ISOTOPIC ANOMALIES: CHEMICAL MEMORY OF GALACTIC EVOLUTION

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ABSTRACT

Because of the differing temporal evolution of primary and secondary products of nucleosynthesis, which I illustrate with a standard family of analytic models of chemical evolution, the isotopic composition of the bulk interstellar medium changes approximately linearly with time. Therefore, any dust component having an age different from that of average dust will be isotopically anomalous. I give three explicit and plausible mechanisms for this that do not rely on special stellar condensates, with special emphasis on C, O, Mg, and Si and on suggestive examples from meteoritic science: isotopically heavy average-stellar condensates of SiC and a correlation of excess $^{16}$O with aluminum. These evolutionary effects add a new dimension to the chemical-memory interpretations of the isotopic anomalies.

Subject heading: nucleosynthesis

I. INTRODUCTION

In this paper I introduce new mechanisms for the chemical memory of isotopic anomalies based on the temporal change during the chemical evolution of the Galaxy of the isotopic composition of the mean ejecta from stars. The words mean ejecta are quite important to this idea, for they ask the reader to suppress temporarily considerations of the wide variations in compositions among individual supernova condensates (SUNOCNs) (Clayton 1978) expected from differing zones of different stars. The key idea instead is that both the bulk isotopic composition of the interstellar medium and the bulk isotopic composition of the total instantaneous ejecta from stars, summed over all types of stars, change monotonically and predictably as the Galaxy ages. Thus any physical mechanism (of which I will give examples) that partially segregates "old ejecta" from "young ejecta" or "old dust" from "young dust" in the interstellar medium will also create macroscopic pools of differing isotopic composition. These pools will become differentially mapped into the spectrum of molecular cloud dust particles by the cyclic interstellar processes that evolve the interstellar dust. The resulting isotopic memory may be recovered to some degree during the selective histories leading to the aggregation of meteoritic samples.

The key nucleosynthesis idea is the distinction between primary and secondary nucleosynthesis products. A primary nucleosynthesis product is synthesized even in stars born from hydrogen and helium only, whereas a secondary nucleosynthesis product is produced in proportion to the concentration of heavier elements (usually C and O) initially present when the star formed. The most abundant nuclear species (viz., $^{12}$C, $^{16}$O, $^{20}$Ne, $^{24}$Mg, $^{26}$Si, $^{32}$S, $^{36}$Ar, $^{40}$Ca, $^{44}$Ca, $^{48}$Ti, $^{52}$Cr, $^{56}$Fe) are all primary products, whereas the least abundant isotopes of those elements are in varying degrees secondary. I here single out $^{13}$C, $^{17}$O, $^{21}$Ne, $^{25}$Mg, $^{29}$Si, and $^{30}$Si as having yields approximately proportional to the initial C plus O content of the stars. This initial C plus O content has itself grown with time, as has therefore the secondary component of the isotopic composition of stellar ejecta. These nucleosynthesis ideas have a long and rich history, but the most useful recent quantitative discussion (Woosley and Weaver 1982) presents the nucleosynthesis comparison of two 25 $M_{\odot}$ stars of differing population.

The new mechanisms augment the general theme of cosmic chemical memory, an astronomical attempt (Clayton 1982) to measure dust formation and evolution by interpreting the isotopic anomalies within meteorites. That approach would be misguided if the isotopic anomalies reflected instead a spatially inhomogeneous admixture into the presolar cloud of anomalous nucleosynthesis products from nucleosynthesis events near the forming Sun (Cameron and Truran 1977; Wasserburg and Papanastassiou 1982; Hinton, Davis, and Scaterna-Wachal 1987). But I believe that a chemical-history interpretation suffers less conflict with isotopic data and is also more plausible, and I therefore enlarge the basis of those interpretations by considering how the temporal evolution of the Galaxy may be reflected in the isotopic compositions of the interstellar dust.

