source abundance is very sensitive to spallation cross section errors; while the large error bar on its adopted source abundance is based on a conservative estimate of these errors, it might still be not entirely definitive.

Sulphur ($Z = 16$), zinc ($Z = 30$), and selenium ($Z = 34$).—These three elements form a group (Fig. 2). All have neighboring values of FIP (10.4, 9.4, and 9.8 eV, respectively) that place them in the “intermediate FIP” region, where the amplitude of the FIP bias seems to be a rapidly varying function of the FIP value. All three also have very similar values of $T_c$ (650, 660, and 680 K, respectively) that make them full-fledged volatile elements (Fig. 3). The source abundances of these elements are reliably determined. Sulphur is one of the best known elements in GCRs (e.g., Engelmann et al. 1990). We have good data on Zn from a clean pre-HEAO C-3 balloon flight, and from both the C-2 and the C-3 instruments on board the HEAO 3 spacecraft, which converge on a source Zn/Fe ratio of $0.43 \pm 0.08$ times solar (Tueller et al. 1979; Lund 1984; Binns et al. 1981; Binns 1995a; M. H. Israel 1996, private communication). Finally, Se is well measured by both the Ariel and the HEAO C-3 instruments (Fowler et al. 1987; Binns et al. 1989; Binns 1995a). The secondary fraction of all three elements is small, and the destruction cross sections of Zn and Se do not differ much from that of Fe, so that the source Zn/Fe and Se/Fe ratios are close to those observed, with a minor error due to interstellar propagation. The GCRS abundances of these three elements are well interpreted in terms of FIP (Fig. 1). In terms of volatility, the lower GCRS abundance of S, as compared with Zn and Se, may at first seem disturbing; it will be interpreted below as a mass effect.

Phosphorus ($Z = 15$).—The FIP of P, another “intermediate FIP” element, is 10.5 eV, i.e., virtually the same as that of S (10.4 eV). But while S is a full-fledged
volatile \( (T_c = 650 \text{ K}) \), P is a rather refractory semivolatile element \( (T_c = 1150 \text{ K}; \text{ Fig. 3}) \). While S is depleted by a factor of 3.4 \( \pm 0.5 \) relative to Si, P seems depleted by a factor of only 1.5 \( \pm 0.7 \) (consistent with being undepleted), so that the P/S ratio is enhanced relative to solar by a factor of 3.0 \( \pm 1.6 \) (Ferrando 1993; DuVernois & Thayer 1996; see also Leske & Wiedenbeck 1993; DuVernois et al. 1996b).

Thus the high P/S ratio represents another hint in favor of volatility's controlling the GCRS composition. There are, however, two caveats. First, like Na, P is a predominantly secondary element in the observed GCRs, so the determination of its source abundance is very sensitive to spallation cross section errors. Second, while P is a clearly siderophile element and does behave as a rather refractory element in its phase, much less than other elements with comparable chemical reprocessing of the grains in the ISM; if this were the case, P could be much more condensed in the grains recently formed in pre-SN stellar winds than in the general ISM depicted in Figure 4.

Copper \((Z = 29)\) and gallium \((Z = 31)\).—Cu and Ga are low-FIP \((7.7 \text{ and } 6.0 \text{ eV})\), respectively semivolatile elements \((T_c = 1040 \text{ and } 920 \text{ K}, \text{ respectively}; \text{ Fig. 3})\). We have, unfortunately, only one observation of these odd-Z elements in GCRs, by the HEAO C-2 experiment, in which these elements seem well resolved (Byrnak et al. 1983; Lund 1984). It yields Cu/Fe \( = 1.14 \pm 0.25 \) and Ga/Fe \( = 1.51 \pm 0.59 \) times solar. These values are consistent with FIP as the relevant parameter.

Germanium \((Z = 32)\).—Ge has virtually the same FIP as Fe \((7.9 \text{ eV})\), but Ge is a volatile element \((T_c = 825 \text{ K})\), while Fe is refractory \((T_c = 1135 \text{ K}; \text{ Fig. 3})\). The C-2 and the C-3 instruments on board the HEAO 3 spacecraft have yielded independent, consistent GCRS Ge/Fe ratios, both significantly lower than solar. All in all, they lead to a GCRS Ge/Fe ratio of 0.57 \( \pm 0.10 \) times solar (Lund 1984; Binns et al. 1989; Garrard et al. 1990; Binns 1995a; M. H. Israel 1996, private communication). In both instruments, the charge resolution is appropriate to safely observe Ge, and possible systematic errors are limited. The errors are predominantly statistical. As for Zn and Se, the source Ge/Fe ratio is close to that measured (only some \( \sim 13\% \) below; Binns et al. 1989), and the error due to interstellar propagation is insignificant. Therefore these considerations of Ge strongly argue in favor of volatility's controlling the GCRS source composition.

