

nature of the temporal and spatial variations in the degree of acidity suggest that the chemical reactions producing and neutralizing acid sulfate take place to a significant extent on a time scale of a few hours. The strong negative correlation between acid sulfate and hydrocarbon would appear to suggest either that hydrocarbon inhibits sulfate formation in some way or that the hydrocarbon species are too volatile in an acidic medium to remain associated with the acid sulfate particulates. Finally, as has been suggested by others (13) on the basis of an apparent upper limit for sulfate concentration in the atmosphere, the observation that the occurrence of acid sulfate is not closely linked with high concentrations of sulfur dioxide suggests that the formation mechanism is one in which other factors control the rate of sulfate formation.

P. T. CUNNINGHAM  
S. A. JOHNSON

Chemical Engineering Division,  
Argonne National Laboratory,  
Argonne, Illinois 60439

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12. The most important effects produced by impactor collection probably involve reactions between particles and changes in water content. Our samples clearly indicate that, in many cases, especially when the relative humidity is high, recrystallization of the sample occurs on the impaction surface; and, on analysis of the sample, its water content is presumably less than that of the ambient aerosol. On the other hand, reaction of the impacted sample with ammonia or sulfur dioxide in the air appears to be less of a problem than for filter samples.
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## Early Irradiation of Matter in the Solar System: Magnesium (Proton, Neutron) Scheme

**Abstract.** *The occurrence of positive and negative  $^{26}\text{Mg}$  anomalies in inclusions of the Allende meteorite is explained in terms of proton bombardment of a gas of solar composition. A significant fraction of  $^{26}\text{Mg}$  in the irradiated gas is stored temporarily in the form of radioactive  $^{26}\text{Al}$  by the reaction  $^{26}\text{Mg}(p,n)^{26}\text{Al}$ . Proton fluxes of  $10^{17}$  to  $10^{19}$  protons per square centimeter per year at 1 million electron volts are inferred. Aluminum-rich materials condensing from the gas phase have positive  $^{26}\text{Mg}$  anomalies, whereas magnesium-rich materials have negative  $^{26}\text{Mg}$  anomalies. The proton flux required to account for the observed magnesium anomalies is used to investigate possible isotopic anomalies in the elements from oxygen to argon. Detectable isotopic anomalies are predicted only for neon. The anomalous neon is virtually pure  $^{22}\text{Ne}$  from  $^{22}\text{Na}$  decay. The predicted amount of anomalous  $^{22}\text{Ne}$  is about  $10^{-8}$  cubic centimeter (at standard temperature and pressure) per milligram of sodium.*

Gray and Compston (1) and Lee and Papanastassiou (2) have reported isotopic anomalies of Mg in inclusions of the Allende meteorite. Their observations are enigmatic because  $^{26}\text{Mg}$  is sometimes enriched and sometimes depleted relative to  $^{25}\text{Mg}$ . These investigators have presented ingenious explanations for their results. Following the suggestion of Gray and Compston (1), we will briefly consider the possible effects of proton bombardment. We believe that it is much too early to rule out any viable idea.

Protons in the energy range of galactic cosmic rays are undoubtedly capable of changing the isotopic composition of Mg by spallation in Al and Si. Judging from the well-studied system Ne-Na-Mg (3), one predicts that such protons will always lower the  $^{24}\text{Mg}/^{26}\text{Mg}$  ratio, will leave the  $^{25}\text{Mg}/^{26}\text{Mg}$  ratio virtually unchanged in Si-rich systems, and will perhaps decrease the  $^{23}\text{Mg}/^{26}\text{Mg}$  ratio in Al-rich systems. However, one must posit vast numbers of galactic protons, on the order of  $10^{21}$  to  $10^{22}$  proton  $\text{cm}^{-2}$  with assumed cross sections  $\sigma$  of 10 millibarns, to account for the observed effects (1, 2). Depletion of  $^{26}\text{Mg}$  is virtually ruled out. From this reasoning we conclude that protons in this energy range are not the cause of the observed Mg anomalies.

