

## The neon alphabet game

D. D. Sabu<sup>1</sup> and O. K. Manuel<sup>1,2</sup>

<sup>1</sup>Department of Chemistry, University of Missouri, Rolla, Missouri 65401

<sup>2</sup>U.S. Geological Survey, Box 20546, Denver Federal Center, Denver, Colorado 80225

**Abstract**—All previously identified components of trapped meteoritic neon—Ne-A, Ne-B, Ne-C, Ne-D, Ne-E, Ne-A1, Ne-A2, Ne-E(L), Ne-E(H), and Ne-O—and differences between the isotopic compositions of atmospheric, meteoritic, and solar-wind-implanted neon can be accounted for by mass fractionation and spallation reactions. These same two well-known processes can account for the experimental data that have previously been assigned to nucleogenetic anomalies in meteoritic neon, although the data cannot refute that hypothesis.

The neon record of events in the early history of the solar system includes evidence of (i) severe fractionation, (ii) spallogenic <sup>21</sup>Ne from an early irradiation of material prior to the compaction of meteorites, (iii) a close association of planetary helium and neon with isotopically anomalous argon, krypton, and xenon, and (iv) the total absence of helium and neon in the noble gas component which contains isotopically normal or planetary-type argon, krypton, and xenon.

Experimental difficulties in the detection of extremely low values of the <sup>20</sup>Ne/<sup>22</sup>Ne ratio are presented in the Appendix.

## INTRODUCTION

The discovery of nucleogenetic anomalies in the isotopes of other elements (see review by Podosek, 1978) has revived interest in possible nucleogenetic anomalies of meteoritic neon. The identification and alphabetical labeling of trapped meteoritic neon components began over a decade ago, with reports that diffusive fractionation is not responsible for any of the observed isotopic variations (Pepin, 1967, 1968).

Early studies of trapped meteoritic neon, which is evolved by stepwise heating of bulk meteorites, identified five isotopically distinct components: Ne-A, Ne-B, Ne-C, Ne-D, and Ne-E (Pepin, 1967, 1968; Black and Pepin, 1969; Black, 1972a,b,c). Recent studies of neon in mineral separates of meteorites have identified subdivisions of Ne-A and Ne-E, *viz.*, Ne-A1, Ne-A2, Ne-E(L), and Ne-E(H) (Alaerts *et al.*, 1979a,b, 1980; Eberhardt, 1978; Eberhardt *et al.*, 1979a,b; Lewis *et al.*, 1979; and references therein), and an eighth neon component, Ne-O, has also been discovered (Eberhardt, 1978).

There are at least two other types of isotopically distinct neon in the solar system, one in the solar wind (Geiss *et al.*, 1972) and the other in the earth's atmosphere (Nier, 1950), and cosmic ray-induced spallation reactions produce

yet another. In spite of frequent interference from these, there has been no respite in the discovery of new types of trapped meteoritic neon. The discovery of so many isotopically distinct components of trapped neon, an element with only *three* stable isotopes, is an intriguing singularity in the study of the elements in meteorites.

Srinivasan and Anders (1978) reported that Ne-E accompanies excess  $^{128-132}\text{Xe}$  isotopes in the etched residue, 1C10, of the Murchison carbonaceous chondrite. The anomalous Xe isotopes were attributed to the *s*-process of nucleosynthesis, and the presence of Ne-E in the same residues seemed to confirm an earlier suggestion that Ne-E might have originated outside the solar system (Black, 1972b; Clayton, 1975; Arnould and Nørgaard, 1978). Other explanations which had been considered for Ne-E include (i) *in situ* ternary fission of a superheavy element (Black, 1972b), (ii) irradiation of the primitive nebula by negative muons (Jeffery and Hagan, 1969) or by neutrons, protons, etc. (Anders *et al.*, 1970; Heymann and Dziczkaniec, 1976; Audouze *et al.*, 1976; Clayton *et al.*, 1977), (iii) *in situ* decay of extinct  $^{22}\text{Na}$  (Black and Pepin, 1969; Clayton, 1975), and (iv) mass fractionation (Kuroda and Manuel, 1970). The latter mechanism has been frequently considered for the less severe enrichments of heavy neon isotopes in meteorites during the three decades since this was first proposed by Suess (1949).

In the following sections, we will show that mass fractionation and spallation can account for differences in the isotopic composition of neon in air, in the solar wind, and in the supposedly isotopically distinct components of trapped meteoritic neon. Readers should be cautioned of a point brought to our attention by an anonymous reviewer: Although the experimental data are permissive of our interpretation, the Ne measurements do not disprove the alternative hypothesis of nucleogenetic anomalies in neon.

## CORRELATED FRACTIONATION EFFECTS

As mentioned in the introduction, mass fractionation has often been considered as a possible explanation for variations in the isotopic composition of neon in the solar system (Suess, 1949; Zähringer and Gentner, 1960; Stauffer, 1961; Manuel, 1967; Zähringer, 1968; Hohenberg *et al.*, 1970; Kuroda and Manuel, 1970; Arrhenius and Alfvén, 1971; Srinivasan and Manuel, 1971; Srinivasan, 1973). A common basis for the rejection of this mechanism has been the absence of correlated variations in the isotopes of argon. Thus, Anders *et al.* (1970) noted that the correlated isotopic variations of helium and neon over the range  $8 \lesssim ^{20}\text{Ne}/^{22}\text{Ne} \lesssim 13$  are consistent with mass dependent fractionation but there is “no detectable variation in  $^{36}\text{Ar}/^{38}\text{Ar}$ .” Black and Pepin (1969) pointed out that fractionation effects in neon are constrained by “the fact that large isotopic variations do not occur in trapped meteoritic argon,” and Zähringer (1968) noted that “the constant  $^{36}\text{Ar}/^{38}\text{Ar}$  ratios cannot be accounted for by this mechanism alone.”

A basic assumption of the above arguments against fractionation—formation of the solar system from a homogeneous nebula of solar composition (Larimer and Anders, 1967; Grossman and Larimer, 1974)—has been discredited by recent

evidence that meteorites condensed from material that was chemically, as well as isotopically, heterogeneous (Manuel and Sabu, 1975, 1977; Sabu and Manuel, 1976, 1980; Boynton, 1978a,b; El Gorse *et al.*, 1978; Manuel, 1979, 1980; Grossman, 1980). The chemical heterogeneities that existed prior to condensation were, in many cases, obscured by the chemical separation of elements into different minerals during condensation. However, a few elements with very similar chemical properties have retained a record of the chemical heterogeneities that existed prior to the redistribution of elements during condensation.

The five noble gases have similar physical properties and are essentially inert to chemical reactions. It is not surprising then that these elements provided the first clear evidence of chemically and isotopically distinct regions in the primitive nebula. The correlations which were first noted between elemental and isotopic ratios of the five noble gases in Allende (Manuel and Sabu, 1975, 1977; Sabu and Manuel, 1976) have since been shown to be a characteristic feature of these elements in meteorites. Abundances of helium and neon in trapped meteoritic noble gases correlate with isotopic ratios of the three heavy noble gases—argon, krypton, and xenon—in the manner that would be expected if we assume that the primitive nebula contained *basically* two very different types of noble gases, X and Y, representing debris from the outer and inner layers, respectively, of a supernova (Manuel, 1978, 1979, 1980; Sabu and Manuel, 1980). These two major categories of gases may be subdivided further as elemental and isotopic characteristics of other components are identified, but it has already been noted (Manuel, 1980) that the large-scale chemical and isotopic heterogeneities manifest in the noble gases provide stringent criteria for models suggested to explain the occurrence of nucleogenetic anomalies in meteorites (Clayton *et al.*, 1973; Clayton, 1975; Manuel and Sabu, 1975; Cameron and Truran, 1977; Lattimer *et al.*, 1978; Manuel *et al.*, 1972).

Differences in the elemental and isotopic compositions of these two types of noble gases led us to suggest that the solar system condensed directly from the debris of a single supernova, with the sun forming on the SN core remnant and the planets condensing from an equatorial disk of material that retained a spatial distribution of heterogeneities within the supernova (Manuel and Sabu, 1975, 1977; Sabu and Manuel, 1976, 1980; Manuel, 1978).

According to this view, type-Y noble gases consist only of the three heavy members—argon, krypton, and xenon—because these were trapped from elements made in the stellar interior where fusion reactions had consumed all low-Z elements. The isotopic compositions of Ar-Y, Kr-Y, and Xe-Y are called “normal” or “planetary” because these are the dominant form of heavy noble gases in the inner planets and in bulk meteorites.

Type-X noble gases represent material from the outer layers of the supernova, and the isotopic compositions of Ar-X, Kr-X, and Xe-X appear “anomalous” to the inhabitants of an inner planet. But type-X gases also contain all of the helium and neon trapped in meteorites, and these are isotopically “normal” or “planetary” because there were no low-Z elements in type-Y gases, i.e., all of the helium and neon were derived from the outer supernova layers. Previous use of the term “planetary” noble gases (Pepin and Signer, 1965 and references therein) thus

referred to helium and neon of type-X gases mixed with argon, krypton, and xenon of type-Y.

Thus, all of the planetary neon in meteorites was trapped with Ar-X, but most of the argon in bulk meteorites is Ar-Y. Therefore, correlated fractionation effects in “planetary” neon and argon are not expected, i.e., the absence of fractionation effects in argon does not exclude the possibility of fractionation effects in neon. Correlated fractionation of neon, Ar-X, Kr-X and Xe-X are expected, and the presently available data do not rule out this possibility. Correlated fractionation effects in the isotopes of helium and neon have been seen, but elemental ratios of these two gases do not vary in the manner expected of diffusive fractionation (Anders *et al.*, 1970; Srinivasan and Manuel, 1971). This suggests the possibility of isotopic fractionation prior to trapping. Severely fractionated helium is, however, difficult to identify because of interference from spallogenic and radiogenic components in an element with only two isotopes.