Lead \((Z = 82)\).—As discussed in \( \S \ 2.1 \), we will be comparing the abundances of the “Pb” elements, with \( Z = 81–83 \), essentially made of low-FIP volatile s-elements, with those of the “Pt” elements, with \( Z = 74–80 \), mostly made of intermediate-FIP refractory r-elements (Figs. 2 and 3). While the observations suggest an excess of most elements with \( Z \geq 40 \) relative to Fe at the sources, the derived source abundances relative to iron are very sensitive to the propagation conditions \( (\S \ 2.1; \text{ Fig. 1}) \). Here we will therefore deal only with the “Pb”/“Pt” ratio, which is much less affected by interstellar propagation. According to standard calculations, the source “Pb”/“Pt” ratio is estimated to be \( \sim 1.65 \) times higher than that observed (Binns et al. 1989). Depending upon the propagation conditions (distribution of short path lengths; \( \S \ 2.1 \)), the adopted partial cross sections, and the source abundances themselves, this factor could actually lie anywhere between \( \sim 1.3 \) and \( \sim 2.6 \) (see, e.g., Clinton & Waddington 1993; Waddington 1996, 1997).

Our current knowledge of the “Pb”/“Pt” ratio comes essentially from the Ariel 6 and the HEAO C-3 spacecraft experiments (Fowler et al. 1987; Binns et al. 1989; Binns 1995a). Recent experiments on board the Long-Duration Exposure Facility have currently yielded only very preliminary results, which do not conflict with the earlier ones (O’Sullivan et al. 1995; Tylka et al. 1995; Domingo et al. 1995). Both available sets of data yield low “Pb”/“Pt” ratios, altogether consistent with an observed “Pb”/“Pt” ratio about 3.9 \( \pm 1.1 \) times lower than solar, which results in a source “Pb”/“Pt” ratio about 2.4 \( \pm 1.3 \) times lower than solar. “Pb” elements are all low-FIP elements \((\sim 7.4 \text{ eV})\), while “Pt” ones are mostly intermediate-FIP elements \((\sim 9 \text{ eV})\). Based on a plain FIP-biased solar source composition (Fig. 1), one would therefore expect a source “Pb”/“Pt” ratio slightly higher than solar, by a factor of \( \sim 1.6 \).

So, the actual source “Pb”/“Pt” ratio is about 3.9 \( \pm 2.0 \) times lower than would be expected, based on an FIP-biased solar source composition (see Fig. 1). This low ratio has been interpreted in terms of an excess of the third r-peak “Pt group” elements in the GCR sources (\( \S \ 2.2 \)). But it seems very difficult to have an excess of the third r-peak nuclei without any excess of the lighter r-nuclei (\( \S \ 2.2 \)). The other possible interpretation is that “Pb” is depleted relative to “Pt” because “Pb” elements are very volatile \((T_c \sim 500 \text{ K})\) while “Pt” elements are refractory \((T_c \sim 1400–1800 \text{ K}; \text{ Fig. 3})\).

In brief, the following picture emerges from this analysis (Fig. 1):

One very solid indicator, Ge, and three less foolproof but still significant indicators, Na, P, and Pb, point to volatility, not FIP, as the relevant parameter governing the GCRS composition.

The other five indicators, S, Zn, Se, Cu, and Ga, are consistent with either the FIP picture or volatility. In terms of volatility, the low S/(Zn, Se) ratio may seem problematic, and semivolatile Cu and Ga seem rather high. In \( \S \ 5 \), these apparent difficulties are interpreted in terms of a mass effect.

5. GCRS COMPOSITION: AN INTERPRETATION IN TERMS OF VOLATILITY AND MASS-TO-CHARGE RATIOS

The outcome of the above analysis of the clue elements is sufficiently suggestive to warrant a plot of the same GCRS overabundances relative to solar as in Figure 1, but this time versus \( T_c \) (Fig. 5). For the “highly volatile” elements, with \( T_c < 400 \text{ K} \), \( T_c \) has no physical relevance, and we have just ordered these elements by mass. Two conclusions can be drawn from Figure 5:

1. By and large, the enhancement of the refractory elements relative to the volatile ones is obvious. More specifically, the two intermediate “semivolatile” and “volatile” classes do tend to show intermediate overabundances, but with quite a large scatter.