The effects of protons of intermediate energy, say 10 to 100 Mev, are difficult to evaluate at this time because of the plethora of possible reactions in Mg, Al, and Si for which cross sections are unknown. Below 10 Mev the number of possible reactions in these elements becomes much smaller, and (p,n) and (p, $^4\text{He}$ ) reactions are probably the most important ones. Here we will consider only (p,n) reactions in Mg. Reactions such as  $^{27}\text{Al}(p,^4\text{He})^{24}\text{Mg}$  and  $^{26}\text{Mg}(p,^4\text{He})^{23}\text{Na}$ , which have  $Q_m$  values (the mass threshold of a nuclear reaction, that is, the positive or negative change in mass between reactants and products) of +1.6 and -1.8 Mev, respectively, must eventually be considered.

The (p,n) reactions in the stable isotopes of Mg are "cyclic" as the reaction products  $^{24}\text{Al}$ ,  $^{25}\text{Al}$ , and  $^{26}\text{Al}$  decay by  $\beta^+$  emission to the target nuclides from which they had been formed. If the radiation remains unchanged for periods longer than the mean lifetime of  $^{26}\text{Al}$ , about  $10^6$  years, a steady state is reached after a few million years in which constant fractions of each of the stable Mg isotopes are "stored" temporarily in the form of their respective radioactive Al-precursors. Because of the very short mean lifetimes of  $^{24}\text{Al}$  and  $^{25}\text{Al}$  (3.0 and 10.4 seconds, respectively), the fractions of  $^{24}\text{Mg}$  and  $^{25}\text{Mg}$  so stored are negligibly small. Because of the cyclic nature of the (p,n) reactions, irradiation of solid grains containing Mg and Al does not result in isotopic anomalies in Mg, as all of the  $^{24}\text{Mg}$ ,  $^{25}\text{Mg}$ , and  $^{26}\text{Mg}$  atoms that are stored temporarily in  $^{24}\text{Al}$ ,  $^{25}\text{Al}$ , and  $^{26}\text{Al}$  are eventually (after a significant number of  $^{26}\text{Al}$  half-lives) restored to the system.

We will consider therefore the effects of the irradiation of a "dust-free" gas (or plasma). When, after several million years, equilibrium is reached, the  $^{26}\text{Al}/^{26}\text{Mg}$  ratio in the gas phase depends only on the proton spectrum and the excitation function  $^{26}\text{Mg}(p,n)^{26}\text{Al}$ , which has a  $Q_m$  value of about 5 Mev (4). The  $^{26}\text{Al}/^{27}\text{Al}$  ratio at equilibrium depends also on the Mg/Al abundance ratio in the gas phase, which we assume here to be 0.1, the solar value (5).

For the calculations we have assumed that the (differential) proton spectra are of the form  $dF/dE = kE^{-\gamma}$ , where  $dF/dE$  is the differential proton flux between  $E$  and  $E + dE$  (in protons per square centimeter per year);  $k$  is a normalization constant;  $E$  is the kinetic energy of the protons (in millions of electron volts); and  $\gamma$  is a parameter representing the steepness of the energy distribution. We have used  $\gamma$  values of 2.5, 3.5, and 4.5, which cover present-day galactic cosmic protons and a substantial range of known  $\gamma$  values in present-day solar flares (6). The  $\sigma$  values for  $^{26}\text{Mg}(p,n)^{26}\text{Al}$  were taken from Furukawa

*et al.* (4), and  $\int (dF/dE)\sigma(E)dE$  was determined by numerical integration from the adopted  $Q_m$  of 5 Mev up to 50 Mev. Because the largest observed  $^{26}\text{Mg}$  anomaly is  $-0.2$  percent, we have first calculated the proton fluxes required to yield  $(^{26}\text{Al}/^{26}\text{Mg})_{\text{equilibrium}} = 2 \times 10^{-3}$ . These are:  $k^{-2.5} = 5.7 \times 10^{17}$ ,  $k^{-3.5} = 3.28 \times 10^{18}$ , and  $k^{-4.5} = 4.32 \times 10^{19}$  proton  $\text{cm}^{-2}$  year $^{-1}$  (superscripts refer to the  $\gamma$  values used).