Finally, it should be acknowledged that the suggested formation of the solar system from the debris of a single supernova (Manuel and Sabu, 1975) has not been widely endorsed (Lewis *et al.*, 1977a; Podosek, 1978; Clayton, 1979.) There is, however, supporting evidence for our views in the chemical and isotopic heterogeneities of other elements. Isotopically anomalous tellurium (Ballad *et al.*, 1979) and uranium (Tatsumoto and Shimamura, 1980) have been reported in the residue fraction of Allende that contains anomalous xenon, krypton and argon, and Grossman (1980) has recently acknowledged that Allende’s inclusions provide evidence for “solar nebular regions of different chemical and isotopic composition.”

## EXCESS SPALLOGENIC NEON

Srinivasan *et al.* (1977) noted that spallogenic  $^{21}\text{Ne}$  appeared to be enriched by a factor of 15 in acid-etched residues of Murchison relative to the bulk meteorite. They reported that recoil of spallation products from other phases could not explain the excess  $^{21}\text{Ne}$  and suggested the possibility of irradiation prior to compaction. Alaerts *et al.* (1980) have argued that the excess  $^{21}\text{Ne}$  is instead evidence of yet another trapped neon, Ne-A2. However, the differences they report in the isotopic compositions of Ne-A1 and Ne-A2 are those expected from the presence of spallogenic neon in the latter.

In this report we will assume that isotopic variations—in the direction expected from fractionation and/or spallation—are evidence of these well-known processes, rather than evidence of isotopically distinct neon components. Our interpretation of the neon data thus agrees with the earlier suggestion that certain phases of meteorites contained spallogenic  $^{21}\text{Ne}$  prior to compaction (Srinivasan *et al.*, 1977).

If the mineral grains of residues were themselves preirradiated, then the chemical compositions of the grains could be used to estimate the amounts of other spallation products, e.g.,  $^3\text{He}$  and  $^{38}\text{Ar}$ , which should accompany the spallogenic

$^{21}\text{Ne}$ . There is evidence, however, that the spallogenic neon was not produced within the current carrier grains. Lewis *et al.* (1977b) reported excess spallogenic neon in a carbon-rich ( $\text{C} \geq 97\%$ ) residue of Allende, where there is a paucity of target elements for the production of neon, and Alaerts *et al.* (1980) observed excess spallogenic  $^{21}\text{Ne}$  in the neon-rich,  $1000^\circ\text{C}$  fractions of Murchison residues, 2C10f and 2C10f140. These observations suggest that the spallation reactions may have occurred in other sites and the neon products may have been trapped, together with planetary neon, in the current carrier grains.

There are two plausible sites for the production of excess spallogenic neon by an early irradiation—in precursor grains enriched in elements with high spallation yields for neon, such as Mg, Al, or Si, or in regions of chemically zoned supernova debris that were enriched in these elements. The latter possibility is particularly attractive from our point of view, since a large flux of cosmic-rays should have occurred near the supernova site (Kuroda, 1979). In fact, spallation and neutron capture reactions in the post-explosion stage may have produced the bulk of the solar system's supply of deuterium, lithium, beryllium, and boron in a manner similar to that suggested earlier by Fowler *et al.* (1962). Clearly, a large fraction of the spallogenic neon could have been produced at that time.

An early irradiation would also be expected to produce spallogenic  $^3\text{He}$  and  $^{38}\text{Ar}$ , although the latter would not necessarily have been produced at the same site as the  $^{21}\text{Ne}$ . If the irradiation occurred prior to trapping of spallogenic neon in the present carrier grains, then the amounts of spallogenic  $^3\text{He}$ ,  $^{21}\text{Ne}$ , and  $^{38}\text{Ar}$  in these grains might depend on the production rates of  $^3\text{He}$ ,  $^{21}\text{Ne}$ , and  $^{38}\text{Ar}$  in the irradiated material, on the relative abundances of helium, neon, and argon in the gas which diluted these spallation products, and on the relative trapping efficiencies of these three gases in the carrier grains.

Thus, an early irradiation would certainly produce other spallation products in addition to  $^{21}\text{Ne}$ , but a particular mineral would not necessarily contain these in the proportions that would be generated by *in situ* spallation reactions. It should be noted that excess  $^{21}\text{Ne}$  is observed mixed with planetary neon (Ne-A) when relatively large amounts of neon are analyzed (Srinivasan *et al.*, 1977; Alaerts *et al.*, 1980). Unlike the evidence to be discussed below for monoisotopic  $^{22}\text{Ne}$ , we differ in the interpretation of, but do not doubt the existence of, excess  $^{21}\text{Ne}$  in certain meteorite minerals.

## MASS FRACTIONATED NEON

Values of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio in air, in the solar wind, and in meteoritic components Ne-A, -B, -C, -D, -A1, -A2 and -O vary by about a factor of two, from  $^{20}\text{Ne}/^{22}\text{Ne} = 14.5 \pm 1.0$  for Ne-D (Black, 1972a) to  $^{20}\text{Ne}/^{22}\text{Ne} \geq 6.4$  for Ne-O (Eberhardt, 1978). Studies of solar-wind-implanted neon in lunar soils have shown that mass fractionation may be responsible for isotopic variations over a similar range,  $8.5 \leq ^{20}\text{Ne}/^{22}\text{Ne} \leq 14$  (Hohenberg *et al.*, 1970; Srinivasan *et al.*, 1972; Megrue and Steinbrunn, 1972; Srinivasan, 1973). The results of these studies



appear to confirm suggestions that mass fractionation may have played an important role in generating variations in the isotopic composition of neon in other parts of the solar system (Suess, 1949; Zähringer and Gentner, 1960; Stauffer, 1961; Manuel, 1967; Zähringer, 1968; Kuroda and Manuel, 1970; Srinivasan and Manuel, 1971; Arrhenius and Alfvén, 1971).

Arrhenius and Alfvén (1971) list seven fractionation mechanisms that might have altered the isotopic compositions of elements in the primitive nebula. Variations in the isotopic composition of neon in lunar soils may represent an example of their multiple desorption process in which gases impinge, diffuse and desorb from solid surfaces.

Several of the fractionation mechanisms suggested by Arrhenius and Alfvén (1971) involve mass separation in an ionized gas, and Frick *et al.* (1979) have recently observed isotopic fractionation that enriched the heavy isotopes of krypton and xenon when these gases were trapped in the kerogen that condensed from an electrically discharged mixture of CH<sub>4</sub>, N<sub>2</sub> and noble gases. Possible fractionation effects in the light noble gases were obscured by meteoritic contamination. The role played by the electrical discharge in separating the isotopes of these heavy noble gases by more than 1% per amu is not understood, but the results of this study demonstrate that fractionation effects in the noble gases may not be restricted to simple diffusive loss.

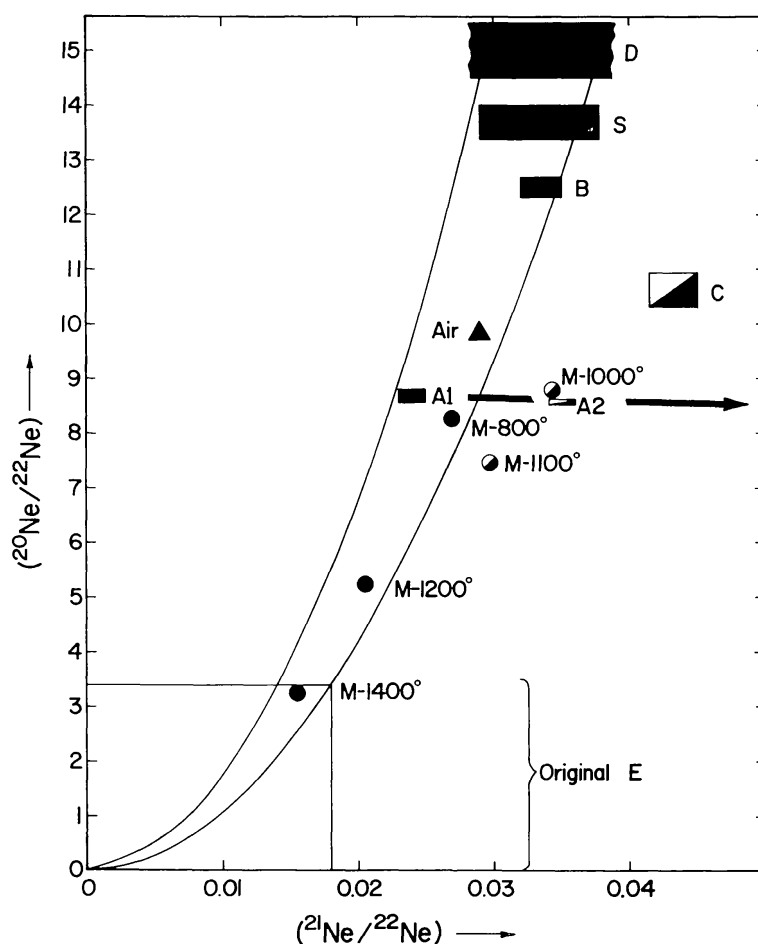
The relative contributions of the various fractionation mechanisms that operated in the early solar system, or for that matter in the laboratory experiments of Frick *et al.* (1979), are not well understood. In spite of this, mass-dependent fractionation of any element, X, with three or more isotopes of mass a, b, c . . . , will produce an anomaly pattern which can be quantitatively represented by the equation (Hohenberg *et al.*, 1970; Srinivasan, 1973),

$$\partial \ln(^aX/^cX) / \partial \ln(^bX/^cX) = (a-c)/(b-c). \quad (1)$$

Isotopically fractionated noble gases in the electric discharge kerogen (Frick *et al.*, 1979) and in lunar soils (Hohenberg *et al.*, 1970; Srinivasan *et al.*, 1972; Megrue and Steinbrunn, 1972; Srinivasan, 1973) are in agreement with Eq. (1). In the case of neon, each stage of fractionation is expected to produce a fractional change in the <sup>20</sup>Ne/<sup>22</sup>Ne ratio that is twice that in the <sup>21</sup>Ne/<sup>22</sup>Ne ratio, so that

$$\partial \ln(^{21}\text{Ne}/^{22}\text{Ne}) / \partial \ln(^{20}\text{Ne}/^{22}\text{Ne}) = 0.5. \quad (2)$$

Equation (2) predicts an almost linear correlation of <sup>20</sup>Ne/<sup>22</sup>Ne vs. <sup>21</sup>Ne/<sup>22</sup>Ne for small degrees of fractionation, but the correlation will show significant departures from linearity when fractionation is severe. This is illustrated in Fig. 1, where the area bounded by the curved lines represents the isotopic compositions predicted by Eq. (2) when planetary Ne-A (Black and Pepin, 1969) is mass fractionated. In calculating the isotopic compositions expected for fractionated Ne-A, we have conservatively assumed that fractional errors will remain constant. In the next section we will show, however, that there are formidable experimental difficulties which greatly increase the experimental uncertainties when severely mass fractionated neon is analyzed.