2. By and large, the overabundances of the “highly volatile” elements seem to be an increasing function of their mass. One exception is H, which is slightly enhanced relative to He at a given energy per nucleon. Further, C and O do not follow the trend. But these are precisely the two elements for which we expect a specific contribution from W-R stars. In the Appendix, this W-R contribution is
roughly estimated for C, based on the W-R nucleosynthetic yields only, i.e., assuming that all the C lies in the gas phase. We have not considered a possible preferential acceleration of a significant fraction of the C that is locked in the grains formed in the C-rich WC wind material, in which C/O > 1 (see § 3). So, our assessment of the W-R C contribution may be an underestimate; that of the non-W-R C abundance, plotted in Figure 5, may therefore be an overestimate.

This leads us to suspect that the overabundances of the elements in the other classes of volatility might also be correlated with mass. We therefore plot in Figure 6 the same overabundance versus element mass, distinguishing the elements in the four classes of volatility. Figure 6 contains the essential conclusions of this paper:

1. The overabundances of most “highly volatile” elements are a strongly increasing function of their atomic mass number, roughly varying as $\propto A^{0.8 \pm 0.2}$.

2. Carbon and oxygen, the two very elements for which we expect a specific contribution from W-R stars (Appendix), are totally out of the correlation. As discussed above, we give in Figure 6 an assessment of the non-W-R C abundance, which may still be an overestimate. It agrees reasonably well with the trend given by the neighboring elements.

3. Hydrogen is also less depleted than expected based on the pattern for He and heavier elements, at least at a given energy per nucleon (the more relevant parameter for acceleration; Paper II). However, as we show in Paper II, a shock can simultaneously accelerate He less efficiently, and heavy volatile elements more efficiently, than H if the shock has a fairly low Mach number (e.g., $\sim 10$ or less) and all elements have the same temperature in the unshocked medium.

4. By contrast, there is only a very weak mass dependence of the refractory element overabundances, or none at all. It is well known, for instance, that the GCRS Fe/Mg ratio is close to solar, enhanced by 20% at the most. The current ultraheavy abundance estimates relative to Fe suggest modest excesses of most elements with $Z > 40$, but these analyses need confirmation (§§ 2.1 and 4).

5. Regarding the two intermediate classes of volatility, they show intermediate overabundances and fit beautifully into the picture. In particular, the low (Na, P)/(Cu, Ga) ratios in the “semivolatile” group, the low $S/(Zn, Se)$ ratio, and the high “Pb” abundance (if confirmed) among the “volatile” class (Fig. 5) are now readily interpreted in terms of a mass effect.

6. With the current errors, it is, of course, not possible to know whether the “volatile” elements behave significantly differently from the “highly volatile” ones or not.

Of course, the mass number $A$ is not a physical parameter capable of governing by itself the acceleration efficiency for the various elements. The observed rough mass dependence
of GCRS overabundances of the more volatile elements most likely just reflects an actual correlation with $A/Q$, i.e., a rigidity dependence of the acceleration efficiency (Paper II).

In any ionization situation, indeed, $A/Q$ is, by and large, a monotonically increasing function of $A$ (with local variations of this generally monotonic increase related to the electronic shell structure). Clearly, the appropriate abscissa scale in would have been $A/Q$, rather than $A$. But plotting an $A/Q$ scale would have required knowledge of the ionization states for all elements in the source gas, which would go far beyond the scope of the present work and will be investigated in a forthcoming paper. We have, however, stressed this point in by denoting the abscissa scale in Figure 6 would have been $A/Q$, rather than $A$. But plotting an $A/Q$ scale would have required knowledge of the ionization states for all elements in the source gas, which would go far beyond the scope of the present work and will be investigated in a forthcoming paper. We have, however, stressed this point in Figure 6 by denoting the abscissa scale “$A \sim (A/Q)^\alpha$,” leaving $\alpha$ unspecified.

