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EQUILIBRIUM PROFILES OF ATOMIC ^{7}Be AND ^{10}Be IN THE ATMOSPHERE ABOVE 100 KM

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Abstract. Theoretical profiles of cosmogenic ^{7}Be in the atmosphere above 100 km were computed in an effort to explain unexpectedly high accumulations of this radionuclide found on the leading surfaces of the Long Duration Exposure Facility (LDEF) spacecraft. Our diffusion calculations suggest that gravitational fractionation is sufficient to explain most, and possibly all, of the observed ^{7}Be enrichment at the recovery altitude of the spacecraft, provided only that the nuclide exists at and above the turbopause predominantly in the form of free atoms. Upward diffusion of ^{7}Be atoms through the turbopause appears to occur rapidly enough to largely offset losses at higher altitudes due to radioactive decay. The same model is used to compute cosmogenic ^{10}Be profiles in order to predict the probable outcome of planned measurements of ^{10}Be accumulations on LDEF surfaces.

Introduction

Recently Fishman et al. (1991) reported that unexpectedly high concentrations ($\sim 5 \times 10^{5}$ atoms cm^{-2}) of the radionuclide ^{7}Be had been found on the leading surfaces of the Long Duration Exposure Facility (LDEF), an unmanned satellite which was recovered in early 1990 after nearly 6 years in orbit. It was determined that the observed ^{7}Be had not been produced by direct cosmic ray action on the materials in the satellite itself; rather, traces of this nuclide had evidently been present in the tenuous gases of the atmosphere at the satellite's orbital altitude and had been collected by impaction.

It has long been known that ^{7}Be is created throughout the atmosphere, especially in and above the stratosphere, as a product of the spallation of oxygen and nitrogen nuclei by energetic cosmic rays. Yet the reported surface concentrations of ^{7}Be on the satellite implied an atmospheric concentration of at least 1.1×10^{-7} atoms cm^{-3}. Depending on the production rate and atmospheric density assumed for the LDEF's recovery altitude of 310 km, this value is between ~500 and ~5000 times larger than the ^{7}Be concentration expected from a steady state balance between radioactive decay and in situ production by cosmic rays. The authors of the initial report concluded that an as-yet unidentified mechanism was responsible for enriching the uppermost atmosphere with respect to ^{7}Be, most probably by transport from lower altitudes. They also noted that any such transport would have to take place on time scales comparable to or shorter than the ^{7}Be half-life of 53 days.

In the following we will show that well-established physical processes may in fact be sufficient to explain the findings reported above. Our explanation is premised on the hypothesis that ^{7}Be atoms at very high altitudes in the atmosphere exist predominantly in the form of free atoms.

Background

At approximately 100 km altitude, a transition known as the turbopause separates the turbulent lower atmosphere (the homosphere) from the remainder of the atmosphere (the heterosphere) in which molecular diffusion dominates (Banks and Kockarts 1973). Below the turbopause, all permanent gaseous constituents are found in nearly constant proportions, and the concentration of each decays exponentially with a common scale height which depends on the mean molecular mass of the mixture of gases. Above the turbopause, on the other hand, each constituent's vertical profile is determined individually by a balance between molecular diffusive forces and gravity, and this balance does not depend on the concentrations of other species. The scale height is thus different for each component and, in particular, varies in inverse proportion to the molecular mass. A well-known result is that light elements such as He and H are observed to constitute an increasingly large fraction of the total atmospheric mass as one progresses to higher altitudes.

A similar fractionation would be expected for the relatively light isotope ^{7}Be, provided only that it is not chemically bonded to another element which would greatly reduce the difference between its molecular mass and the mean molecular mass of the other atmospheric gases. This provision runs somewhat counter to the usual assumptions concerning the chemical behavior of cosmogenic Be in the atmosphere, as these atoms normally oxidize rapidly to BeO following their creation and then attach to nearby aerosol particles. While the process described clearly prevails in the stratosphere, where the bulk of cosmogenic ^{7}Be originates and where there are plenty of ambient aerosol particles, the intense ionizing radiation and long molecular mean free path in the thermosphere may dictate an equilibrium condition which favors free atomic Be.