**Fig. 1.** A comparison of the isotopic compositions of neon expected from fractionation and spallation with those in air, in the solar wind (S), in various temperature fractions of the Murchison 1C10 residues, and in the postulated components of trapped meteoritic neon, Ne-A1, -A2, -B, -C, -D, and -E. Fractionation of “planetary” neon, Ne-A, generates isotopic compositions within the area bounded by the two curved lines, and the original limits on the isotopic composition of Ne-E are represented by the rectangle in the lower left corner. Spallogenic neon lies far to the right, in the direction indicated by the arrow from Ne-A1. The filled symbols represent neon that can be related to planetary Ne-A by simple mass-dependent fractionation, and the half-filled symbols represent neon that may be a mixture of fractionated and spallogenic neon. Ne-O, which lies far to the right at  $^{20}\text{Ne}/^{22}\text{Ne} > 6.4$  and  $^{21}\text{Ne}/^{22}\text{Ne} > 0.12$ , can also be understood as a mixture of spallogenic and fractionated neon.

It can be seen from Fig. 1 that the isotopic compositions of several trapped neon components—Ne-A, Ne-A1, Ne-D, Ne-B, and the original upper limits on Ne-E (Black and Pepin, 1969; Black, 1972b)—are mathematically related to the isotopic composition of neon in air (Nier, 1950) and in the solar wind (Geiss *et al.*, 1972) in the manner expected from mass-dependent fractionation. Fractionation also accounts for the isotopic composition of neon in the 800°C, 1200°C, and 1400°C fractions of the Murchison 1C10 residues (Srinivasan and Anders, 1978). These fractions are shown with filled symbols to indicate that they can be explained by fractionation and thus can, by widely accepted criteria (e.g., Clayton

and Mayeda, 1977; Lee *et al.*, 1978), provide no evidence of isotopically distinct neon components.

A reviewer has asked that we cite an independent example of a mass-dependent isotopic fractionation of this size. Aside from such obvious examples as the diffusive separation of molecular  $^{235}\text{UF}_6$  from  $^{238}\text{UF}_6$  in the early 1940's, electromagnetic separations of ions in mass spectrometers, etc., relatively large differences in the isotopic compositions of planetary and solar helium and neon fit the pattern expected from mass-dependent fractionation (Srinivasan and Manuel, 1971). It seems reasonable to consider the possibility that the fractionation which altered the  $^3\text{He}/^4\text{He}$  ratio in these two major gas reservoirs by a factor of three could, by repetitive stages, generate minor gas fractions with much larger fractionation effects.

It is, of course, impossible to disprove the existence of isotopically distinct neon components, even if they lie within the area bounded by the curved lines in Fig. 1. It is worthy of note, however, that other studies have not confirmed all of the trapped neon components reported by Black and Pepin (1969) and Black (1972b). Thus, Smith *et al.* (1978) report variations in the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio of trapped neon in three carbonaceous chondrites—Murchison, Mokoia, and Cold Bokkeveld—ranging from 6.9 to 13.7 with “no clear evidence of preferred intermediate compositions such as Ne-A.”

In Fig. 1, the neon in the 1000°C and 1100°C fractions of the 1C10 residues and that in three other trapped neon components, Ne-A2, Ne-C, and Ne-O, contain excess  $^{21}\text{Ne}$  from spallation. The dark arrow from Ne-A1 extrapolates to spallogenic neon at  $^{20}\text{Ne}/^{22}\text{Ne} \approx ^{21}\text{Ne}/^{22}\text{Ne} \approx 1$ . The addition of spallogenic neon to fractionated Ne-A can account for the neon components represented by the half-filled symbols. Spallation plus fractionation can also account for the isotopic composition of the other trapped component, Ne-O (not shown), which lies far to the right of Fig. 1 at  $^{20}\text{Ne}/^{22}\text{Ne} > 6.4$ ,  $^{21}\text{Ne}/^{22}\text{Ne} > 0.12$  (Eberhardt, 1978).

Thus, differences in the isotopic compositions of neon in air, in the solar wind, and in the meteoritic components identified as Ne-A, -B, -C, -D, -A1, -A2 and -O are those expected from fractionation or fractionation plus spallation. Clearly these provide no evidence for distinct nucleogenetic components of neon, and we will now turn our attention to the extremely  $^{22}\text{Ne}$ -rich forms of neon.

In using Eq. (2) to look for evidence of nucleogenetic anomalies of neon in data showing extreme enrichments of  $^{22}\text{Ne}$ , it is advantageous to examine the isotopic ratios in the manner shown in Fig. 2. Srinivasan and Anders (1978) reported that noble gases released at high temperatures from the Murchison 1C10 residue were enriched in Ne-E and in s-products of krypton and xenon. The neon released from the residue at 800°C is similar to Ne-A in composition and its isotopic ratios display a linear relationship in Fig. 2. The addition of spallogenic neon would cause the largest fractional increase in the abundance of  $^{21}\text{Ne}$ , producing an anomaly pattern with the general features indicated by the upper dashed line in Fig. 2. However, the addition of monoisotopic  $^{22}\text{Ne}$ , which was recently suggested as the composition of Ne-E (Jungck and Eberhardt, 1979; Meier *et al.*,



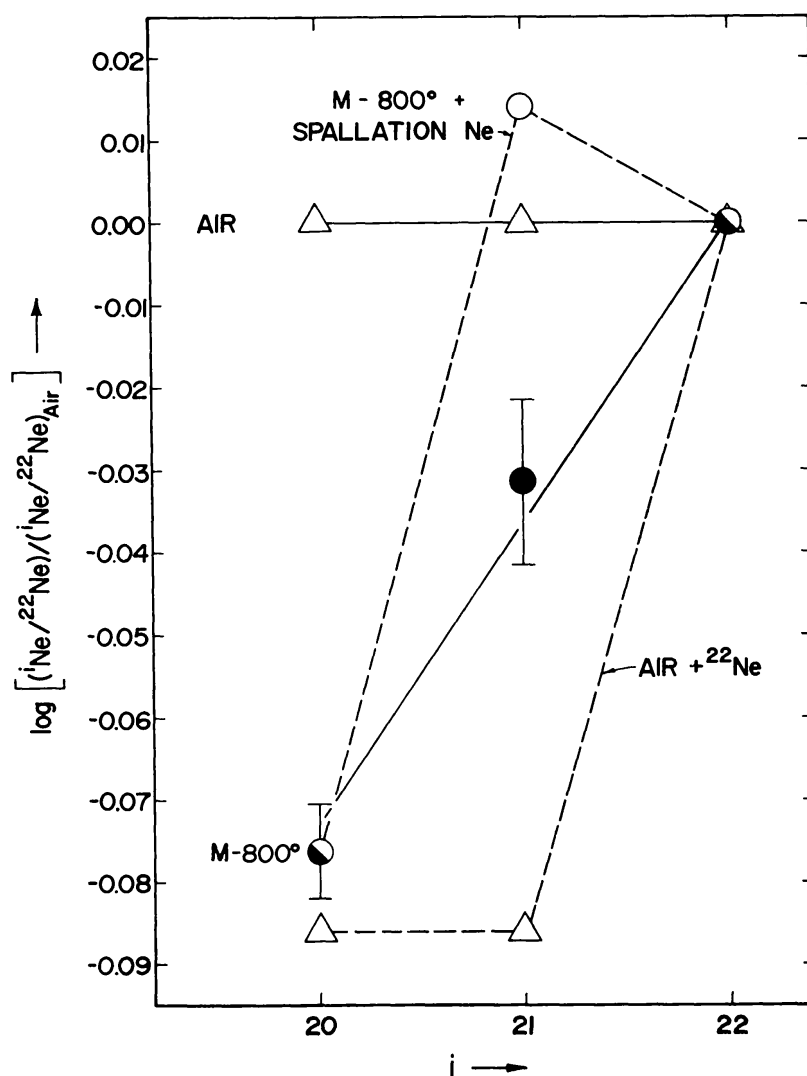


Fig. 2. A comparison of the isotopic composition of neon in air with that expected by fractionation (the solid diagonal line passing through neon in the 800°C fraction of the Murchison 1C10 residue), by the addition of spallogenic neon to that in the 800°C fraction of Murchison 1C10 (upper dashed line), and by the addition of monoisotopic <sup>22</sup>Ne to atmospheric neon (lower dashed line).

1980), would produce the opposite anomaly pattern indicated by the lower dashed line in Fig. 2. Although not shown in Fig. 2, the addition of <sup>20</sup>Ne would also produce an anomaly pattern in which <sup>21</sup>Ne would appear to be depleted relative to a fractionation line defined by the abundances of <sup>20</sup>Ne and <sup>22</sup>Ne. When Eq. (2) is used to examine the isotopic composition of meteoritic neon in the manner illustrated in Fig. 2, it allows us to distinguish anomalies that may have been produced by spallation and fractionation from those that might indicate the presence of distinct nucleogenetic components.