Qualitatively, we can say that the accelerated gas cannot be a purely collisionally ionized gas around $\sim 10^4$ K since, in such a gas, Ne and He, for example, would be entirely neutral and hence not accelerated. It could be hot, $\sim 10^5$ K, gas in which grains have been somehow preserved, in which case the charge states $Q$ are a rather smooth function of $A$ and we obtain, very roughly, $A/Q \approx A^{0.4}$ (Arnaud & Rothenflug 1985). It might also be $\sim 10^4$ K gas photoionized by stellar UV radiation, in which case most elements will have charges of $Q = +1$ or $+2$, so that $A/Q = (0.5-1)A$; the pure mass scale on Figure 6 would then be relevant as an $A/Q$ scale, to within a leftward shift by a factor of 2 for the points representing the elements with $Q = +2$, i.e., those with a low second ionization potential. The somewhat low $N/^{20}\text{Ne}$ ratio (two elements with neighboring masses) could be understood in this context if $N$ was predominantly in the $N^{+\text{+}}$ state and $^{20}\text{Ne}$ in the $^{20}\text{Ne}^{+\text{+}}$ state. By contrast, whatever the charge state of He, $^{4}\text{He}^+$ or $^{4}\text{He}^{++}$, the high $H/\text{He}$ ratio cannot be understood in these terms, but it can be understood as a direct effect of shock acceleration (Paper II).

Note that $A/Q$-dependent abundance enhancements similar to those observed among the GCR volatiles are reported to exist in several heliospheric accelerated-particle populations:

1. Cummings & Stone (1996) claim that the “anomalous cosmic rays,” accelerated by the solar wind termination shock in the outer heliosphere, show an $A/Q$ enhancement, and they attribute this to the same effect of shock smoothing as discussed in Paper II for GCRs.

2. Smooth, $A/Q$-dependent enhancements are clearly found in gradual SEP events accelerated by coronal mass ejection–associated shocks in the corona and interplanetary medium (Mogro-Campero & Simpson 1972; Meyer 1985a; Breneman & Stone 1985; Stone 1989; Garrard & Stone 1993; Reames 1995). The $A/Q$ enhancements are found superposed upon the FIP bias of the coronal and solar wind composition relative to the photosphere (Meyer 1993, 1996); this FIP bias actually accounts for part of the bulk heavy-element enhancements relative to the photosphere noted early on (Mogro-Campero & Simpson 1972). Here, however, heavier, higher $A/Q$ elements, while most frequently enhanced, are also sometimes depleted relative to...
lighter ones. The depletions of heavier elements are mostly observed when the spacecraft is poorly connected to the flare site, while the enhancements are generally found in well-connected events (Cane, Reames & von Rosenvinge 1991). So shock smoothing, which can produce only heavier element enhancements (Paper II), is certainly not always the dominant factor shaping the accelerated solar particle composition. For instance, acceleration by weaker, less smoothed shocks, in a parallel or a perpendicular geometry, wave generation and saturation, variations in shock geometry, particle trapping and escape, and contributions of stochastic and resonant wave acceleration may all play important roles.

3. Note that the heavy-element enhancements found in impulsive, $^4$He-rich SEP events have quite different, specific characteristics and are to be explained in terms of resonant wave acceleration (see, e.g., Reames, Meyer, & von Rosenvinge 1994; Steinacker et al. 1997 and references therein).

4. Shock acceleration at the Earth bow shock has been studied in detail with in situ spacecraft measurements and comprehensive modeling (e.g., Jones & Ellison 1991). Clear evidence for nonlinear effects from efficient shock acceleration has been reported, including the $A/Q$ enhancement of diffuse heavy ions accelerated at the quasi-parallel portion of the shock (e.g., Ellison, Möbius, & Paschmann 1990). While the observed enhancements are modest compared with those seen in GCRs, they are fully consistent with nonlinear shock acceleration theory and, to our knowledge, have not been successfully explained by any alternative model.

6. SUMMARY AND CONCLUSIONS

We have shown that the GCR source (GCRS) abundances of all elements are best described (Fig. 6) in terms of (1) a general enhancement of the refractory elements relative to the volatile ones, and (2) among the volatile elements, an enhancement of the heavier elements relative to the lighter ones; this general trend accounts, in particular, for the well-known low abundances of $^4$He, $^6$He, and $^8$N in the GCRS. Besides $^4$C and $^16$O, for which a specific contribution is expected from the Wolf-Rayet He-burning material component responsible for the $^{22}$Ne excess, only $^4$H, slightly enhanced relative to $^6$He at a given energy per nucleon, does not entirely fit into this pattern; this mass dependence most likely reflects a mass-to-charge ($A/Q$) dependence of the acceleration efficiency. Among the refractory elements, there is no such enhancement of heavier species, or only a much weaker one.

This conclusion is based on a detailed analysis of the GCRS composition, in terms of both FIP and volatility. In particular, the GCRS $^24$Na/$^26$Mg, $^44$Ge/$^46$Ge, $^54$Pb/$^58$Pt, and $P/S$ ratios between elements of comparable FIP and mass, but widely different volatilities, are very difficult to interpret in terms of an FIP fractionation. Specifically, a combination of an FIP and a mass (or $A/Q$) fractionation could not account for them (see further discussion in Paper II).

