

MODEL SIMULATIONS OF CARBON ISOTOPE VARIABILITY IN THE MANTLE.

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The $\delta^{13}\text{C}$ sampling frequency distributions of kimberlitic diamonds from southern Africa and Russia are indistinguishable. The summary distribution for the two continents may have thus broader significance and has been used to test geochemical models that can be proposed to explain carbon isotope variations in the mantle. The distribution has a weighted mean of -7 ‰, a major mode at -5.5 ‰, is highly skewed towards negative $\delta^{13}\text{C}$ values, and may have a minor second mode between -15 and -19 ‰. Hence it differs significantly from the normal $\delta^{13}\text{C}$ distributions of carbonatite and kimberlite carbonates whose means and modes all coincide at -5.5 ‰.

Chemical fractionation models which allow for changes in pressure, temperature, oxygen fugacity, and depletion of a carbon reservoir can explain systematic variations of $\delta^{13}\text{C}$ of a few ‰. However, they are not capable to account for the very high negative skewness of the diamond $\delta^{13}\text{C}$ distribution.

Subduction of sedimentary, organic carbon has been considered as a cause for the existence of ^{13}C depleted diamonds. The consequences of such a process have been examined with the aid of models simulating isotope exchange and mixing. Because at this time no evidence has been presented for an isotopic composition difference between the crustal and mantle carbon reservoir the mean isotopic composition of the subducted carbon is assumed to be -7 ‰. The extent to which the carbon isotopic composition of the sedimentary end members (organic carbon $\delta^{13}\text{C} = -25$ ‰, limestones $\delta^{13}\text{C} = 0$ ‰) are preserved or altered through isotope exchange and homogenization during subduction is unknown. It has been assumed in the model that the retention of the endmember isotopic compositions is as probable as the establishment of any intermediate isotopic composition through isotopic exchange. The only restriction assumed is that the weighted mean isotopic composition of the carbon compounds remained -7 ‰. The volume over which the carbon isotopic composition is homogenized during subduction is also important for the resulting $\delta^{13}\text{C}$ sampling distribution. If the volume is very small it is more likely that the extreme sedimentary carbon isotopic compositions are retained in the mantle, if the volume is very large the weighted mean value of -7 ‰ must be attained. The model was set up so that varying degrees of homogenization over a variable range of volumes could be examined.

Over 100 different combinations of exchange and homogenization scales have been studied. Emphasis was placed on an examination of those models which would most closely simulate the relative abundance of the low $\delta^{13}\text{C}$ diamonds which have been proposed as indicators of subducted carbon. The results have been compared with the summary diamond $\delta^{13}\text{C}$ distribution.

Those model $\delta^{13}\text{C}$ distributions which have the highest abundance of low $\delta^{13}\text{C}$ diamonds and at the same time an abundance at the major mode which matches that of the observed diamond distribution, predict lower relative abundances of ^{13}C depleted diamonds than actually observed. Models which predict correctly the observed relative abundance of low $\delta^{13}\text{C}$ diamonds predict, however, also that there should be 50% fewer diamonds with the isotopic composition of the major mode, and a very much higher abundance of diamonds of isotopic compositions between 0 and -3 ‰ than is actually found.

In all cases the mode of the model distribution occurs at -7 ‰, rather than at -5.5 ‰. The success to match more than one feature of the model distribution with the observed

diamond distribution has been limited. The model computations suggest that if the low $\delta^{13}\text{C}$ diamonds were to be the result of subduction of reduced sedimentary carbon that this carbon would be subducted in preference to carbonate carbon. Consequently a difference in the carbon isotopic composition between crustal and mantle carbon reservoirs would have to be postulated.

Models simulating the effect of the removal of ^{13}C enriched CO_2 from the mantle as a result of degassing have also been constructed. The fractionation factor for such a process can be deduced from the skewness of the diamond $\delta^{13}\text{C}$ distribution. The estimate ($\alpha = 1.0038$) is much larger than expected on the basis of known isotope effects and recent experiments. A large number of model frequency distributions were investigated, using different assumptions for the initial reservoir isotopic composition, reservoir variability, and fractionation factor. It was found to be impossible to match all major features of any of the model distributions with those of the observed diamond $\delta^{13}\text{C}$ distribution.

The $\delta^{13}\text{C}$ variability of diamonds has probably multiple causes which may include: 1. chemical isotope effects related to temperature, pressure, and oxygen fugacity; 2. reservoir depletion effects; 3. existence of carbon isotope reservoirs of differing isotopic composition which could remain from accretion or be introduced by subduction. It is unlikely that any one of these causes alone can produce the observed diamond carbon isotopic composition distribution. The relative importance of the various causes remains to be established.

If the limited data we have accumulated to date on the ^{13}C depletion of asthenospheric South African diamonds would reflect a general mantle feature, one could propose that the mantle is for several reasons heterogeneous in $\delta^{13}\text{C}$, and that with increasing depth the relative abundance of low $\delta^{13}\text{C}$ carbon increases. This concept could explain: 1. the common mode in $\delta^{13}\text{C}$ of carbonatite and diamond carbon isotopic compositions; 2. the significant difference in mean $\delta^{13}\text{C}$ between carbonatites (-5.5) and diamonds (-7 ‰); 3. a higher relative abundance of ^{13}C depleted asthenospheric diamonds; 4. the common features of diamond and meteorite $\delta^{13}\text{C}$ distribution; and 5. the apparent difference in the mean carbon isotopic composition of meteorite and mantle carbon. The proof of this type carbon isotope distribution within the mantle - which would have to be established through further work - would significantly alter our concepts of the geochemical cycle of carbon.