The report that Ne-E accompanies s-products of xenon and krypton in the 1C10 residue of Murchison (Srinivasan and Anders, 1978) played a major role in reviving interest in the possibility of nucleogenetic anomalies in neon (Black,

1972b). To demonstrate that the 1C10 residue contains no evidence of neon with a nucleogenetic history which differs from that found in the rest of the solar system, the isotopic composition of neon in the three spallation-free fractions, 800°C, 1200°C, and 1400°C, are shown in Fig. 3, together with the isotopic compositions of neon in air and in the solar wind. If stellar nuclear reactions produced the neon in these three fractions, then they did a remarkably good job of mimicking the isotopic ratios that Eq. (2) predicts for mass fractionation.

The isotopic compositions of neon in the 1000°C and 1200°C fractions are shown in Fig. 4. The anomaly pattern is that expected from fractionation and spallation, as illustrated earlier by the upper dashed line of Fig. 2. Neon in the 1600°C fraction shows a similar anomaly pattern, but it has not been plotted because of large experimental uncertainties.

The presence of monoisotopic  $^{22}\text{Ne}$  in the 1C10 residue of Murchison would be indicated by an anomaly pattern such as that shown by the lower dashed line in Fig. 2. The data shown in Figs. 3 and 4 clearly demonstrate that fractionation

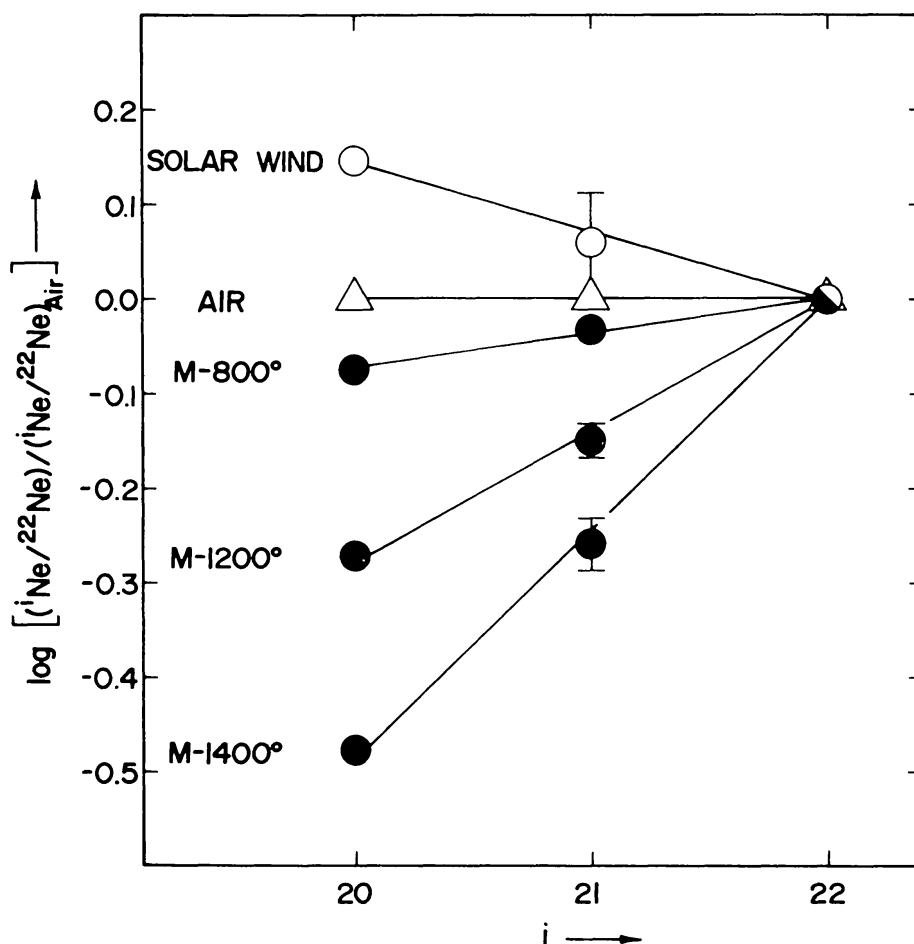


Fig. 3. Neon in the solar wind and in the 800°C, 1200°C, and 1400°C fractions of the Murchison 1C10 residues displays the isotopic compositions expected by fractionation of atmospheric neon. The isotopic ratios do not match the pattern expected from the addition of monoisotopic  $^{22}\text{Ne}$  (see lower dashed line in Fig. 2).

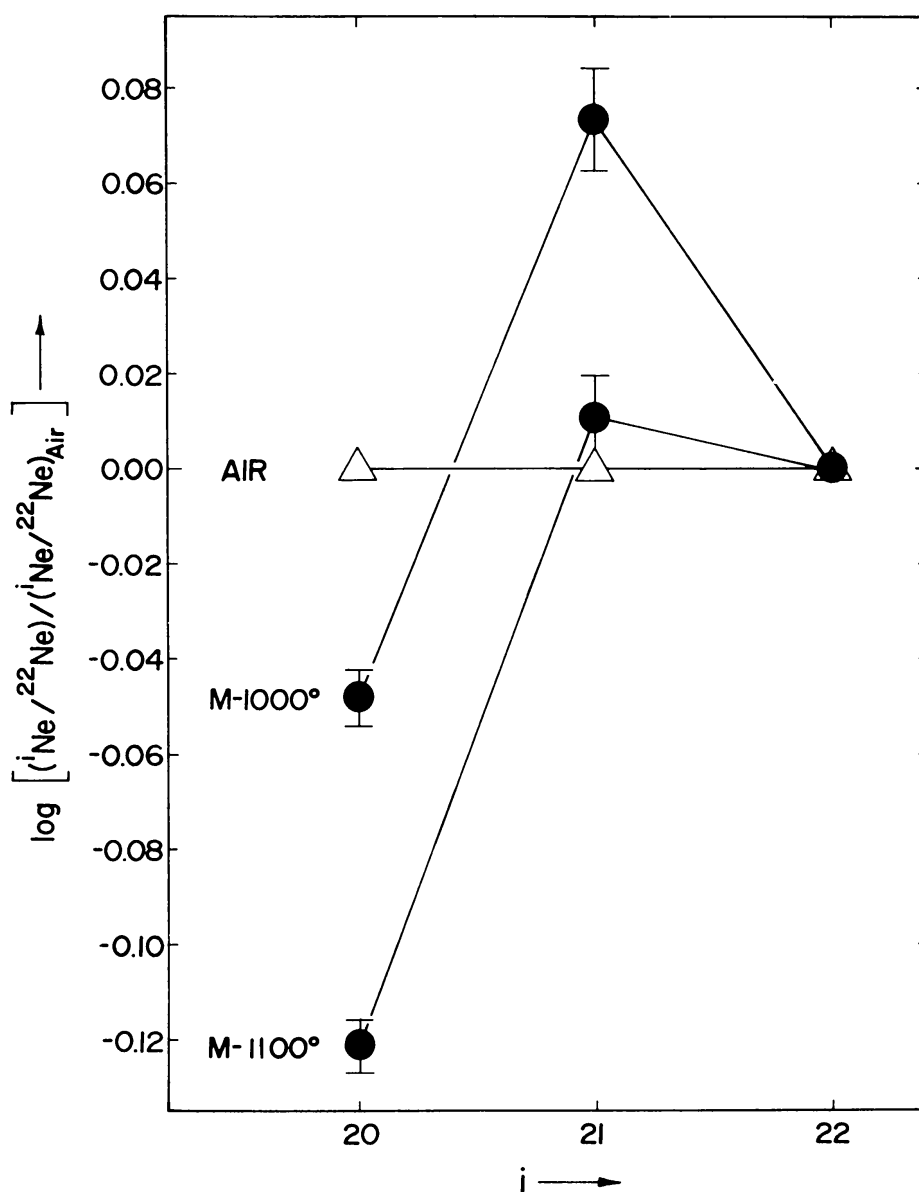


Fig. 4. Neon in the 1000°C and 1100°C fractions of the Murchison 1C10 residues displays the isotopic anomaly pattern expected by spallation and fractionation, as illustrated by the upper dashed line in Fig. 2. There is no evidence for the presence of monoisotopic  $^{22}\text{Ne}$  here (see lower dashed line of Fig. 2).

and/or fractionation plus spallation can account for the neon isotopes in 1C10. The neon in the 1C10 residue therefore provides no evidence of isotopic anomalies which might require a separate nucleogenetic origin.

It could be argued, however, that the presence of monoisotopic  $^{22}\text{Ne}$  has been obscured by some fortuitous correlation of cosmogenic  $^{21}\text{Ne}$  with values of the  $^{22}\text{Ne}/^{20}\text{Ne}$  ratio. If low values of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio in the 1C10 residues of Murchison are assigned to the presence of monoisotopic  $^{22}\text{Ne}$ , and if the fractionation trends in the neon isotopes released at 800°C, 1200°C, and 1400°C are attributed to the chance presence of spallogenic neon which “masks the unam-

biguous evidence for mono-isotopic  $^{22}\text{Ne}$  addition," as suggested by a reviewer, then spallogenic neon is being released at all extraction temperatures and amounts to  $2.3 \times 10^{-8}$  cc STP/g. On the other hand, if low values of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio are assigned to fractionation and spallation, then spallogenic  $^{21}\text{Ne}$  is only being released in the  $1000^\circ\text{C}$  and  $1100^\circ\text{C}$  fractions and amounts to  $1.4 \times 10^{-8}$  cc STP/g. Because of the possible presence of spallogenic  $^{21}\text{Ne}$  from an early irradiation prior to compaction, the amounts of excess  $^{21}\text{Ne}$  indicated by these calculations do not rule out either possibility. The reader will have to use other criteria, such as the likelihood of events assumed in these two calculations, to decide their merits.