We regard the strong similarity between the volatility-biased GCRS composition and the FIP-biased composition of the solar corona, wind, and energetic particles as coincidental. Note that this similarity is, indeed, not complete: crucial elements such as Na and P ($\S$ 4) do seem to behave differently in GCR sources and SEPs, where they clearly follow the FIP pattern (Garrard & Stone 1993; Reames 1995). By contrast, the $A/Q$-dependent enhancements found among the GCR volatiles and in various heliospheric accelerated-particle populations should, in several instances, have the same causes.

To confirm or disprove these views, new determinations of the GCRS abundances of all low-FIP volatile and high-FIP moderately volatile elements (in the bottom left and top right portions of Fig. 2, respectively) would be essential. In addition to the key elements already studied, i.e., Na, P, S, Cu, Zn, Ga, Ge, Se, and Pb, the following elements, whose GCRS abundances may become accessible in the future, can serve as clues: Cl, K, Mn, As, Br, Rb, Ag, Cd, In, Sn, Sb, Te, Cs, Au, Ti, and Bi.

In a companion paper (Paper II), this GCRS composition is interpreted in terms of an acceleration of interstellar and/or circumstellar gas and dust material by SNR shock waves. Such shock waves, smoothed by the feedback pressure of the very accelerated particles, preferentially inject and accelerate the higher rigidity ions. Among the ISM gas-phase volatile elements, they therefore enhance the higher $A/Q$, and hence in practice the more massive, elements (early suggestions of this effect were given by Eichler 1979, Ellison 1981, and Ellison, Jones, & Eichler 1981). The same shock waves treat the dust grains as extremely high $A/Q$ "ions" and accelerate them very efficiently to $\sim 0.1$ MeV nucleon$^{-1}$ energies, where friction and sputtering become important. The sputtered ions form a population of $\sim 0.1$ MeV nucleon$^{-1}$ refractory elements that can be further accelerated by the shock, and for which the crucial early acceleration phases have taken place while the ion was a member of the entire grain, hence independently of its own individual mass. So, both the presence of a strong mass dependence of the abundance enhancements among the volatile elements and its absence among the refractories may be understood consistently. Contrary to the earlier models in terms of FIP, such a picture accelerates the GCR ions in a single step in a single site. It also accounts naturally for the presence of an $^{22}$Ne-$^{12}$C-$^{16}$O excess in GCRs, since the shocks associated with the most massive supernovae accelerate their own pre-SN $^{22}$Ne-$^{12}$C-$^{16}$O-rich Wolf-Rayet wind material.

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APPENDIX

THE $^{22}$Ne-$^{12}$C-$^{16}$O-RICH COMPONENT

There exists one important exception to the absence of a signature of specific nucleosynthetic processes in the GCRS composition (§ 2.2). A large GCRS excess of the isotope $^{22}$Ne is derived from observations. The source ratio $^{22}$Ne/$^{20}$Ne $\approx 0.335 \pm 0.065$ (Łukasiak et al. 1994; DuVerneil et al. 1996a; Neher, Lukasiak, & McDonald 1997) is enhanced by a factor of about 4.4 $\pm$ 0.9 relative to the low solar reference ratio $^{22}$Ne/$^{20}$Ne$_{\odot}$ $\approx 0.076 \pm 0.005$, upon which planetary Ne-B, solar wind, and recent SEP and local ISM values derived from “anomalous cosmic ray” data now converge (Anders & Grevesse 1989; Selesnick et al. 1993; Wiedenbeck, Leske, & Cummings 1996; and references therein).

This large $^{22}$Ne excess, associated with the high GCRS C/O ratio (1.7 times solar), seems a clear signature of the presence of an He-burning material component in GCRs (Meyer 1981a, 1985b). The absence of observed signatures of other, unrelated, specific nucleosynthetic processes in the GCRS composition (§ 2.2) suggests an origin of the material in Wolf-Rayet star wind in which pure He-burning material is being expelled into space during the WC and WO stages, without other large anomalies (Cassé & Paul 1982; Maeder 1983; Prantzos, Arnould, & Arcoragi 1987; Maeder & Meynet 1993).