The question addressed here, therefore, is simply whether the equilibrium scale height of unbonded atoms with mass 7 is large enough relative to that of the ambient air, and whether upward diffusion through and above the turbopause is rapid enough, to explain most or all of the observed enrichment of ^{7}Be at 310 km.

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Diffusion Model

Calculation of the steady state vertical profile of a conservative trace substance in the heterosphere is straightforward and follows equations given by Banks and Kockarts (1973). For a cosmogenic radionuclide like ^{7}Be, such calculations are complicated somewhat by the need to consider, in addition to gravitational equilibrium, the balance between production and decay and hence the speed with which ^{7}Be atoms can diffuse from relative sources to relative sinks. The applicable governing equations are then

$$\lambda n + \frac{\partial}{\partial z}(nw) = P \tag{1}$$

$$w = -D\left[\frac{\partial \ln n}{\partial z} + \frac{m_1 g}{RT} + (1+\alpha_T)\frac{\partial \ln T}{\partial z}\right] \tag{2}$$

where $n(z)$ is the number of atoms of ^{7}Be per unit volume at an altitude z, $T(z)$ is the absolute temperature, $g(z)$ is the gravitational acceleration, m_1 is the atomic mass of ^{7}Be, R is the molar gas constant, and α_T is the thermal diffusion factor, which we have taken to be -0.38. The production term P in (1) is the rate at which ^{7}Be is created in a unit volume of air. Accordingly, this rate is given by $P = SY\rho$, where ρ is the air density, and we have taken the cosmic ray 'star' production rate S to be 1×10^{-2} g^{-1}s^{-1} and the ^{7}Be yield Y to be 0.045 atoms per star (O'Brien 1979). The decay rate λ is simply ln(2) divided by the half-life. The diffusion coefficient for ^{7}Be in air is given by an expression of the form $D = AT^s/n_a$ (Banks and Kockarts 1973), where n_a is the ambient concentration of air molecules and we have taken A and s to be 1.5×10^{17} and 0.75, respectively. Although no data are available on the diffusion of atomic Be in air, these values are rough estimates based on the examples of monoatomic gases such as He and Ar and taking into account the dependence of D on mass.

Using standard methods, the time-independent diffusion equation (1) was discretized and numerically solved for $n(z)$ for two model profiles of temperature and atmospheric molecular mass (Figure 1) adapted from the tables of Banks and Kockarts (1973). The first assumes a 750 K thermopause and is a reasonable model for the upper atmosphere during periods of low solar activity. The second model assumes a 2000 K thermopause and is more typical of a period of very high solar activity, such as the one which led to the premature decay of the LDEF orbit. The decreasing mean molecular mass with altitude in both cases (Figure 1b) reflects not only the gravitational fractionation of atmospheric constituents by mass but also the increasing tendency for compounds such as molecular oxygen and nitrogen to be dissociated into monoatomic forms at higher altitudes.

As a lower boundary condition, the concentration of ^{7}Be at the turbopause was held constant at a value equal to its expected equilibrium concentration (P/λ) in the well-mixed region below the turbopause. A zero flux upper boundary condition was enforced by requiring the value of n at the top of the model domain (in this case 3000 km) to satisfy $w = 0$. This boundary condition expresses the fact that cosmic ray production and radioactive decay are too slow, relative to the molecular diffusion speed, to significantly distort the shape of the equilibrium concentration profile at that altitude. While there is undoubtedly a slow loss of ^{7}Be atoms whose velocities permit them to escape the gravitational field, this flux is negligible for the purposes of our calculations.