These fluxes are several orders of magnitude larger than the annual averages of solar flare protons at 1 A.U. during the last few solar cycles. Where did these protons come from? The sun during its pre-main sequence (T Tauri) phase is a possible source. During this stage, the star exhibits a vastly increased surface activity as shown by its substantial fluctuations in luminosity. Fowler *et al.* (7) have considered the T Tauri phase of the sun for the nucleosynthesis of D, Li, Be, and B in the solar system. Although these ideas have been largely abandoned today, their hypothesis does call for vast proton (and neutron) fluxes, comparable to the numbers given above.

Kuhi (8) has shown that T Tauri stars lose mass at the staggering rate of about  $3.7 \times 10^{-8}$  solar mass per year (albeit in the form of atoms and ions with energies in the range of 1 kev per nucleon). He has suggested that the ionization in the expanding stellar envelope might be due to high-energy protons, and has deduced a flux of  $1.6 \times 10^{22}$  proton  $\text{cm}^{-2}$  year $^{-1}$  (protons of energy 3 Mev). Herbig (9) has pointed out that the Li abundance in FU Ori requires either that the high-energy proton flux at the surface of this T Tauri star be some  $10^{11}$  to  $10^{12}$  times present solar levels or that the invisible prestellar source of this object contained abundant high-energy protons.

The isotopic Mg anomalies have been observed in a solid object, the Allende meteorite. Hence we must consider the consequences of condensation and accretion. According to current thinking (10), the first compounds to appear in a cooling gas of solar composition are enriched in Al/Mg relative to the gas phase, with Mg-enriched compounds such as enstatite appearing only much later and at lower temperatures. Let us first consider the unrealistic case of the condensation of all Al (in the form of Al-containing minerals), followed later by the condensation of all Mg. Accretion of the condensates into larger objects yields the simple prediction that inclusions of the Allende meteorite in which the Al/Mg ratio is lower than that of the gas—presumed to be about 0.1, the solar value—must show depletion of  $^{26}\text{Mg}$  and inclusions in which the Al/Mg ratio is greater than that of the gas must show

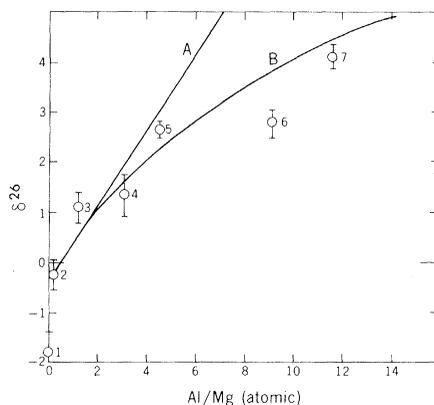


Fig. 1. Plot of  $\delta^{26}$  as a function of Al/Mg (atomic) for samples from the Allende meteorite. Data are from (1, 2). Point 1, average of four values for sample C1 (2); point 2, average of three negative values for sample B12 (2); point 3, value for sample B31 (2); point 4, value for sample B32 (2); point 5, average of three values for sample B29 (2); point 6, average of three values for sample B30 (2); point 7, value in inclusion 1 (1). Curve A is a linear array of points 2, 3, 4, and 5; proton flux,  $4 \times 10^{16}$  proton  $\text{cm}^{-2}$  year $^{-1}$  at 5 Mev. Curve B is one possible curve for the gradual removal of Al and Mg from an irradiated gas (see text); proton flux,  $\sim 10^{15}$  proton  $\text{cm}^{-2}$  year $^{-1}$  at 5 Mev.

