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Novel Chemical Methodology for Identifying Origin of Archeological Bitumen: Chasing the Trade Routes along the Japanese Archipelago and Sakhalin Island in Prehistory

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We invented a novel methodology for identifying origin of archaeological bitumen by use of field-ionization mass spectrometry (FI-MS). In the FI-MS method, fragmentation of molecular ions is minimal and there is a unit charge on each molecule. Thus, the observed mass spectra directly reflect the distribution of the molecular weights of the alkane components in bitumen. The distribution could be a molecular criterion for characterizing the bitumen sources from which each bitumen sample was derived.
Actually, we decomposed the FI-MS spectra by Z-numbers into several components: the Z-number refers to z in the formula CnH2n+z and 2n-z is equivalent to the deficit number of H atoms when compared to the corresponding saturated hydrocarbon, which, in turn, is correlated to the ring number in alkanes. The integrated intensities of the component spectra corresponding to the Z-number were compared to each other. The difference in the observed spectra is reflected by the difference in concentration of alkane groups with different Z-number. In this way, the intensities data of the component spectra were used as indexes to search for the origin of the bitumen.
FI-MS measurements were performed on 67 samples from five different bitumen sources and 41 bitumen samples excavated from archaeological sites in Honshu and Hokkaido, the largest and the second largest island in Japan, and Sakhalin island in Russia. By use of the spectral intensities of the seven alkane components in each sample, multiple discriminant analysis was employed for the data of raw bitumen samples and excavated samples from archaeological sites. The GC-MS chromatograms obtained from the archaeological samples from the Honshu area were all consistent with the results obtained by multivariate analysis, and thus the validity of the newly developed Z-number analysis was confirmed.
As for the archaeological bitumen samples in Hokkaido, It was found that bitumen from Niigata, one of the main sources in Honshu, spread to the north in 2000 BC. It reached a small island near the north end of Hokkaido. Bitumen from Sakhalin reached the central lowland in Hokkaido, but it did not go into Honshu. Bitumen from Akita, another main source in Honshu, was predominated in the northeastern part of Honshu and the Oshima peninsula located at the southeastern end of Hokkaido. The story is consistent with a strong cultural tie between the Oshima peninsula and the northern Honshu throughout the Jomon period, the long lasting cultural period in Japanese prehistory. The long trade route along the coast of the Sea of Japan is being argued due to the recent archaeological findings obtained by excavations. Our results will shed more light on the geopolitical situation in the Jomon period of the area.