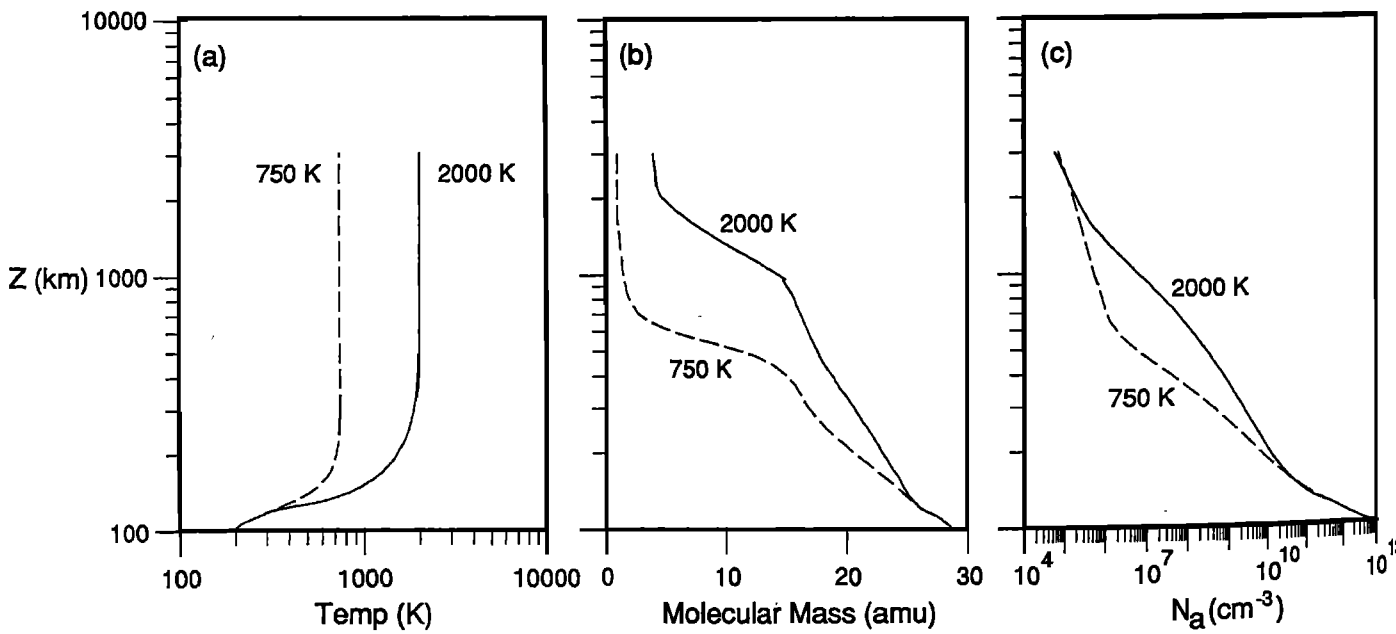

Fig. 1. Model atmospheres used in diffusion calculations. Dashed curves correspond to atmosphere with 750 K thermopause (inactive sun); solid curves correspond to 2000 K thermopause temperature (active sun): (a) temperature (b) mean molecular mass (c) molecular concentration.

Results

Results of the ^{7}Be profile calculations are shown in Figure 2. At the LDEF's recovery altitude of 310 km, predicted concentrations are 1.2×10^{-8} atoms per cm^3 and 2.9×10^{-8} atoms per cm^3, respectively, for the inactive sun and active sun thermal profiles. *The latter value is within a factor of 4 of the concentration inferred from the LDEF measurements and thus would account for most of the ^{7}Be enrichment reported by Fishman et al.*

Interestingly, setting both λ and P to zero above the lower boundary of the model domain had only a modest effect on the computed profiles. This suggests that the concentration of ^{7}Be in the heterosphere is governed largely by the concentration at the turbopause and that diffusion from the turbopause to higher altitudes takes place on time scales which are indeed rather short compared with the half-life. It may therefore be inferred that the profiles computed above are relatively insensitive to the exact specification of the diffusion coefficient D.