II. CHEMICAL (ABUNDANCE) EVOLUTION

Let $Z_{1}(t)$ be the bulk interstellar concentration of a primary nucleosynthesis product, and $Z_{2}(t)$ that of a secondary nucleosynthesis product. To achieve explicit expressions for these concentrations, Clayton (1984, 1985) has constructed exact analytic models having time-dependent infall $f(t)$ onto the solar annulus of the Galactic disk with the aid of these plausible physical assumptions: star formation rate $\psi(t)$ proportional to gas mass $M_{d}(t)$; instantaneous recycling approximation; and constant yield $y_{1}$ of primary nucleosynthesis products. Clayton and Pantelaki (1986) have added the exact solutions of secondary nucleosynthesis products if their yields $y_{2} = \beta Z_{2}(t)$ are proportional to the concentration of the primary seed nuclei. With metal-free infall $f(t)$, each abundance satisfies

$$\frac{dZ}{dt} = y_{1} - \frac{Z_{1}(t)}{M_{d}(t)} f(t),$$

where $y_{1}$ is the appropriate yield and the constant $\omega$ is defined by the linear dependence of star formation $\psi(t) = [\omega(1 - R)]M_{d}(t)$, where $R$ is the return fraction. I will illustrate the ideas of this paper in terms of Clayton's (1985) standard-infall models which are defined by the time-dependent infall $f(t) = [kM_{d}(0)/\Delta][1 + \Delta]/\Delta e^{-\omega t}$, with $k$ an integer and $\Delta$ the time parameter to shape infall. Within this family the exact solutions in terms of the time variable
with the average isotopic composition of stellar ejecta at that time. At \( t = t_0 \), just immediately before solar birth, the average interstellar isotopic composition is \( Z_1/Z_0 = (Z_1)_0/(Z_0)_0 \); but the average isotopic composition currently being ejected from stars is different. That average ejecta can easily be analyzed in the linear star formation models with instantaneous recycling, because the general differential equation (viz., Clayton 1984, eq. [7]) for the interstellar mass growth of element \( Z \),

\[
\frac{d}{dt}(ZM_d) = \gamma(1 - R)\Psi + RZ\Psi - Z\Psi + Z_f f = \frac{d}{dt}(M_Z),
\]

(4)

yields immediately the new nucleosynthesis and the return from stars as the first and second terms, respectively. The ratio of the rates of injection of primary and secondary nuclei is then (with metal-free infall \( Z_f = 0 \))

\[
\left( \frac{dM_1}{dt} \right)_{\text{inj ecta}}^1 = \frac{y_1 + rZ_1}{y_s + rZ_s},
\]

(5)

where \( r = R/(1 - R) \) \approx 0.5 if the return fraction \( R \) is approximately \( \frac{1}{2} \). This may be combined with equation (1) to give

\[
\left( \frac{dM_1}{dt} \right)_{\text{inj ecta}} = \frac{dZ_1}{dt} + Z_1(\rho + f\lambda M_d),
\]

\[
= \frac{dZ_1}{dt} + Z_1(\rho + k(t + \Delta)),
\]

(6)

where the second equality follows from \( f\lambda M_d = k(t + \Delta) \) in the two-parameter representation of infall in the Clayton (1985) standard model. Although equation (6) is exact, a simpler approximation for this introductory discussion results from the quasi-linear growth of \( Z_1 \) and the quasi-parabolic growth of \( Z \), by noting that with zero initial metallicity \( dZ_1/dt \approx Z_1/t \) and \( dZ_2/dt \approx 2Z_1/t \) at the time the solar system was forming. Near \( t = t_0 \) an approximate version of equation (6) is therefore

\[
\left( \frac{dM_1}{dt} \right)_{\text{inj ecta}} \approx \frac{1}{2} \left( \frac{Z_1}{Z_0} \right)^{1 + k + \rho \omega t_0}/(1 + k + \rho \omega t_0)/2.
\]

(7)

Consider first the infall-free \( (k = 0) \) galaxy. If it was \( t_0 = 6 \) Gyr old when the Sun formed, the value of \( \omega t_0 \) can be estimated in two ways: (1) \( \omega t_0 \approx 2 \) if the astration time of the interstellar medium (ISM) is estimated as \( \omega^{-1} \approx 3 \text{ Gyr} \); (2) \( \omega t_0 \approx 1.3 \) if the exponential gas decline must give 10% gas today at \( t_0 + 4.6 \text{ Gyr} \). Thus \( \omega t_0 \approx 0.8 \) if \( \omega \approx 0.5 \) and \( \omega t_0 \approx 1.6 \). With this estimate \( dM_1/dt \) at \( t = t_0 \) near \( t = t_0 \). As an example, the isotopic composition of silicon being ejected from stars shortly prior to solar birth is \( d(28,30)\text{Si})/d(28\text{Si}) = \)

