

Stable carbon and hydrogen isotope ratios in atmospheric methane over Siberia

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The stable hydrogen isotope ratio (D/H) in methane (CH₄) would be a useful tracer for both identifying the source of atmospheric CH₄ and constraining the atmospheric CH₄ budget as well as the stable carbon isotope ratio (¹³C/¹²C) and the radiocarbon content (¹⁴C). However, the measurement of the D/H ratio in atmospheric CH₄ and its sources has so far been less developed, compared to that of ¹³C/¹²C ratio. This is due to the difficulties in the preparation of samples that are required before analysis by isotope ratio mass spectrometry. Recently a gas chromatography / high-temperature conversion / isotope ratio mass spectrometry (GC/TC/IRMS) technique, which allows high-precision measurement of the D/H ratio of atmospheric methane using only a small sample, has been established.

Only a few isotopic measurements have been attempted in the Siberian region, in spite of its important source region for global atmospheric CH₄. We applied the GC/TC/IRMS technique to CH₄ in tropospheric air samples over Siberia collected at the air flight (Surgut; altitude, 500m, 7030m and 7100m) and ground (Plotnikovo; 8m) experiments in the summer of 1994.

The atmospheric CH₄ in air samples over wetlands (Plotnikovo) were depleted in D compared to the atmospheric background. From the results observed, the D/H ratio of CH₄ emitted from wetlands into the atmosphere were estimated to be ~-360‰. The corresponding ¹³C/¹²C ratio of CH₄ also showed the typical value of wetlands. The D/H ratio of atmospheric CH₄ in air samples over oil wells and pipelines as well as wetlands (Surgut) were also depleted in D compared to the atmospheric background, showing a correlation with the CH₄ concentration. From the relationship between the CH₄ concentration and D/H ratio, CH₄ added to the background CH₄ was as close as single source and was isotopically light value, indicating the wetlands source. However, the corresponding ¹³C/¹²C ratio of CH₄ was higher than the atmospheric background, exhibiting a significant contribution from sources of oil wells and pipelines. We will discuss the explanation for the disagreement between ¹³C/¹²C ratio and D/H ratio observed.

Comparison between (U-Th)/He and K/Ar ages from layered young rhyolites in Kyushu, Japan.

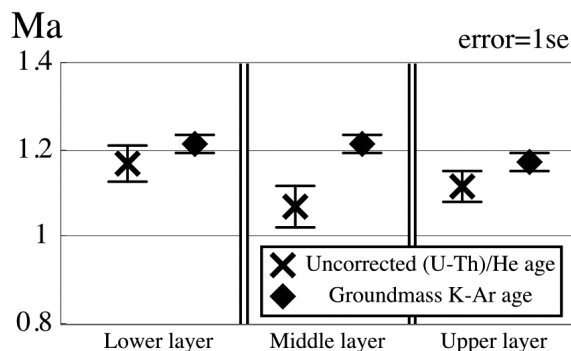
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The (U-Th)/He dating method is suitable for measuring young ages due to the rapid decay rate and the low blank He (only 5 ppm in atmosphere). However, it will need some suitable corrections for measuring ages less than about 1 Ma because of the effects from the disequilibrium of ²³⁸U decay series. One of these corrections is offered by Farley et al. (2002), nevertheless there is few studies so far that compared young (U-Th)/He ages with other reliable isotopic ages. Therefore in this study, we report new (U-Th)/He ages of zircons from three layers of acidic plateau lava, for which K/Ar ages were determined previously. Secondly we compared them and estimated the suitability of the disequilibrium correction on young (U-Th)/He age with zircons.

We analyzed zircon crystals separated from Yamakogawa rhyolite on Hohi volcanic zone, middle of Kyushu Island, Japan. This unit comprises of lavas or strong welded tuffs, containing three major layers and is thought that it was rapidly cooled after eruption and not reheated. Additionally it is thought that all layers erupted continuously from observation of the outcrops.



The diagram shows a plot of uncorrected or apparent zircon (U-Th)/He and groundmass K-Ar ages. It shows that all of zircon ages are slightly younger than K-Ar ages. This trend is qualitatively consistent with the possible effects of ²³⁸U decay series disequilibrium. However, we have no idea for the consistency quantitatively. Therefore we will measure D230's of Yamakogawa rhyolite and evaluate the adequateness of the correction until August.

Reference

Farley, K.A., Kohn, B.P. and Pillans, B., (2002). EPSL., **201**, 117-125.