We have examined all available data for evidence of a neon component with less  $^{21}\text{Ne}$  than predicted by Eq. (2), i.e., we sought evidence for an anomaly pattern like that represented by the lower dashed line in Fig. 2. Such a finding would discredit a mass fractionation origin for Ne-E. However, the abundance of  $^{21}\text{Ne}$  is, within the  $1\sigma$  error limits, always equal to or greater than that predicted by Eq. (2), except in one measurement.

In the  $800^\circ\text{C}$  fraction of neon released from the 2C10f residues of Murchison (Alaerts *et al.*, 1980), there appears to be a deficiency of  $^{21}\text{Ne}$ . Although neon in the  $800^\circ\text{C}$  fraction of the 2C10f140 residues do not confirm this deficiency of  $^{21}\text{Ne}$ , Alaerts *et al.* (1980) have averaged the isotopic compositions of neon in the  $800^\circ\text{C}$  fractions of 2C10f and 2C10f140 to obtain the component which they call Ne-A1. They treat the statistics as if the analyses of 2C10f and 2C10f140 represent independent measurements of the same neon, and thus obtain small statistical errors on the isotopic composition of Ne-A1 and an apparent deficiency of  $^{21}\text{Ne}$  in this calculated component. Alaerts *et al.* (1980) include in their Table 3 the isotopic compositions of neon in the  $800^\circ\text{C}$  fractions of Murchison residues 1C5 and 1C10, but the neon in these is not deficient in  $^{21}\text{Ne}$  and was not used in calculating the isotopic composition of Ne-A1.

The one measurement showing less  $^{21}\text{Ne}$  than predicted by Eq. (2) provides little evidence, however, for the presence of monoisotopic  $^{22}\text{Ne}$ . In the  $800^\circ\text{C}$  fraction of 2C10f residues, the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio is high, 8.73, and ironically Alaerts *et al.* (1980) calculate a negative amount of Ne-E,  $(-3.1 \pm 1.9) \times 10^{-8}$  cc STP/g, in this neon fraction.

If Ne-E were monoisotopic  $^{22}\text{Ne}$ , or even if it consisted of the extremely  $^{22}\text{Ne}$ -rich gas reported in D1\* (Jungck and Eberhardt, 1979; Meier *et al.*, 1980), then mixtures of Ne-E with planetary neon would produce an anomaly pattern similar to that shown by the lower dashed line in Fig. 2. The absence of such an anomaly pattern in the data shown in Figs. 3 and 4 or in any other reports on Ne-E, except for the report of negative Ne-E noted above, suggests an inconsistency between all earlier neon analyses and the recent report on neon in the D1\* mineral separates of Orgueil (Jungck and Eberhardt, 1979; Meier *et al.*, 1980). It therefore seems clear that the addition of monoisotopic  $^{22}\text{Ne}$  is not required for differences in the isotopic composition of meteoritic neon over the range  $3 < ^{20}\text{Ne}/^{22}\text{Ne} < 15$ . We will therefore focus our attention on recent evidence for extreme forms of Ne-E with  $^{20}\text{Ne}/^{22}\text{Ne} < 3$ .

## EXTREMES OF NEON-E

### Detection limits

Although there has been considerable speculation in recent years about an extra-solar origin of  $^{22}\text{Ne}$  in the Ne-E component (Niederer and Eberhardt, 1977; Arnould and Nørgaard, 1978; Srinivasan and Anders, 1978; Eberhardt, 1978; Clayton, 1979; Eberhardt *et al.*, 1979a,b; Lewis *et al.*, 1979; Jungck and Eberhardt, 1979; Alaerts *et al.*, 1979a,b, 1980; Meier *et al.*, 1980), it should be stressed that there have been very few experimental details published for those analyses in which  $^{22}\text{Ne}$  is enriched by more than a factor of 10. Many mass spectrometers contain blanks of atmospheric neon with  $^{20}\text{Ne}/^{22}\text{Ne} \sim 9.8$ , and the blank contribution at mass 20 may be appreciable if the value of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio is low, particularly when the sample size and its neon content are extremely low. This problem is treated quantitatively, within the limits of available information, in the Appendix.

The calculations presented there are not intended to cast doubt on the experimental work at Chicago and Bern, but rather to demonstrate the experimental problems encountered in measuring low  $^{20}\text{Ne}/^{22}\text{Ne}$  ratios in very small samples. Jungck and Eberhardt (1979) and Meier *et al.* (1980) stress that their data on D1\* are preliminary, but this has done little to silence the research news reports on the discovery of extra-solar  $^{22}\text{Ne}$  in meteorites (Fallick and Pillinger, 1979; Hutchison, 1980). Variations of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio by two orders of magnitude in the gas released at different temperatures from the 100- $\mu\text{g}$  sample of D1\* suggest the possibility of instrumental or other fractionation effects that are peculiar to the analysis of such minute amounts of gas. There have been no reports on calibration of the mass spectrometer and the associated clean-up system with such minute amounts of neon of known composition. In view of these experimental problems, we suggest that there is at present little or no experimental evidence for a trapped neon component in meteorites with  $^{20}\text{Ne}/^{22}\text{Ne} < 1$ .

Before turning our attention to other aspects of Ne-E, it should be noted that the isotopic composition of neon reported in D1\* separates of Orgueil (Jungck and Eberhardt, 1979; Meier *et al.*, 1980) is so highly enriched in  $^{22}\text{Ne}$  that mixtures of this neon with atmospheric or planetary neon would produce an anomaly pattern similar to that shown by the lower dashed line in Fig. 2. The neon anomaly pattern in other reports of Ne-E (e.g., the putative Ne-E of Murchison 1C10 residues shown in Figs. 3 and 4) cannot be explained as a simple mixture of planetary Ne-A with the Ne-E that has been reported in D1\*.

### Trends in isotopic limits on Ne-E

Except for the report on the Murchison 2C10 residues (Alaerts *et al.*, 1980) numerical values have not been reported for isotopic ratios in individual temperature fractions of the most Ne-E rich mineral separates of meteorites (those with



$^{20}\text{Ne}/^{22}\text{Ne} < 1$ ). However, upper limits on values of the  $^{20}\text{Ne}/^{22}\text{Ne}$  and  $^{21}\text{Ne}/^{22}\text{Ne}$  ratios in Ne-E are usually given, and the upper limits on both isotopic ratios have steadily declined in recent years. Thus, the upper limit of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio has declined from 3.4 (Black and Pepin, 1969; Black, 1972b) to the present value of 0.01 (Jungck and Eberhardt, 1979; Meier *et al.*, 1980).

Upper limits on the  $^{21}\text{Ne}/^{22}\text{Ne}$  ratio have also declined during recent years, from a value of 0.018 (Black, 1972b) to 0.001 (Jungck and Eberhardt, 1979; Meier *et al.*, 1980). Although the low values observed for both isotopic ratios have been interpreted as evidence for the presence of monoisotopic  $^{22}\text{Ne}$  (Jungck and Eberhardt, 1979; Alaerts *et al.*, 1980; Meier *et al.*, 1980), it can be shown that the upper limits on these two isotopic ratios in Ne-E are remarkably close to the values expected from simple mass-dependent fractionation.

In the absence of numerical values for the isotopic ratios of neon in the most Ne-E rich mineral separates of meteorites, we will consider the upper limits on the isotopic ratios of Ne-E in the G5a (Eberhardt *et al.*, 1979a,b) and in the D1\* (Jungck and Eberhardt, 1979; Meier *et al.*, 1980) mineral separates of Orgueil. In Fig. 5, these upper limits on the isotopic composition of Ne-E are compared with the isotopic composition of atmospheric neon and with earlier upper limits

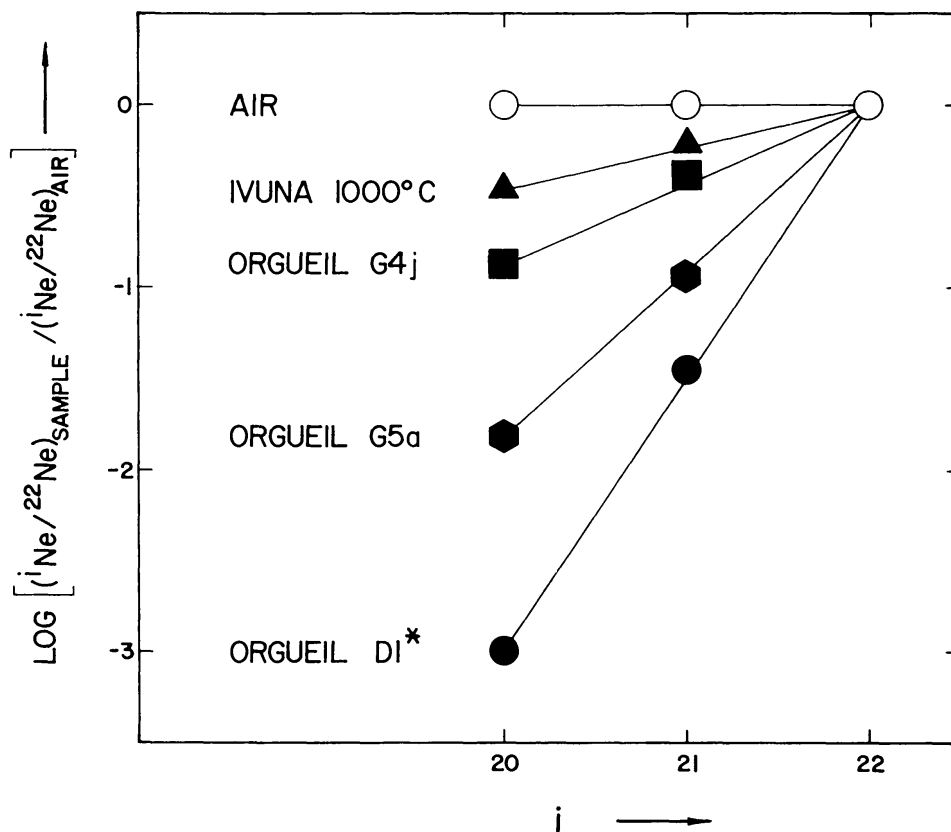


Fig. 5. Upper limits on the values of  $^{20}\text{Ne}/^{22}\text{Ne}$  and  $^{21}\text{Ne}/^{22}\text{Ne}$  ratios in Ne-E over the past several years vary in exactly the manner expected from simple mass-dependent fractionation of atmospheric neon. In no case do the limits on Ne-E provide evidence for the anomaly pattern expected if Ne-E were monoisotopic  $^{22}\text{Ne}$ .

on the isotopic composition of Ne-E from the 1000°C fraction of the bulk Ivuna carbonaceous chondrite (Black and Pepin, 1969; Black, 1972b) and from the G4j mineral separates of Orgueil (Eberhardt, 1978).