### TABLE 1

| PRIMARY AND SECONDARY METALLICITIES* (CLAYTON 1985 STANDARD MODEL) |
|-----------------------------|-----------------------------|
| Infall                      | \( Z_1/Z_0 \)               | \( Z_0/Z_0 \)               |
|-----------------------------|-----------------------------|
| \( k = 0 \) (closed box)    | \( t_0 \)                   | \( (t_0)^2 \)               |
| \( k = 1 \) (exponential)   | \( t \) \( (t + 2\Delta) \) \( t_0 + \Delta \) \( t_0 + 2\Delta \)  \( t \) \( t_0 + \Delta \) \( t_0 + 3\Delta \)  \( t \) \( t_0 + \Delta \) \( t_0 + 3\Delta \)  |
| \( k = 2 \) (e^{-\omega})   | \( t \) \( (t^2 + 2\Delta \text{A} + 3\Delta^2) \) \( t_0 + \Delta \) \( t_0 + 2\Delta \text{A} + 3\Delta^2 \)  \( t \) \( t_0 + \Delta \) \( t_0 + 2\Delta \text{A} + 3\Delta^2 \)  |

* Derived from results in Clayton and Panteelaki 1986.
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1.56(29,30Si/28Si): i.e., current silicon ejection from all stars (assuming 29,30Si to be entirely secondary, a modest exaggeration [cf. Woosley and Weaver 1982, Table 5]) carries a 56% enrichment of the heavy-isotope fractions of solar silicon. This is a very large isotopic anomaly 29,30δ = 560 in the parts-per-thousand deviations common in meteoritics literature. For larger integer values of k (more infall) the excess becomes smaller. Retaining root 2 = 0.8, they would be 36% for k = 1 and 26% for k = 2.

To illustrate the idea even further, image that some fixed percentage (from all stellar types) of silicon ejecta condenses as refractory SiC, and that the carbon also carries the mean isotopic composition of its current bulk ejection. That SiC would then be isotopically heavy in both Si and C. This could be related to the isotopically heavy SiC discovered by Zinner, Ming, and Anders (1988), even though in detail the true situation must be more complicated because the measured Si anomalies are smaller than the measured C anomaly in those samples. Jorgensen (1988) addresses this, for example, by arguing for specific condensates from carbon star atmospheres, one of the many specific-condensate anomalies that are superposed on the mean ejecta. I agree with that approach. STAR-DUST (stellar matter thermally condensed during ejection from stars) will condense the ambient isotopic composition within each star, and the mean isotopic composition of STAR-DUST will not equal the mean isotopic composition of the bulk ejecta. But my purpose here is to clarify how the mean current ejecta is already anomalous.

It should be added that one does expect thermal refractory condensates from stars to be much younger than the mean interstellar atoms. Sputtering and astration destroy the specific stellar condensates, whereas the old atoms are cyclically sputtered and reaccreted as grain mantles. Liffman and Clayton (1988) have studied these cycles in numerical detail. But it follows from the youth of the mean refractory condensates that they are isotopically heavy with respect to the much older bulk nuclei that have partaken in the sputtering cycles of the ISM.

For the cosmochemically important element Mg one notes that, owing to the 24Mg deficiency in the bulk recent ejecta, young ejecta could be erroneously interpreted as bearing large positive mass-dependent fractionation on the basis of the 25/24 ratio alone. If a sample containing it were to be renormalized to (25/24)$_0$, according to experimental custom, it would then appear deficient in 25Mg. The question of both in situ and fossil radiogenic 26Mg (Clayton 1986a) would then be more complicated than is normally assumed. Some meteorite samples having small anomalies of these correlated signs do exist, although it is not my purpose to argue this mechanism to be their cause.

b) Mechanism 2: Surface-correlated Accretion of Current Ejecta

Suppose instead that the current ejecta from bulk stars is entirely gaseous. It carries excess heavy isotopes, again according to equation (6); but now concentrate on totally gaseous ejection from stars. That current ejecta (at t = t$_{ej}$) will be accreted by preexisting interstellar grains before the molecular cloud collapse that formed the solar system. The grains will accrete this gas in proportion to their surface areas, so that the smallest grains will, in comparison with their masses, have the largest component of isotopically heavy mantles. Clayton (1980) has discussed this general idea. As an illustrative reinterpretation of the preceding example, the isotopically heavy SiC might then be interpreted as SiC made from an aggregate of the smallest particles. Although I do not argue that this is the correct interpretation of those SiC data, I do point out that dynamic collapse and aggregation can easily perform some sorting of grains according to size. Analogous remarks apply to magnesium.

c) Mechanism 3: Different Age Spectrum of Differing Dust Types

For simplicity in comprehending this mechanism, take all stellar ejecta to be gaseous, and assume that the entire spectrum of interstellar dust is the natural result of the sputtering and fragmentation, followed by reaccretion, interspersed with chemical migrations and annealing that is the logical consequence of a cyclic ISM, with all its varied environments. During such cyclical histories, some chemical structures will quite naturally survive longer than others. The refractory structures so formed will quite naturally have a greater average age than the more volatile and more transient structures, and they will therefore be slightly deficient in secondary isotopes. Consider first in this regard the element aluminum, whose great depletion factors from interstellar gas confirm that elemental Al spends a smaller fraction of its time in interstellar gas than does an average element: i.e., the Al structures in dust are on average older than structures composed of other elements.