$^{26}\text{Mg}$  enrichment. In Fig. 1 we have plotted the observed  $^{26}\text{Mg}$  anomalies against the Al/Mg ratio in the sample. The Al/Mg ratio for the samples with the largest negative anomalies (point 1) is not known; we have placed this point arbitrarily on the ordinate even though the aggregate from which these samples come has been described as "Ca-Al chondrules, extremely alkali poor" (2). Because of the small  $\delta^{26}$  values,

$$\delta^{26} \equiv \left[ \frac{(^{26}\text{Mg}/^{25}\text{Mg})_{\text{sample}}}{(^{26}\text{Mg}/^{25}\text{Mg})_{\text{standard}}} - 1 \right] \times 100$$

one expects the points to form a linear array. They do not. Only points 1, 6, and 7 form a linear array with  $(\text{Al}/\text{Mg})_{\text{gas}} = 2.7$ . Points 2, 3, 4, and 5 form a linear array (curve A) with  $(\text{Al}/\text{Mg})_{\text{gas}} = 0.5$ . Both  $(\text{Al}/\text{Mg})_{\text{gas}}$  ratios are unacceptable because they imply that the gas phase was enriched in Al/Mg relative to the solar value. Points 3, 4, 5, 6, and 7 fall near a straight line, but this line implies a negative  $(\text{Al}/\text{Mg})_{\text{gas}}$  ratio, which is unacceptable.

A gradual rather than sudden removal of Al and Mg from the gas has interesting consequences as long as the rate of removal is commensurate with the mean lifetime of  $^{26}\text{Al}$  such that the steady state is only very slightly disturbed ("batch" removal with sufficient time elapsed between the batches for reestablishment of the steady state is an acceptable alternative). The main result is that Mg remaining in the gas phase becomes increasingly depleted in  $^{26}\text{Mg}$  beyond  $\delta^{26} = -0.2$  percent, the value

adopted above, an effect well known in isotope separation. This conclusion is equivalent to saying that the proton flux can be relaxed. If the samples in the upper right-hand corner of Fig. 1 (points 6 and 7) are the "earliest condensates," one might expect the points to fall on a curve which becomes increasingly concave toward the abscissa for decreasing Al/Mg ratios (curve B is an example). Samples with the "solar" Al/Mg ratio (point 2) are permitted to be depleted in  $^{26}\text{Mg}$ ! This hypothesis is certainly grossly consistent with the observations. The required proton flux is now determined by point 7 with  $\delta^{26}$  about  $+0.4$  percent and Al/Mg about 10, which implies a 50-fold decrease of the  $k$  values given above.

The irradiation of a "dusty" gas, one that contains solid Mg- and Al-containing grains at the onset of the irradiation leads to an almost limitless number of variations with an almost unlimited freedom of speculation. As in the earlier case, isotopic Mg anomalies are produced only in the gas phase. Both Al and Mg are gradually removed from the gas either by the growth of preexisting grains, by the growth of new grains, or by both. As a result, Mg-anomalous materials are mixed with Mg-normal materials; hence the Al/Mg ratio measured in Allende aggregates becomes irrelevant, and the relevant ratio  $(\text{Al}/\text{Mg})_{\text{gas}}$  at the time of condensation cannot be known. The correlation between  $\delta^{26}$  and Al/Mg could be almost random. The trend in Fig. 1 appears to favor the irradiation of a virtually dust-free gas.

Flux variations can bring apparently deviant results into line; however, the long mean lifetime of  $^{26}\text{Al}$  is a powerful "buffer" on flux variations for an era we assume to have lasted for only  $10^7$  years. On the other hand, if we assume constant proton flux, it is possible that Al and Mg removal from the gas began before the  $^{26}\text{Al}$  steady state was reached. Straight lines connecting points 1 and 6 and 7, 1 and 5, and so forth in Fig. 1 progressively increase in slope. Hence, Fig. 1 might represent a record of the first  $\sim 5 \times 10^6$  years of the irradiation with only point 1 representing steady-state conditions. This hypothesis raises the  $k$  values to the numbers given above.