It can be seen in Fig. 5 that the upper limits reported for the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio of Ne-E covary with the upper limits reported for the  $^{21}\text{Ne}/^{22}\text{Ne}$  ratio of Ne-E in the manner expected from mass fractionation. Since the values shown there are only upper limits, they do not provide conclusive evidence against the presence of monoisotopic  $^{22}\text{Ne}$ . However, if pure  $^{22}\text{Ne}$  is trapped in meteorites, then it must be a remarkable coincidence that all the available numerical values on isotopic ratios of Ne-E and the upper limits estimated for the isotopic compositions of Ne-E in all other reports on this strange neon (except in the one report of negative Ne-E), always show the anomaly pattern expected from fractionation and/or fractionation plus spallation (see Figs. 1-5).

In view of the low probability of this remarkable coincidence, possible analytical problems discussed earlier, and the absence of any experimental data showing the anomaly pattern expected from the addition of monoisotopic  $^{22}\text{Ne}$  (see lower dashed line of Fig. 2), we conclude that there is at present no evidence of monoisotopic  $^{22}\text{Ne}$  or other alien nucleogenetic components of neon—unless addition of the latter produces the fractionation/spallation-type anomaly pattern.

## CONCLUSIONS

Neon in meteorites contains an important record of events in the early solar system, including evidence that (i) planetary neon was initially associated with planetary helium and with isotopically anomalous argon, krypton and xenon but not with the argon, krypton and xenon of “normal” or “planetary” composition (Sabu and Manuel, 1980); (ii) mass fractionation has enriched  $^{22}\text{Ne}$  relative to  $^{20}\text{Ne}$  by as much as an order of magnitude (Fig. 3), and perhaps by as much as three orders of magnitude (Fig. 5); and (iii) some phases contain spallogenic  $^{21}\text{Ne}$  that was produced prior to compaction of the meteorite (Fig. 1, Fig. 4, and Srinivasan *et al.*, 1977).

When the available data are examined for the isotopic anomaly patterns expected from fractionation, spallation, and/or the addition of monoisotopic  $^{22}\text{Ne}$  (Fig. 2), no evidence of the latter is observed. Experimental problems are noted in the measurement of extremely low  $^{20}\text{Ne}/^{22}\text{Ne}$  ratios when the blanks are atmospheric in composition. Further, the isotopic compositions observed in most studies on Ne-E cannot be explained as mixtures of planetary Ne-A with the extremely  $^{22}\text{Ne}$ -rich component that has been tentatively identified in the D1\* mineral separate of Orgueil.

**Acknowledgments**—We gratefully acknowledge helpful comments by the following members of the U.S. Geological Survey in Denver: Mitsunobu Tatsumoto, Dan Unruh, Jon Patchett, and Bill White. The manuscript also benefited by suggestions made by Gregory Herzog, Tony Zaikowski and two anonymous reviewers. This research was supported by a grant from the National Science Foundation (NSF-EAR78-09251-01).

## REFERENCES

- Alaerts L., Lewis R. S., and Anders E. (1979a) Host phases of neon-E and s-process xenon in the Murchison C2 chondrite (abstract). In *Lunar and Planetary Science X*, p. 12–14. Lunar and Planetary Institute, Houston.
- Alaerts L., Lewis R. S., Matsuda J., and Anders E. (1979b) Noble gas components in the Murchison C2 chondrite. In *Programme of 42nd Annual Meeting of the Meteoritical Society*. Abstract no. 146, p. 93, Heidelberg, Germany.
- Alaerts L., Lewis R. S., Matsuda J.-I., and Anders E. (1980) Isotopic anomalies of noble gases in meteorites and their origins—VI. Presolar components in the Murchison C2 chondrite. *Geochim. Cosmochim. Acta* **44**, 189–209.
- Anders E., Heymann D., and Mazor E. (1970) Isotopic composition of primordial helium in carbonaceous chondrites. *Geochim. Cosmochim. Acta* **34**, 127–131.
- Arnould M. and Nørgaard H. (1978) Thermonuclear origin of Ne-E. *Astron. Astrophys.* **64**, 195–213.
- Arrhenius G. and Alfvén H. (1971) Fractionation and condensation in space. *Earth Planet. Sci. Lett.* **10**, 253–267.
- Audouze J., Bibring J. P., Dran J. C., Maurette M., and Walker R. M. (1976) Heavily irradiated grains and the neon isotope anomalies in carbonaceous chondrites. *Astrophys. J. Lett* **206**, L185–L189.
- Ballad R. V., Oliver L. L., Downing R. G., and Manuel O. K. (1979) Isotopes of tellurium, xenon, and krypton in Allende meteorite retain record of nucleosynthesis. *Nature* **277**, 615–620.
- Black D. C. (1972a) On the origins of trapped helium, neon and argon isotopic variations in meteorites-I. Gas rich meteorites, lunar soil and breccia. *Geochim. Cosmochim. Acta* **36**, 347–375.
- Black D. C. (1972b) On the origins of trapped helium, neon and argon isotopic variations in meteorites-II. Carbonaceous chondrites. *Geochim. Cosmochim. Acta* **36**, 377–394.
- Black D. C. (1972c) The A, B, C's of trapped helium, neon and argon in meteorites and lunar samples. In *Symposium on the Origin of the Solar System* (H. Reeves, ed.), p. 237–244. Edition du Centre National de la Recherche Scientifique, Paris.
- Black D. C. and Pepin R. O. (1969) Trapped neon in meteorites II. *Earth Planet. Sci. Lett.* **6**, 395–405.
- Boynton W. V. (1978a) The chaotic solar nebula: Evidence for episodic condensation in several distinct zones. In *Protostars and Planets* (T. Gehrels, ed.), p. 427–438. Univ. Ariz. Press, Tucson.
- Boynton W. V. (1978b) Rare earth elements as indicators of supernova condensation (abstract). In *Lunar and Planetary Science IX*, p. 120–122. Lunar and Planetary Institute, Houston.
- Cameron A. G. W. and Truran J. W. (1977) The supernova trigger for formation of the solar system. *Icarus* **30**, 447–461.
- Clayton D. D. (1975) Na<sup>22</sup>, Ne-E, extinct radioactive anomalies and unsupported Ar<sup>40</sup>. *Nature* **257**, 36–37.
- Clayton D. D. (1979) Supernovae and the origin of the solar system. *Space Sci. Rev.* **24**, 147–226.
- Clayton D. D., Druck E., and Woosley S. E. (1977) Isotopic anomalies and proton irradiation in the early solar system. *Astrophys. J.* **214**, 300–315.
- Clayton R. N., Grossman L., and Mayeda T. K. (1973) A component of primitive nuclear composition in carbonaceous meteorites. *Science* **182**, 485–488.
- Clayton R. N. and Mayeda T. (1977) Correlated oxygen and magnesium isotopic anomalies in Allende inclusions: I. Oxygen. *Geophys. Res. Lett.* **4**, 295–298.
- Eberhardt P. (1978) A neon-E rich phase in Orgueil: Results of stepwise heating experiments. *Proc. Lunar Planet. Sci. Conf. 9th*, p. 1027–1054.
- Eberhardt P., Jungck M. H. A., Meier F. O., and Niederer F. (1979a) Neon-E: New limits for isotopic composition. Two host phases (abstract)? In *Lunar and Planetary Science X*, p. 341–343. Lunar and Planetary Institute, Houston.
- Eberhardt P., Jungck M. H. A., Meier F. O., and Niederer F. (1979b) Presolar grains in Orgueil: Evidence from neon-E. *Astrophys. J.* **234**, L169–L171.
- El Goresy A., Nagal K., and Ramdohr P. (1978) Fremdlinge and their noble relatives. *Proc. Lunar Planet Sci. Conf. 9th*, p. 1279–1303.