They are, however, somewhat more sensitive to certain other assumptions, especially the placement of the turbopause. For example, increasing the turbopause height z_T in the model from 100 km to 120 km reduces the predicted ^{7}Be concentration at 310 km by a factor of 10. On the other hand, a minor reduction in z_T could easily bring the active sun profile into exact agreement with the observed concentration. Because the transition from the turbulence-dominated to diffusion-dominated regimes is not discontinuous, but takes place over an interval of perhaps 20–30 km, beginning near 90 km, our choice of 100 km as the effective height of a discrete turbopause in our simple model is arbitrary, though not unreasonable.

Other parameters in the model are also subject to some uncertainty, including the high-altitude star production rate, which ranges from less than 0.5×10^{-2} g^{-1} s^{-1} near the equator to at least 5.0×10^{-2} g^{-1} s^{-1} near the poles for typical levels of solar activity (O'Brien 1979). The solar cycle introduces another factor of two variability in the production rate. Since horizontal diffusion in the thermosphere is very rapid, we have taken the view that a global average star production rate estimated at $\sim1.0 \times 10^{-2}$ g^{-1} s^{-1} is more appropriate for our calculations than would be the somewhat lower production rate ($\sim0.3 \times 10^{-2}$ g^{-1} s^{-1}) prevailing at the low latitudes of the LDEF orbit.

Conclusions

To well within the uncertainties of our atmospheric diffusion calculations, the LDEF measurements reported by Fishman et al. (1991) are consistent with the hypothesis that cosmogenic Be exists mainly as free atoms in the upper atmosphere. The next step should be to establish whether any independent theoretical or empirical data relevant to the chemical behavior of Be in the upper atmosphere exist and, if so, whether this evidence is consistent with our proposed explanation.

We wish to emphasize that the one-dimensional diffusion model employed here is a relatively crude approximation to reality. A two-dimensional diffusion model would be required to properly account for meridional diffusion and for latitudinal variations in production rate, and a much more complex dynamic model might be necessary

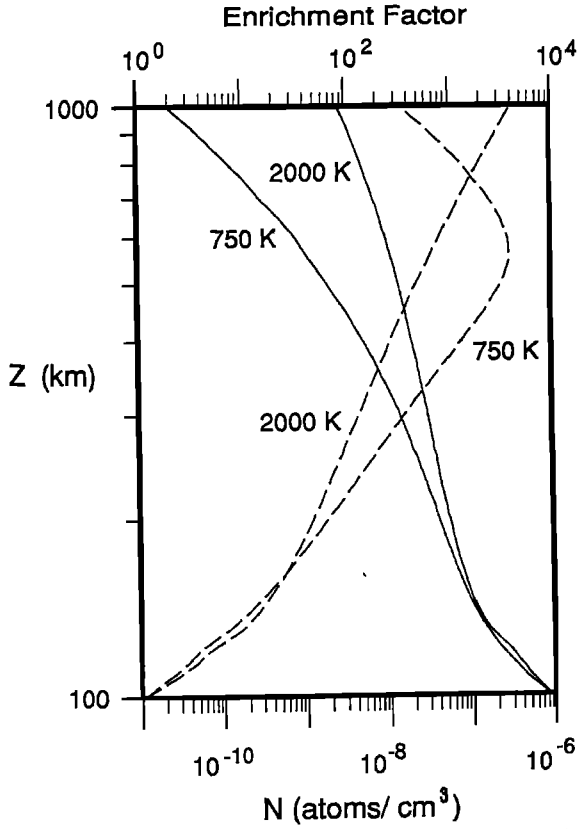

Fig. 2. Concentrations (solid curves) and enrichment factors relative to turbopause (dashed curves) of ^{7}Be computed for the two model atmospheres.