Finally, if the (p,n) scheme with the gradual removal of radioactive precursors is correct, we can make predictions concerning observable isotopic effects in other elements. We will restrict our discussion to elements with two or more stable isotopes from O to Ar. The case of O is complicated by the fact that  $^{16}\text{F}$  (half-life  $t_{1/2} \sim 10^{-19}$  second) is not a  $\beta^+$  emitter but is proton-unstable and decays via  $^{15}\text{O}$  ( $t_{1/2} = 124$  seconds) to  $^{15}\text{N}$ . Both  $^{17}\text{F}$  and  $^{18}\text{F}$  are  $\beta^+$  emit-

ters ( $t_{1/2} = 66$  seconds and 110 minutes, respectively). Because of the short mean lifetimes of  $^{17}\text{F}$  and  $^{18}\text{F}$ , it is very unlikely that the (p,n) scheme could result in observable isotopic effects in O. We do not think that the large (up to 5 percent) isotopic O effects seen in certain carbonaceous chondrites (11, 12) can be explained in terms of the (p,n) scheme.

The case of Ne is more promising because solid objects in the solar system are very strongly fractionated in Ne; hence very small quantities of proton-produced stable Ne isotopes may be detectable. We have estimated the  $R_{22/23} = (^{22}\text{Na}/^{23}\text{Na})_{\text{equilibrium}}$  ratios for the reaction  $^{22}\text{Ne}(p,n)^{22}\text{Na}$  ( $^{22}\text{Na}$ ,  $t_{1/2} = 2.6$  years) in a gas of solar composition as  $R_{22/23}^{-2.5} = 1.3 \times 10^{-10}$ ,  $R_{22/23}^{-3.5} = 1.7 \times 10^{-11}$ , and  $R_{27/23}^{-4.5} = 2.4 \times 10^{-12}$ . For the case of sudden condensation of Na, the quantities of  $^{22}\text{Ne}$  that result from subsequent  $^{22}\text{Na}$  decay in Na-containing solid phases are  $7.4 \times 10^{-9}$ ,  $5.6 \times 10^{-9}$ , and  $1.0 \times 10^{-8} \text{ cm}^3$  at standard temperature and pressure per milligram of Na for the three equilibrium ratios given above, which must be detectable in carbonaceous chondrites which typically contain 0.5 percent (by weight) of Na. Because of the short half-lives of  $^{21}\text{Na}$  ( $t_{1/2} = 23$  seconds) and  $^{20}\text{Na}$  ( $t_{1/2} = 0.4$  second), the  $(^{21}\text{Na}/^{22}\text{Na})_{\text{equilibrium}}$  and  $(^{20}\text{Na}/^{22}\text{Na})_{\text{equilibrium}}$  ratios in the gas are extremely low. We have considered all possible reactions in Na, Mg, Al, and Si that lead to  $^{20}\text{Na}$ ,  $^{21}\text{Na}$ , and  $^{22}\text{Na}$  and have concluded that neither of the two equilibrium ratios can be greater than  $10^{-6}$ . Since the radioactive F-precursors  $^{20}\text{F}$ ,  $^{21}\text{F}$ , and  $^{22}\text{F}$  have half-lives of 11, 4.4, and 4 seconds, respectively, our scheme results in a Ne component that is virtually pure  $^{22}\text{Ne}$ . Perhaps this is the origin of neon-E [for a discussion of neon E, see (13)].

The  $\beta^+$ -emitting precursors of the stable Si isotopes have half-lives shorter than 2.5 minutes ( $^{30}\text{P}$ ); hence we expect no isotopic anomalies in Si. The  $\beta^+$ -emitting precursors of the stable S isotopes have half-lives shorter than 30 minutes ( $^{34m}\text{Cl}$ ); hence we expect no isotopic anomalies in S.