This conclusion follows from the assumption that the number N(Z) of gas atoms of refractory element Z are created by a steady state between production by sputtering and fragmentation of grains with rate λ$_+$ (Z) and depletion by sticking with efficiency E onto grains having surface area σ:

$$\frac{dN(Z)}{dt} = \lambda_+ - E\sigma N(Z). \quad (8)$$

If it is assumed that the refractory atoms stick with equal efficiencies E, the steady state solution yields N(Z) = λ$_-$, so that the largest depletions (Al, Ca, Ti) occur for those elements that are volatilized with lowest efficiency λ$_-$ by sputtering and fragmentation. But it is equally clear that the mean time spent in a grain is proportional to λ$_-$, so those elements with smallest fractional gas abundance N(Z) have on average resided longer between volatilizations. Aluminum itself has but a single stable isotope; however, it binds itself energetically with oxygen as Al$_2$O$_3$ structures within oxidized grains. If it is assumed that this oxygen has the isotopic composition of average interstellar oxygen at the time the Al$_2$O$_3$ structures formed, the average age of that oxygen structure will be greater than that of average oxygen structures, which means that it will be smaller in $^{17}$O and $^{18}$O content, considering that those two heavy oxygen isotopes are secondary nucleosynthesis products. By this means interstellar Al is associated with $^{16}$O-rich oxygen. Such an association is suggested by meteoritic data showing that the aluminum-rich inclusions of carbonaceous meteorites are $^{16}$O-rich (Clayton et al. 1977). Meteoriticists have not themselves endorsed the idea that $^{16}$O excess was carried into the parents of the inclusions as Al$_2$O$_3$; but I have long argued (Clayton 1982, 1986b) that it is so, primarily because the SUNOCONs of newly synthesized aluminum are expected to be $^{16}$O-pure. The present idea also associates $^{16}$O excess with aluminum, but in this case because aluminum oxide is the oldest oxide within interstellar dust. This is the first argument associating $^{16}$O with aluminum that has not related
on the injection initially into the ISM of $^{16}$O-pure Al-rich SUNOCONS. Such surviving SUNOCONS are here seen only to strengthen a correlation that would nonetheless exist even in their absence. Furthermore, the magnitudes are reasonable. Because the $^{17}$O/$^{16}$O ratio increases rather linearly in time, a 5% excess of $^{16}$O (as observed in meteorites) within supernova ejecta would require only that they be formed from dust components that are older than bulk interstellar dust by 5% of the age of the Galaxy. This is easily justified by observing that Al resides roughly 10-fold longer in dust than do other elements because it is 10-fold less gaseous than other elements. Therefore, this mechanism must be regarded as being at least as attractive as the beautiful earlier explanation based on $^{16}$O-rich, Al$_2$O$_3$ SUNOCONS. I personally find the existence of two extremely attractive reasons for associating $^{16}$O excess with interstellar Al altogether remarkable.

Lifman and Clayton (1988) have quantitatively modeled the idea that refractory grain cores have, as entities, lifetimes considerably longer than the bulk mass lifetime of dust against sputtering. $\tau_{\text{sputter}} = M/M_{\text{sputter}}$. They constructed Monte Carlo histories of layered interstellar particles. Although their astrophysical model was oversimplified in its neglect of shattering and of carbonaceous mantles, their computations nonetheless showed that about 20% of the refractory cores (their phase A) formed during $6 \times 10^9$ yr of interstellar evolution survive even though the mass lifetime $\tau_{\text{sputter}}$ of bulk refractory atoms is only $2.61 \times 10^8$ yr (in their case 1b for thermal sputtering). Setting their 20% survival equal to $\tau_0 (1 - e^{-T/\tau})$ as the approximate definition of a mean survival time of their phase A cores yields $\tau_0 = 1.2 \times 10^9$ yr, considerably greater than $\tau_{\text{sputter}} = 2.61 \times 10^8$ yr. Therefore, if those phase A cores formed in the dust medium from ambient oxygen (rather than being injected as SUNOCONS), their $^{16}$O excess would be more than adequate. Although the oxygen within Al$_2$O$_3$ phase A cores is expected by this mechanism to carry the largest $^{16}$O excess, I note that even the bulk refractory dust is significantly enriched. Even if the distinction of the phase A cores is suppressed, a mass lifetime of $2.6 \times 10^9$ yr implies that the bulk refractory dust has an age that is several percent of that of the Galaxy, giving it a comparable percentage excess of $^{16}$O with respect to the bulk gas. So it is not necessary to identify Al as the only carrier—just as the most abundant carrier.