- Fallick A. E. and Pillinger C. T. (1979) Meteorites—life begins at seventy. *Nature* **281**, 527–528.
- Fowler W. A., Greenstein J. L., and Hoyle F. (1962) Nucleosynthesis during the early history of the solar system. *Geophys. J. Roy. Astron. Soc.* **6**, 148–220.
- Frick U., Mack R., and Chang S. (1979) Noble gas trapping and fractionation during synthesis of carbonaceous matter. *Proc. Lunar Planet. Sci. Conf. 10th*, p. 1961–1973.
- Geiss J., Bühler F., Cerutti H., Eberhardt P. and Filleux Ch. (1972) Solar wind composition experiment. *Apollo 16 Prelim. Sci. Rep.* NASA SP-315, p. 1–10.
- Grossman L. (1980) Refractory inclusions in the Allende meteorite. *Ann. Rev. Earth Planet. Sci.* **8**, 559–608.
- Grossman L. and Larimer J. W. (1974) Early chemical history of the solar system. *Rev. Geophys. Space Phys.* **12**, 71–101.
- Heymann D. and Dziczkaniec M. (1976) Early irradiation of matter in the solar system: Magnesium (proton, neutron) scheme. *Science* **191**, 79–81.
- Hohenberg C. M., Davis P. K., Kaiser W. A., Lewis R. S., and Reynolds J. H. (1970) Trapped and cosmogenic rare gases from stepwise heating of Apollo 11 samples. *Proc. Apollo 11 Lunar Sci. Conf.*, p. 1283–1309.
- Hutchison R. (1980) Interstellar grains in meteorites? *Nature* **283**, 813.
- Jeffery P. M. and Hagan P. J. (1969) Negative muons and isotopic composition of the rare gases in the Earth's atmosphere. *Nature* **223**, 1253.
- Jungck M. H. A. and Eberhardt P. (1979) Ne-E in Orgueil density separates. *Meteoritics* **14**, 439–441.
- Kuroda P. K. (1979) Isotopic anomalies in the early solar system. *Geochem. J.* **13**, 83–90.
- Kuroda P. K. and Manuel O. K. (1970) Mass fractionation and isotope anomalies in neon and xenon. *Nature* **227**, 1113–1116.
- Larimer J. W. and Anders E. (1967) Chemical fractionations in meteorites—II. Abundance patterns and their interpretation. *Geochim. Cosmochim. Acta* **31**, 1239–1270.
- Lattimer J. M., Schramm D. N., and Grossman L. (1978) Condensation in supernova ejecta and isotopic anomalies in meteorites. *Astrophys. J.* **219**, 230–249.
- Lee T., Papanastassiou D. A., and Wasserburg G. J. (1978) Ca isotopic anomalies in the Allende meteorite. *Astrophys. J. Lett.* **220**, L21–L25.
- Lewis R. S., Alaerts L., Matsuda J.-I. and Anders E. (1979) Stellar condensates in meteorites: Isotopic evidence from noble gases. *Astrophys. J.* **234**, L165–L168.
- Lewis R. S., Srinivasan B., and Anders E. A. (1977a) Reply to comment by O. K. Manuel and D. D. Sabu. *Science* **195**, 209–210.
- Lewis R. S., Gros J., and Anders E. A. (1977b) Isotopic anomalies of noble gases in meteorites and their origins: 2: Separated minerals from Allende. *J. Geophys. Res.* **82**, 779–792.
- Manuel O. K. (1967) Noble gases in the Fayetteville meteorite. *Geochim. Cosmochim. Acta* **31**, 2413–2424.
- Manuel O. K. (1978) Review of meteoritic noble gases. In *Proceedings of the Robert A. Welch Foundation, Conferences on Chemical Research. XXI Cosmochemistry* (W. O. Milligan, ed.), p. 263–272. The Robert A. Welch Foundation, Houston.
- Manuel O. K. (1979) Comment on 'Isotopic anomalies of noble gases in meteorites and their origins. 2: Separated minerals from Allende' by Roy S. Lewis, Jacques Gross and Edward Anders. *J. Geophys. Res.* **84**, 5683–5684.
- Manuel O. K. (1980) The enigma of helium and anomalous xenon. *Icarus* **41**, 312–315.
- Manuel O. K., Hennecke E. W., and Sabu D. D. (1972) Xenon in carbonaceous chondrites. *Nature* **240**, 99–101.
- Manuel O. K. and Sabu D. D. (1975) Elemental and isotopic inhomogeneities in noble gases: The case for local synthesis of the chemical elements. *Trans. Mo. Acad. Sci.* **9**, 104–122.
- Manuel O. K. and Sabu D. D. (1977) Strange xenon, extinct superheavy elements and the solar neutrino puzzle. *Science* **195**, 208–209.
- Megrué G. H. and Steinbrunn F. (1972) Classification and source of lunar soils, clastic rocks and individual mineral rock and glass fragments from Apollo 12 and 14 samples as determined by the

- concentration gradients of the helium, neon and argon isotopes. *Geochim. Cosmochim. Acta, Suppl.* **3**, 1899–1916.
- Meier F. O., Jungck M. H. A., and Eberhardt P. (1980) Evidence for pure neon-22 in Orgueil and Murchison (abstract). In *Lunar and Planetary Science XI*, p. 723–725. Lunar and Planetary Institute, Houston.
- Niederer F. and Eberhardt P. (1977) A neon-E rich phase in Dimmitt. *Meteoritics* **12**, 327–331.
- Nier A. O. (1950) A redetermination of the relative abundances of the isotopes of neon, krypton, rubidium, xenon and mercury. *Phys. Rev.* **79**, 450–454.
- Pepin R. O. (1967) Trapped neon in meteorites. *Earth Planet. Sci. Lett.* **2**, 13–18.
- Pepin R. O. (1968) Neon and xenon in carbonaceous chondrites. In *Origin and Distribution of the Elements* (L. H. Ahrens, ed.), p. 379–386. Pergamon, N.Y.
- Pepin R. O. and Signer P. (1965) Primordial rare gases in meteorites. *Science* **149**, 253–265.
- Podosek F. A. (1978) Isotopic structures in solar system materials. *Ann. Rev. Astron. Astrophys.* **16**, 293–334.
- Sabu D. D. and Manuel O. K. (1976) The xenon record of element synthesis. Manuscript distributed during presentation of paper no. P58, 1976 Annual Meeting of the American Geophysical Union, 35 pp. (available from authors).
- Sabu D. D. and Manuel O. K. (1980) Noble gas anomalies and synthesis of the chemical elements. *Meteoritics* **15**, 117–138.
- Smith S. P., Huneke J. C., and Wasserburg G. J. (1978) Neon in gas-rich samples of the carbonaceous chondrites Mokoia, Murchison, and Cold Bokkeveld. *Earth Planet. Sci. Lett.* **39**, 1–13.
- Srinivasan B. (1973) Variations in the isotopic composition of trapped rare gases in lunar soil 14318. *Proc. Lunar Sci. Conf. 4th*, p. 2049–2064.
- Srinivasan B. and Anders E. (1978) Noble gases in the Murchison meteorite: Possible relics of s-process nucleosynthesis. *Science* **201**, 51–56.
- Srinivasan B., Gross J., and Anders E. (1977) Noble gases in separated meteorite minerals: Murchison (C2), Ornans (C3), Karoonda (C5) and Abee (E4). *J. Geophys. Res.* **82**, 762–778.
- Srinivasan B., Henneke E. W., Sinclair D. E., and Manuel O. K. (1972) A comparison of noble gases released from lunar fines (#15601.64) with noble gases in meteorites and in the earth. *Proc. Lunar Sci. Conf. 3rd*, p. 1927–1945.
- Srinivasan B. and Manuel O. K. (1971) On the isotopic composition of trapped helium and neon in carbonaceous chondrites. *Earth Planet. Sci. Lett.* **12**, 282–286.
- Stauffer H. (1961) Primordial argon and neon in carbonaceous chondrites and ureilites. *Geochim. Cosmochim. Acta* **24**, 70–82.
- Suess H. E. (1949) Die Häufigkeit der Edelgas auf der Erde und im Kosmos. *J. Geol.* **57**, 600–647.
- Tatsumoto M. and Shimamura T. (1980) Evidence for live curium-247 in the early solar system. *Nature*. In press.
- Zähringer J. (1968) Rare gases in stony meteorites. *Geochim. Cosmochim. Acta* **32**, 209–237.
- Zähringer J. and Gentner W. (1960) Uredelgase in einigen Steinmeteoriten. *Z. Naturforsch.* **15a**, 600–602.

## APPENDIX

### The blank problem

The available information on instrumental blanks at the laboratories in Chicago and Bern, the two laboratories where the recent work on Ne-E has been done, suggests a possible problem. To demonstrate this, we have computed the number of atoms of each neon isotope in the temperature fractions of Murchison separates 1C10 (Srinivasan and Anders, 1978), 2C10c, and 2C10c200 (Alaerts *et al.*, 1980) and Orgueil separates G4j (Eberhardt, 1978) and D1\* (Jungck and Eberhardt, 1979; Meier *et al.*, 1980) which show the greatest enrichments of Ne-E Ne-E(L), or Ne-E(H). The results are tabulated in Table 1 as atomic abundances so that the reader can appreciate the minute amounts of gas that are being analyzed when extremely low values of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio are being reported. Also shown in parentheses are values of blanks, B, and when available, the uncertainties on the blanks.



**Table A-1.** Atomic abundances of neon and blanks in temperature fractions of Orgueil and Murchison separates which show the greatest enrichments of Ne-E, Ne-E(L), or Ne-E(H)§.

| Sample    | $^{20}\text{Ne}/^{22}\text{Ne}$<br>( $\times 10^2$ ) | $^{22}\text{Ne}$ ( $^{22}\text{B}$ )<br>( $\times 10^8$ atoms) | $^{20}\text{Ne}$ ( $^{20}\text{B}$ )<br>( $\times 10^9$ atoms) | $^{21}\text{Ne}$ ( $^{21}\text{B}$ )<br>( $\times 10^6$ atoms) |
|-----------|--|--|--|--|
| Murchison |  |  |  |  |
| 1C10      |  |  |  |  |
| 1400°C    | 327  | 120.5 (19.2 $\pm$ 13.7)  | 39.4 (18.8 $\pm$ 13.4)   | 164.0 (55.7 $\pm$ 39.8)  |
| 2C10c     |  |  |  |  |
| 800°C     | 45   | 25.7 (19.2 $\pm$ 13.7, 2.2)                                    | 1.2 (18.8 $\pm$ 13.4, 2.2)                                     | 7.7 (55.7 $\pm$ 39.8, 6.5)                                     |
| 1200°C    | 40   | 14.5 (19.2 $\pm$ 13.7, 2.2)                                    | 0.6 (18.8 $\pm$ 13.4, 2.2)                                     | 31.9 (55.5 $\pm$ 39.8, 6.5)                                    |
| 2C10c200  |  |  |  |  |
| 1200°C    | 100  | 5.2 (19.2 $\pm$ 13.7, 3.0)                                     | 0.5 (18.8 $\pm$ 13.4, 3.0)                                     | 93.5 (55.7 $\pm$ 39.8, 8.8)                                    |
| 1400°C    | 89   | 11.7 (19.2 $\pm$ 13.7, 3.0)                                    | 1.0 (18.8 $\pm$ 13.4, 3.0)                                     | 105.1 (55.7 $\pm$ 39.8, 8.8)                                   |
| Orgueil   |  |  |  |  |
| G4j       |  |  |  |  |
| 600°C     | 241  | 16.967 (0.037)   | 4.082 (0.053)  | 115.21 (0.11)  |
| 1230°C    | 148  | 14.608 (0.037)   | 2.162 (0.053)  | 32.72 (0.11)   |
| D1*       |  |  |  |  |
| Av. Temp. | 1  | 0.645 (0.037)  | 0.0006 (0.053)   | 0.03 (0.11)  |

§ Sources of data are given in the text.