The  $\beta^+$ -emitting precursors of  $^{36}\text{Al}$  and  $^{38}\text{Ar}$  ( $^{40}\text{Ar}$  effects in solid objects are strongly masked by radiogenic  $^{40}\text{Ar}$  from the decay of natural  $^{40}\text{K}$ ) have half-lives shorter than 7.7 minutes ( $^{38}\text{K}$ ). Although Ar, like Ne, is strongly fractionated in solid objects, our preliminary estimates indicate that  $^{38}\text{Ar}$  enhancements are probably undetectably small.

DIETER HEYMANN

MARLENE DZICZKANIEC

Department of Space Physics and  
Astronomy and Department of Geology,  
Rice University, Houston, Texas 77001

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14. I thank Dr. D. D. Clayton for stimulating discussions, Dr. Huneke for allowing me to quote from his unpublished paper, and the two unknown reviewers for their constructive criticism. This research was supported by NASA grant NGL-44-006-127.

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## Laboratory Simulation of Thermal Convection in Rotating Planets and Stars

**Abstract.** *Because of dynamical constraints in a rotating system, the component of gravity perpendicular to the axis of rotation is the dominant driving force of convection in liquid planetary cores and in stars. Except for the sign, the centrifugal force closely resembles the perpendicular component of gravity. Convection processes in stars and planets can therefore be modeled in laboratory experiments by using the centrifugal force with a reversed temperature gradient.*

Convection driven by thermal buoyancy is a major source of fluid motions in planets and in stars. In general, buoyancy forces result from superadiabatic temperature gradients in the nearly spherically symmetric gravity fields of celestial bodies. Progress in the understanding of natural convection processes has depended to a large extent on the interaction of theoretical analysis and experimental observation (1). Because of the difficulty of simulating spherical gravity in the laboratory, experimental investigations have been restricted to convection in a plane layer heated from below. Both this case and convection in a spherical fluid shell of a nonrotating planet or star share the property of horizontal isotropy. When rotation becomes important the spherical symmetry is lost and the analogy with the laboratory convection layer no longer holds. The most significant difference arises from the fact that the angle between gravity and the vector  $\Omega$  of angular velocity varies in self-gravitating bodies, while the two vectors must be parallel in the laboratory experiment. When  $\Omega$  is inclined with respect to the vertical, gravity gives rise to a nonstationary body force in the rotating system. Although it may appear that the experimental simulation of convection in rotating self-gravitating spherical systems is even more difficult than in the nonrotating case, the presence of the centrifugal force in a rotating laboratory apparatus provides a new possibility for simulation experiments, as we outline in this report.

The importance of rotation for the dynamics of fluids is measured by two non-dimensional parameters, the Ekman num-

ber and the Rossby number. The latter represents the ratio between the vorticity of motions relative to the rotating system and the angular velocity  $\Omega$ . The Ekman number is defined by

$$E = \nu/L^2\Omega$$

where  $\nu$  is the kinematic viscosity of the fluid and  $L$  is a typical length, say the radius of a planetary liquid core. In many applications both the Ekman and the Rossby number are very small. In the limit where these parameters vanish the Proudman-Taylor theorem for a stationary velocity field  $\mathbf{v}$  in a homogeneous incompressible fluid holds

$$\Omega \cdot \nabla \mathbf{v} = 0 \quad (1)$$

This equation states that the velocity cannot vary in the direction of the axis of rotation. The theorem also holds for a barotropic fluid if the velocity vector is replaced by the momentum vector.

Motions with a nonvanishing radial component in a fluid sphere or spherical shell cannot satisfy Eq. 1 exactly because the normal velocity component must vanish at the boundary. On the other hand, strongly time-dependent flows, which do not have to obey Eq. 1, are inefficient in releasing potential energy because of the phase lag between thermal buoyancy and radial motion. The theoretical investigation (2) of the problem shows that in the realized convective flow the constraint of Eq. 1 is overcome by the combination of a slow time dependence and viscous forces associated with the small length scale in the azimuthal direction. As shown in Fig. 1, the motions occur in the form of thin