I must add that an exceedingly fascinating parallel exists to magnesium. MgO units join Al$_2$O$_3$ units to form highly refractory spinel, MgAl$_2$O$_4$. Spinel crystals found in meteorites are the most $^{16}$O-rich minerals found there ($^{16}$O $= +50$ parts per thousand). But by the same reasoning the Mg atoms in interstellar spinel should be older structures than those containing the remaining Mg atoms. This leads to a remarkable expectation for the Mg isotopic composition within spinel—namely, that it be $^{24}$Mg-rich at the approximate level $^{24}$Mg $\approx 1.5$. Such a large $^{24}$Mg excess has never been seen in meteorite samples; but it could be one component of meteoritic Mg showing small $^{24}$Mg excess. My point is not to press that identification but rather to note that if such a sample were to be renormalized by mass-dependent isotopic fractionation on the assumption that the ratio $^{24}$Mg/$^{25}$Mg is solar (a procedure often used in measurements on meteorites), the $^{24}$Mg excess would appear to be instead a $^{26}$Mg excess with $^{26}$Mg $= 1.5$. This is the first time that interstellar history has predicted an "apparent $^{26}$Mg excess" in interstellar Al without benefit of $^{26}$Al decay. Although I do not claim that this new effect, rather than $^{26}$Al decay, is the source of the $^{26}$Mg richness found in certain meteoritic Al-rich inclusions (Wasserburg and Papanastassiou 1982), I do suggest that it could play a role in the complicated and poorly understood Mg isotopic systematics (Clayton 1986a). Clayton and Leising (1987) show that the gamma-ray measurements themselves imply that $^{26}$Al also must play a large role in that systematics. My brief discussion in this work calls out only the dramatic consequences that chemical and abundance evolution can introduce.

IV. DISCUSSION

One must remember that the interstellar medium contains highly fractionated components, both chemically and isotopically. Clayton (1978) argued that this fractionated state was remembered in many ways by the partially closed chemical reorganizations within the meteorites, a mechanism named "cosmic chemical memory" by Clayton (1982). These papers emphasized the role of very isotopically anomalous SUNOCONS. Clayton (1978) had the temerity to present a table of refractory SUNOCONS and their isotopic constitutions. Their huge isotopic anomalies dwarf in magnitude those found in meteorites; but the macroscopic meteorite samples dwarf in mass the microscopic interstellar SUNOCONS. The idea was that small anomalous elements in macroscopic meteorite samples are what remain following the chemical exchange and dilution involved in aggregating and annealing a large number of small particles within a warm solar accretion disk.

The present discussion suppresses these dramatic variations within small individual particles, not because they do not exist but because it is easier to then envision the systematic isotopic evolution of the bulk ISM and the systematic isotopic anomalies that may thereby be preserved in different classes of dust particles. Considering that the term "isotopically anomalous" means nothing more than "differing from the bulk Sun," one realizes that the entire ISM was isotopically anomalous $10^9$ yr, say, before the solar system formed. I have presented three very plausible mechanisms by which these systematic anomalies may infect whole classes of particles. These considerations provide the baseline upon which the isotopically anomalous SUNOCONS and STARDUST are superposed as especially anomalous components. What I have shown by examples is that some of the most exciting and fundamental isotopic anomalies are potentially impacted by these new mechanisms, which therefore demand continuing study as part of the total fabric of cosmic chemical-memory theories of the isotopic anomalies. One of the advantages of the new mechanisms is that they allow one to see how particles may be sorted by dynamic effects (not addressed here) or how different components of dust may be sorted by subsequent chemical reorganization effects (also not addressed here) to leave macroscopic samples that are isotopically anomalous. It has been a difficult challenge to accomplish this within the theory based on SUNOCONS and STARDUST alone, although it has been possible to do so. Lifman and Clayton (1988) present especially clear and detailed discussion of how this may be effected by the remapping algorithms established by sputtering and reaccretion cycles. But it must be admitted that these new mechanisms have a startling simplicity.

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