Values of blanks are not presented in the recent reports on Ne-E (Alaerts *et al.*, 1980; Jungck and Eberhardt, 1979; Meier *et al.*, 1980). The Chicago group (Alaerts *et al.*, 1980) report uncertainties in blanks but not the values of the blanks themselves, and the Bern group earlier reported values of blanks but not their uncertainties (Eberhardt, 1978). However, an earlier report on noble gases in Allende residues (Lewis *et al.*, 1977b) indicates that the neon blanks were of atmospheric composition and contained  $^{20}\text{Ne} = (7 \pm 5) \times 10^{-10}$  cc STP. Since attempts to obtain numerical values for blanks in the more recent analyses were unsuccessful, we have shown values of the blanks reported by Lewis *et al.* (1977b) and Eberhardt (1978) in Table 1. Alaerts *et al.* (1980) report lower blank uncertainties than Lewis *et al.* (1977b), and the former values are shown as the second entry for blank uncertainties in Table 1.

The amount of neon in D1\* has not been reported (Jungck and Eberhardt, 1979; Meier *et al.*, 1980). The method that we used to estimate the amount of neon in an average temperature fraction of the D1\* separates will be presented below.

### Neon-E in Murchison residues

It can be seen in Table 1 that the current of neon ions in the analysis of gas released from Murchison 1C10 residues of Murchison (Srinivasan and Anders, 1978) exceed those in the blank at all mass numbers. It was shown earlier in Fig. 3 that the low value of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio, 3.27, in this gas is accompanied by a  $^{21}\text{Ne}/^{22}\text{Ne}$  ratio with the value predicted by Eq. (2) for mass fractionation.

In their report on the analysis of Murchison residues, Alaerts *et al.* (1980) state that the “ $^{22}\text{Ne}$  blank corrections for 2C10c and 2C10c200 had uncertainties of  $0.4 \times 10^{-8}$  and  $0.7 \times 10^{-8}$  cm<sup>3</sup>/g.” Atomic abundances were calculated from these blank “uncertainties”, and the values are shown in Table 1 as the second blank uncertainty on  $^{22}\text{Ne}$ . Alaerts *et al.* (1980) also report that “The data have been corrected for procedural blanks, and the errors for isotopic ratios were increased by either the magnitude of the shift caused by the blank correction (Ar and Xe) or by the measured variation in the blank (Ne).” These authors do not report actual values of blanks or values of “uncertainties” in blanks at masses 20 or 21. We have assumed that the blank uncertainties are in atmospheric proportions in estimating the blank uncertainties listed in Table 1.

The data in Table 1 provide three points of interest concerning the possible occurrence of Ne-E in Murchison residues 2C10c and 2C10c200:

(i) The ion current at mass 20 from the blank “uncertainties” is itself larger than the ion current of  $^{20}\text{Ne}$  in those gas fractions that are reported to show extremely low values, 0.4–1.0, of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio (Alaerts *et al.*, 1980). It should be noted that the actual blank at mass 20 (Lewis *et al.*, 1977b) may be an order of magnitude higher than the  $^{20}\text{Ne}$  signal in these gas fractions.

(ii) If we have greatly overestimated the blank or the blank “uncertainty” at masses 20 and 21, because the blank itself is highly enriched at mass 22, then the evidence for enriched  $^{22}\text{Ne}$  in these analyses is dubious.

(iii) Alaerts *et al.* (1980) observed the lowest  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio in the gas released from the 2C10c residue of Murchison at 1200°C, where it was reported that  $^{20}\text{Ne}/^{22}\text{Ne} = 0.40 \pm 0.34$ . The blank “variation” in the mass 20 ion current alone appears from Table 1 to be about 4 times larger than the sample  $^{20}\text{Ne}$ , unless we have overestimated the blank “variation” at mass 20 for the reason noted above.

Taken together, these three points suggest that blanks may constitute an appreciable fraction of the ion current in the recent reports of Ne-E in residues of Murchison (Alaerts *et al.*, 1979a,b, 1980; Lewis *et al.*, 1979).

### Neon-E in Orgueil minerals

Table 1 shows only the most Ne-E-rich fractions released from Orgueil separate G4j at low, Ne-E(L), and high, Ne-E(H), extraction temperatures. It can be seen that the blanks given by Eberhardt (1978)

constitute only 1–2% of the  $^{20}\text{Ne}$  ion current in the gas released at these temperatures. However, this sample weighed 18 mg and contained a total of  $1.7 \times 10^{-9}$  cc STP  $^{22}\text{Ne}$ . The lowest value observed for the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio in G4j was only  $1.48 \pm 0.02$ .

Numerical values for isotopic ratios, sample weights, blanks, etc., have not yet been reported for the more Ne-E-rich fractions of Orgueil G5a which is reported to yield a limit of  $^{20}\text{Ne}/^{22}\text{Ne} < 0.62$  in Ne-E (Eberhardt *et al.*, 1979a,b) and D1\* which is reported to yield a limit of  $^{20}\text{Ne}/^{22}\text{Ne} < 0.01$  (Jungck and Eberhardt, 1979; Meier *et al.*, 1980). On the basis of the latter value, Eberhardt suggested in his presentation at the Eleventh Lunar and Planetary Science Conference that single-stage diffusive fractionation would require the loss of 60 orders of magnitude of neon to explain the  $^{22}\text{Ne}$  enrichment in Ne-E. In response to questions, he also acknowledged that the sample weight of D1\* was about 100  $\mu\text{g}$ .

As noted earlier, there is no *a priori* reason to assume that the fractionation of neon isotopes was limited to diffusive gas loss. Even if other fractionation mechanisms (Arrhenius and Alfvén, 1971; Frick *et al.*, 1979) were not known, there would be no rational basis for assuming that the diffusion of neon was limited to a single fractionation stage. However, an understanding of the experimental problems in measuring values of  $^{20}\text{Ne}/^{22}\text{Ne} < 0.01$  in the gas released by stepwise heating of a sample weighing about 100  $\mu\text{g}$  is more relevant to the subject at hand than are arguments about the degree of fractionation that may be necessary to explain the results.

Eberhardt *et al.* (1979a) report that “The Ne-E concentration in G5a is  $24 \times 10^{-8}$  cm<sup>3</sup> STP  $^{22}\text{Ne}$ /g . . .” This is unclear because the analyses provided only a lower limit on the  $^{22}\text{Ne}$  content of Ne-E. If the stated value represents the total  $^{22}\text{Ne}$  in G5a, then we estimate that G5a contains about 2 times the  $^{22}\text{Ne}$  content of G4j but only about 1/2 of the  $^{20}\text{Ne}$  content of G4j. Eberhardt *et al.* (1979a,b) report that the G5a separates ( $< 2.3$  g cm<sup>-3</sup>) contain about 3 to 7 times more cm<sup>3</sup> STP  $^{22}\text{Ne}$ /g than the next 7 more dense fractions, G5b-G5h (2.3–4.05 g cm<sup>-3</sup>). We will not attempt to estimate the blank contribution to the 20-mass peak in the analyses of G5a, except to note that the fractional blank contribution at mass 20 would be about twice that in G4j *if the sample weights were identical* (18 mg) and appreciably greater if the sample weight of G5a was appreciably lower than 18 mg.

Even fewer experimental details are published for the analysis of D1\* separates ( $< 2.5$  g cm<sup>-3</sup>). The neon contents shown in Table 1 for D1\* were estimated by assuming that its  $^{22}\text{Ne}$  content is comparable to that of G5a, the least dense ( $< 2.3$  g cm<sup>-3</sup>) and most  $^{22}\text{Ne}$ -rich of the G5a-G5h separates (Eberhardt *et al.*, 1979a, b). In that case, the 100  $\mu\text{g}$  sample of D1\* would contain about  $24 \times 10^{-12}$  ccSTP  $^{22}\text{Ne}$ .

The amounts of  $^{20}\text{Ne}$  and  $^{21}\text{Ne}$  in the bulk neon of D1\* were then calculated from a plot of the neon isotopic ratios in these separates (Jungck and Eberhardt, 1979; Meier *et al.*, 1980), e.g.,  $^{20}\text{Ne}/^{22}\text{Ne} \sim 1 \times 10^{-2}$  for the bulk neon in D1\* so its  $^{20}\text{Ne}$  content is about  $24 \times 10^{-14}$  cc STP. This neon was released in “ten or more temperature steps” (Jungck and Eberhardt, 1979), and the average amount of  $^{20}\text{Ne}$  in each temperature fraction would be, by our estimate, about  $2.4 \times 10^{-14}$  cc STP, i.e.,  $6 \times 10^5$  atoms. As shown in Table 1, the  $^{20}\text{Ne}$  abundance in an average temperature fraction of D1\* is expected to be almost two orders of magnitude less than the blank value that Eberhardt (1978) reported at mass 20.

Thus, the data base for extreme anomalies of neon in mineral separates of Murchison and Orgueil seems to be characterized by analyses in which blanks constitute a large part of the ion current. The possibility that very low values of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio might be an artifact of data reduction cannot be fairly evaluated in the absence of experimental details on uncorrected ratios, blanks, blank variations, etc.