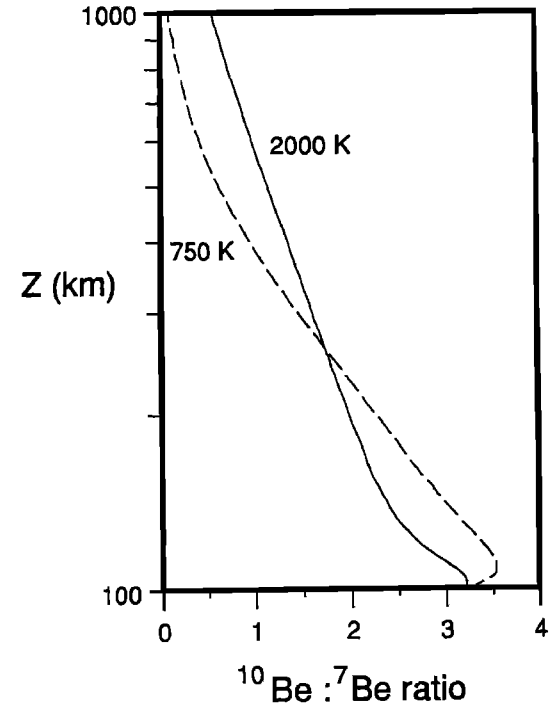

Fig. 3. Predicted ratio of ^{10}Be concentration to ^{7}Be concentration for the two model atmospheres.

to investigate the effect of diurnal solar heating of the upper atmosphere on cosmogenic radionuclide distributions. We have also not considered the implications of any ionization of the ^{7}Be atoms in the upper atmosphere, though it is possible that many or most of them are ionized. While it is evidently not *necessary* to consider two or three dimensional geometry, or to invoke ion-specific transport mechanisms, in order to account for the bulk of the observed ^{7}Be enrichment, we cannot rule out the possibility that a more comprehensive physical model might give significantly different results.

An Opportunity for Further Verification

At the time of this writing, samples from the LDEF were being prepared for Accelerator Mass Spectrometry (AMS) measurements of ^{10}Be (B.A. Harmon, personal communication). This isotope is produced in the atmosphere in exactly the same way as ^{7}Be, though with a yield per star which is smaller by about a factor of two (O'Brien 1979). The half-life of ^{10}Be is 1.5×10^{6} yr, however, so that measurements on the LDEF will effectively represent total accumulations over the entire 5.8 yr mission. Most of this accumulation will have occurred near 480 km altitude, rather than at the 310 km altitude from which the satellite was recovered.

The measured ^{10}Be accumulations, when they become available, will offer another opportunity for qualitatively testing the physical model proposed above. If the simple upward diffusion of unattached Be atoms is in fact the principal enrichment mechanism at work, then we expect ^{10}Be:^{7}Be ratio profiles similar to those in Figure 3, based on an assumed effective atmospheric removal rate (inverse residence time) $\lambda' = 1.0\ yr^{-1}$. While this value is known to be approximately correct for Be isotopes in the stratosphere (Reiter 1975), residence times at higher altitudes are uncertain. However, a longer estimated residence time than we have assumed here can be accommodated by allowing a directly proportional increase in the predicted ^{10}Be concentration at all levels.

Between 310 km and 480 km, expected ^{10}Be concentrations are within a factor of two of, but generally larger than, those computed for ^{7}Be. At 310 km and for the active sun profile, the ratio is 1.6. As a minimum, therefore, we expect about 8×10^{5} atoms cm^{-2} to have accumulated during the 77 days prior to LDEF recovery (the effective period represented by the observed ^{7}Be accumulations). For the remaining 5.5 yr of the mission, the ^{10}Be concentration encountered may have ranged over an order of magnitude, namely from about 1.6×10^{-9} atoms cm^{3} to 1.8×10^{-8} atoms cm^{3}. Based on the very limited orbital and atmospheric data available to us at the time of these calculations, we estimate that the total LDEF accumulation of ^{10}Be should be found to fall roughly between 2×10^{6} and 6×10^{6} atoms cm^{-2}, with a slight possibility of values as high as 10×10^{6} atoms cm^{-2}. As noted above, these values should be multiplied by the best prevailing estimate of the upper atmospheric residence time (in years) for cosmogenic Be.

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