THE URANIUM CONTENT OF THE LAVAS OF LASSEN VOLCANIC NATIONAL PARK, CALIFORNIA

by

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Our work at Lassen Volcano is part of a program to investigate the volcanic phase of the geochemistry of uranium. This program is particularly concerned with the following points: the average abundance of uranium in volcanics; the role, if any, of radioactivity in the thermal mechanisms of vulcanism; the possible dispersal of uranium in the gases and fine solids produced by volcanoes; and finally, the selective leaching of uranium from volcanic tuffs.

Lassen Volcano was selected for early study because of its accessibility; because of the large amount of chemical and petrographic work that had been done by earlier researchers, particularly Professor Howell Williams; and because of presence of fluorite. One of our primary aims, then, was to see how uranium fitted into the chemical and petrographic patterns worked out by these earlier researchers.

Samples were collected in late June, 1952, by the authors. Close attention was paid to the lava types mapped and petrographically described by Williams. The degree of weathering and the megascopic homogeneity were also noted. A total of forty-one samples were collected, nineteen of which will be discussed here. Slide One shows the location of Lassen Volcanic National Park in the state of California and the localities within the park from which our samples were taken. Lassen Peak and Cinder Cone are also indicated.

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The samples were analyzed for uranium by fluorimetric techniques modified from those described in the reports of the Trace Elements Laboratory of the United States Geological Survey. The uranium present is usually in the range from 0.1 to 10.0 parts per million by weight. After over two years of development and several thousand analyses in our laboratories, our average precision in the determination of uranium in volcanics is about $\pm 8\%$ of the uranium present. By use of spikes at various steps in the procedure, we estimate our average accuracy with volcanics to be about $\pm 8\%$ of the uranium present. Fifty-seven uranium determinations were made on the nineteen samples under discussion.

The relative alpha particle activity of each of the samples was measured by a scintillation counter technique. The counting was continued long enough to give a statistical precision of at least $\pm 20\%$ at the 90% confidence level. A total of forty-five counts were made on the nineteen samples.

The potassium and sodium contents of the samples were determined with a flame photometric technique. The average precision and accuracy of the alkali analyses are within a few per cent of the amount present. Standard samples from the United States Bureau of Standards were used as standards and checks on the technique. The alkalis were determined so that we could correlate the uranium analyses with the variation diagram for the Lassen suite. A simplified variation diagram for Lassen (after Williams) is shown in Slide Two. Note that soda passes through a maximum, while potash increases consistently with increasing silica.

A plot of uranium in weight parts per million versus soda in weight per cent is shown in Slide Three. The soda values are plotted as points because the analytical uncertainty is so low. The uranium values are shown as lines to express an analytical

uncertainty of about $\pm 10\%$ of the uranium present. The analytical uncertainty with uranium is probably somewhat less than $\pm 10\%$. There is not a very good correlation between soda and uranium. One would not expect a close correlation unless the soda to uranium ratios in the crystalline and non-crystalline phases remained constant.

A plot of uranium metal in weight parts per million versus potash in weight per cent is shown in Slide Four. The correlation here is quite definite. The slight deviations from the main trend are probably due as much to analytical error as to real variations in the samples. This curve shows that the net effect of the differentiation of the Lassen suite was to concentrate both uranium and potash at about the same rate in the more siliceous lavas. The slope of the trend lines shown here is about 0.75, indicating that the uranium was concentrated about one-third faster than the potash. This curve can be interpreted most conveniently in the familiar terms of fractional crystallization with the exclusion of potash and uranium from the crystalline phases that formed and the separation of the crystalline and non-crystalline phases as differentiation proceeded. It is difficult to propose any other interpretation, because the foremost property that uranium and potassium have in common in this situation is that their ionic space requirements and coordination numbers with respect to oxygen do not permit them to substitute easily for other elements in the crystalline phases of early differentiation. This is less true for potassium than for uranium, since uranium concentrates faster. Some of the potassium was certainly taken into such minerals as biotite. The percentage of uranium that separated into any gas phase must be several times smaller than the small percentage of potassium that separated into the crystalline phase, otherwise the slope of this curve would be greater. These data indicate that the radioactivity of potassium-forty and of uranium are both concentrated more and more as the high silica members

of the Lassen suite are approached. The only other abundant, long-lived radioactive substance to be considered is thorium.

The thorium series together with the uranium series is responsible for the alpha particle activity of these samples. The alpha particle activity in relative alpha counts per hour is plotted against uranium in weight parts per million in Slide Five. Both the alpha counts per hour and the uranium in parts per million are plotted as lines to express the analytical uncertainty. There is a fair correlation here. The major deviations from the main trend may be due to several things: the deviations may be due to analytical error; they may be real; the deviations may be due to radioactive disequilibrium, particularly if uranium has been selective leached from the samples in the last few thousand years. Leaching of uranium would displace a point mostly to the left and a little downward. Selective leaching is also indicated by the fact that the material from the 1915 mud flow is one of the major deviations; another of the major deviations is the Mansanita dacite, which is one of the oldest dacite flows and the outcrop we sampled was partially weathered. Assuming radioactive equilibrium in the other samples, any significant partition of thorium and uranium between the crystalline and non-crystalline phases should be reflected in this curve. The fact that the relative alpha counts per hour to uranium ratio remains fairly constant throughout the entire range, is evidence that the thorium to uranium ratio also remains fairly constant. We are now trying to develop independent analytical methods for determining the thorium to uranium ratios in these samples. This curve indicates that both the thorium and uranium were being concentrated in the non-crystalline phases or residual magma during differentiation.

The same general trend is shown in a plot of potash in weight per cent versus relative alpha counts per hour, as in Slide Six. Here again the major deviations from the main trend are samples like the 1915 mud flow, the Mansanita dacite, and some rather unconsolidated pumice. Indeed, the 1915 mud flow does not qualify as a pure and simple volcanic and it is used here as an example of material that has been somewhat leached. These data indicate that the thorium to uranium ratio as determined by relative alpha activity did not change much during differentiation.

In conclusion, the uranium content of the Lassen volcanics varies by a factor of about four from 0.8 parts per million to 3.6 parts per million by weight. The thorium probably varies also by about a factor of four. The uranium versus potash curve is essentially linear for the Lassen suite. However, differentiation at Lassen is not very extreme, and one would expect this linearity to break down in situations where potassium rich phases, such as orthoclase, began to form. We hope to trace uranium through a more extreme differentiation. At Lassen, all major sources of radioactivity, potassium-forty, uranium, and thorium, were three to four times more abundant in the most silicious lava studied than they were in the least siliceous lave studied.

6

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Slide 1

Variation diagram of the Lassen Peak region
(after Williams)

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Slide 2

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Slide 6