In massive stars such as W-R’s, the CNO cycle first turns the entire initial CNO into $^{14}$N. At the onset of He burning, this entire $^{14}$N in the He-burning layer is briefly turned into $^{18}$O, which itself is rapidly turned into $^{22}$Ne. The latter remains stable through most of the He-burning phase. Meanwhile, the $^4$He is progressively turned into $^{12}$C, and later into $^{16}$O by addition of another $^4$He. The $^{22}$Mg and $^{26}$Mg isotopes essentially start being formed only after the He-burning phase, largely through $^{22}$Ne destruction. In the case of W-R stars, huge winds peel off the star to the point that first its N-rich H-burning zone (WN phase), and then its C- and, later, O-rich He-burning core material, is being blown off into space (WC and WO stages; Prantzos et al. 1986, 1987; Maeder 1992; Schaller et al. 1992; Maeder & Meynet 1994; Meynet et al. 1994).

The time-averaged $^{22}$Ne and $^{12}$C enhancements relative to solar in the He-burning material can be roughly estimated from the initial stellar abundances (Meyer 1981a, 1985b). We now update the earlier estimates of these enhancements, based on new GCRS and reference solar $^{22}$Ne/$^{20}$Ne ratios (about half the previous SEP ratios that were often used in earlier work). As reference solar abundances, we adopt Grevesse et al.’s (1996) elemental abundances and the above $^{22}$Ne/$^{20}$Ne ratio, yielding $\Delta H \approx 2.75 \times 10^6$, $\Delta ^{16}$O $\approx 0.27 \times 10^6$, $\Delta ^{18}$O $\approx 980$, $\Delta$ CNO $\approx 3280$, and $\Delta ^{22}$Ne $\approx 23.4$, on a scale where $\Delta ^{16}$O $\equiv 100$. With these values, the conversion of all the initial CNO into $^{14}$N and then into $^{22}$Ne yields an $^{22}$Ne enhancement by a factor of CNO$_{\odot}/^{22}$Ne$_{\odot} \approx 3280/23.4 \approx 140$ in the He-burning layer. In the same He-burning layer, the maximal possible $^{12}$C enhancement is reached if all the initial $^4$He and $^{12}$C is converted into $^{12}$C by the $^4$He + $^4$He reaction, yielding a maximal possible $^{12}$C enhancement factor of $(H/4 + ^{12}$C)/$^{12}$C $\approx (2.75/4 + 0.27) \times 10^6/(3 \times 980) \approx 325$. This value is, however, not reached, because $^{12}$C starts being turned into $^{16}$O in the later stages of the He-burning phase. In addition, the $^{12}$C enhancement builds up progressively, so that the averaged enhancement during the He-burning phase should be about half the maximal value reached. We will therefore adopt an averaged $^{12}$C enhancement in the He-burning layer of about $35\% \pm 10\%$ of the above maximal possible value, i.e., about $115 \pm 33$.

To obtain the GCRS $^{22}$Ne/$^{20}$Ne enhancement factor of $4.4 \pm 0.9$, the He-burning component with $^{22}$Ne/$^{20}$Ne enhancement of 140 must be highly diluted in a component with solar $^{22}$Ne/$^{20}$Ne, by a factor of $140/(4.4 \pm (0.9-1)) \approx 44 \pm 12$. With the same dilution factor, the predicted $^{12}$C excess in GCRS sources is $1 \pm (115 \pm 33)/(44 \pm 12) \approx 4.0 \pm 1.6$. As a result, the carbon abundance in the main GCRS component, not affected by W-R He-burning nucleosynthesis, is $\sim 1/(4.0 \pm 1.6)$, i.e., $\sim 18\%$–$42\%$ of the total GCRS carbon abundance. This rough estimate has been plotted in Figures 1, 5, and 6. It is based on W-R nucleosynthesis properties only. If, in addition, a fraction of the WC wind carbon is preferentially accelerated because it has condensed in solid carbon compounds in the C-rich WC star wind material, the fraction of the remaining GCRS carbon originating in the main, non-W-R, GCR component may be even smaller.

Which accompanying composition anomalies should one expect? At the onset of He burning, $^{14}$N is first turned into $^{18}$O. The smaller the stellar mass, the longer this $^{18}$O survives before being converted into $^{22}$Ne. We might therefore find an associated $^{18}$O excess, if some stars are peeled off before reaching the W-R core before $^{18}$O has been destroyed. Further, if WC-type W-R star wind material contributes to GCRs, we expect similar contributions from the wind material of the preceding and subsequent N-rich WN and O-rich WO phases. The expected associated N and O excesses in GCRs, however, cannot be evaluated with any certainty. The W-R star wind yields of N and O, relative to those of $^{22}$Ne and C, indeed depend critically upon the still-unsettled time profile of the mass-loss rate as the star evolves, and upon the degree of mixing between stellar layers (Maeder 1992; Maeder & Meynet 1994; G. Meynet 1996, private communication). In addition, the efficiency of wind material acceleration into GCRs may also be a function of stellar mass. In the WN phase, CNO-cycle H-burning material is being expelled in the wind. In this material, the N enhancement is equal to CNO$_{\odot}/^{14}$N $\approx 3280/257 \approx 12.8$ only. The actual expected N excess depends upon the relative strengths of the winds in the WN and WC phases (see, e.g., Maeder 1992, Maeder & Meynet 1994). If the WN and WC wind contributions are very roughly comparable, as could be the case, this N-rich component, also diluted by a factor of order of $\sim 44$, yields a GCRS N excess by a factor of $\sim 1 + 12.8/44 \approx 1.29$ only. In the WO phase, O-rich gas, due to the $^{12}$C + $^{18}$O reaction in the later stages of He burning, is being expelled in the Wolf-Rayet wind. So, we also expect an $^{16}$O yield in the wind depends critically upon the degree of nuclear evolution of the He core at the time it gets significantly peeled off by the winds. Finally, the conversion of the $^{22}$Ne into $^{25}$Mg and $^{28}$Mg takes place only very late in the He-burning phase and can probably be neglected.
in the W-R wind material.\(^2\)

The GCRS isotopic ratios derived from observations are consistent with the above features: recent analyses suggest a low \(^{13}\text{C}/^{12}\text{C}\) ratio (i.e., a \(^{12}\text{C}\) excess) and possibly a high \(^{18}\text{O}/^{16}\text{O}\) ratio (yet to be confirmed) and indicate the absence of a significant \((^{25}\text{Mg} + ^{26}\text{Mg})/^{24}\text{Mg}\) enhancement (Ferrando 1994; Lukasiak et al. 1994; DuVernois et al. 1996a; Webber et al. 1997).

\(^2\) In a more precise, but more model dependent, approach, we can consider the calculated total yields of \(^{12}\text{C},^{14}\text{N},^{16}\text{O},\) and \(^{22}\text{Ne}\) from W-R star winds, by integrating the wind contribution of all W-R stars over the stellar initial mass function. Normalized to the above \(^{12}\text{C}\) yield, we find that the GCRS isotopic ratios derived from observations are consistent with the above features: recent analyses suggest a low \(^{13}\text{C}/^{12}\text{C}\) ratio (i.e., a \(^{12}\text{C}\) excess) and possibly a high \(^{18}\text{O}/^{16}\text{O}\) ratio (yet to be confirmed) and indicate the absence of a significant \((^{25}\text{Mg} + ^{26}\text{Mg})/^{24}\text{Mg}\) enhancement (Ferrando 1994; Lukasiak et al. 1994; DuVernois et al. 1996a; Webber et al. 1997).
by a factor of 196

and 26

in contrast to our suggestion in the text [unless the accepted thermonuclear cross sections for 

12

D

star nucleosynthesis, although recent assessments have yielded uncomfortably low amounts of phases of He burning. This suggested that the observed O excess in the GCRS could presumably be accounted for by W-R 22

ratio from W-R nucleosynthesis is rather model independent in the W-R ejecta (Appendix).

22

AIP], 245 [1989], scaled to O/Si throughout the ISM (J.-P. Meyer, in AIP Conf. Proc. 183, Cosmic Abundances of Matter, ed. C. J. Waddington [New York: AIP], 72). However, some 15% of the O is certainly trapped in silicates, the principal constituents of the grain cores of our interest in the text [unless the accepted thermonuclear cross sections for 

12

C

25

Mg, and 

26

Mg are made and expelled at about the same time, so that the expected 

16

O/(23

Mg, 24

Mg) ratio from W-R nucleosynthesis is rather model independent (Maeder 1983; Prantzos et al. 1986, 1987; Maeder & Meynet 1993). We have now realized that all these calculations yield comparable 

16

O, 25

Mg, and 26

Mg excesses, within a factor of ~2. The lack of significant GCRS excesses of 22

Ne and 24

Mg (Ferrando 1994; Lukasiak et al. 1994; DuVernois et al. 1996a; Webber et al. 1997) therefore indicates that the O excess probably cannot be interpreted in terms of W-R star nucleosynthesis, in contrast to our suggestion in the text [unless the accepted thermonuclear cross sections for 

12

C(x, g)16

O and 

22

Ne(x, n)25

Mg, known to within factors of ~3, are very incorrect].

On the other hand, while we have considered the likely trapping of a large fraction of C in solid carbon compounds within the C-rich WC atmosphere (§3, 5), we overlooked in the text any possible trapping of O in ISM grains. The trapping of a large fraction of O is actually excluded, i.e., ice mantles do not survive in the environments we consider, and the amount of O trapped in organic refractory mantles seems negligible (W. A. Schutte, Ph.D. thesis, Univ. Leiden [1988]; W. A. Schutte & J. M. Greenberg, in Dust in the Universe, ed. M. E. Bailey & D. A. Williams [Cambridge: Cambridge Univ. Press], 403 [1988]). However, some 15%–20% of the O is certainly trapped in silicates, the principal constituents of the grain cores throughout the ISM (J.-P. Meyer, in AIP Conf. Proc. 183, Cosmic Abundances of Matter, ed. C. J. Waddington [New York: AIP], 245 [1989], scaled to O/Si = 20.4 from Grevesse et al. 1996). Now, from the abundances of N, 20

Ne, and the refractories in Figure 6, we estimate that grain material is accelerated ~10 times as efficiently as the gas-phase O (like 20

Ne, N is probably not significantly trapped in grains; see, e.g., B. Mason, Data of Geochemistry/Cosmochemistry/Meteorites, Geol. Surv. Professional Pap. 440-B-1 [Washington: GPO] 1979; C. M. Sharp & G. J. Wasserburg, Geochem. Cosmochem. Acta, 59, 1633 [1995]; Mathis 1996). With 15%–20% of the O in grains, which gets 10-fold preferentially accelerated, the O excess by a factor of ~3 is readily accounted for within the framework of our model.

Note added in proof.—A remark by M. DuVernois has led us to reconsider our interpretation of the GCRS oxygen excess by a factor of ~3 relative to the neighboring volatile species, N and 

20

Ne (Fig. 6). As discussed in the Appendix, 

16

O, 23

Mg, and 26

Mg are not produced by W-R stars at the same stage as 

12

C and 

22

Ne. Ratios such as 

16

O or 

23

Mg, 26

Mg/12

C or 

22

Ne are therefore quite model dependent, strongly affected by the relative timescales and wind strengths during the various phases of He burning. This suggested that the observed O excess in the GCRS could presumably be accounted for by W-R star nucleosynthesis, although recent assessments have yielded uncomfortably low amounts of 

16

O, with 

16

O/12

C ~ 9% only in the W-R ejecta (Appendix).

By contrast, 

16

O, 23

Mg, and 26

Mg are made and expelled at about the same time, so that the expected 

16

O/(23

Mg, 24

Mg) ratio from W-R nucleosynthesis is rather model independent (Maeder 1983; Prantzos et al. 1986, 1987; Maeder & Meynet 1993). We have now realized that all these calculations yield comparable 

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O, 25

Mg, and 26

Mg excesses, within a factor of ~2. The lack of significant GCRS excesses of 22

Ne and 24

Mg (Ferrando 1994; Lukasiak et al. 1994; DuVernois et al. 1996a; Webber et al. 1997) therefore indicates that the O excess probably cannot be interpreted in terms of W-R star nucleosynthesis, in contrast to our suggestion in the text [unless the accepted thermonuclear cross sections for 

12

C(x, g)16

O and 

22

Ne(x, n)25

Mg, known to within factors of ~3, are very incorrect].

On the other hand, while we have considered the likely trapping of a large fraction of C in solid carbon compounds within the C-rich WC atmosphere (§3, 5), we overlooked in the text any possible trapping of O in ISM grains. The trapping of a large fraction of O is actually excluded, i.e., ice mantles do not survive in the environments we consider, and the amount of O trapped in organic refractory mantles seems negligible (W. A. Schutte, Ph.D. thesis, Univ. Leiden [1988]; W. A. Schutte & J. M. Greenberg, in Dust in the Universe, ed. M. E. Bailey & D. A. Williams [Cambridge: Cambridge Univ. Press], 403 [1988]). However, some 15%–20% of the O is certainly trapped in silicates, the principal constituents of the grain cores throughout the ISM (J.-P. Meyer, in AIP Conf. Proc. 183, Cosmic Abundances of Matter, ed. C. J. Waddington [New York: AIP], 245 [1989], scaled to O/Si = 20.4 from Grevesse et al. 1996). Now, from the abundances of N, 20

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