

The evaluation of the 100 best natural water sources in Heisei period by trace elements and application for the origin estimation of foods

*Naoya Yanagisawa¹, Nami Sasamoto¹, Yoshinari Abe¹, IZUMI NAKAI¹

1.Tokyo University of Science Graduate School of Chemical Sciences and Technology

Recently, fraudulent claiming of origin and safety control of foods have become a social problem in Japan, there is a growing need for a traceability of food's origin and manufacturing process. We have previously developed estimation methods for origin of grains, vegetables and beverages using inorganic chemical composition and isotope ratios. In the present study, we newly focused on Japanese natural water, especially water designated as "the best 100 natural water sources in Heisei period". The water is an essential resource having commitment in our life with wide range of application. Using trace elements solved in the water as a tracer, we examined regional characterization of waters with a view to application for origin estimation of foods.

Natural water samples were collected from 20 points (5 rivers 15 springs) over Japan using a previously-cleaned polyethylene container and have been stored in a cold dark place (~4°C). Deposits in water samples were removed by a filtration using a membrane filter (pore size 0.45 µm) prior to the analysis. While light elements (Na, Mg, K, Ca, Si) were analyzed by an inductively coupled plasma emission spectrometer (SPS3520UV), other trace elements were analyzed using a quadrupole inductively coupled plasma mass spectrometer (Agilent 7500c). ¹¹⁵In was added to each water samples as an internal standard element, concentrations of more than 20 elements solved in the waters were quantified by a calibration method.

At first we considered possible sources of trace elements in water samples detected by our analyses. Concentrations of Li was characteristically high in river water samples from the Saitama and Yamanashi Prefectures. This is because the riverhead of these rivers runs over a granite layer containing Li. On the other hand, high concentration of V is characteristic in water samples from the Yamanashi and Shizuoka Prefectures. The *Natsukari Spring* in the Yamanashi Prefecture and the *Wakudama Pond* in the Shizuoka Prefecture are spring waters in the region consist of V-rich basalt rock. Similarly, the *Genbei River* is a river belonging to the *Sagami Riverine* system in which basalt rocks are widely distributed. Concentrations of Li and V of other water samples were not so high because geology of upper stream and waterbearing stratum of these waters are not granitic or basaltic. In the case of Cu, waters sampled from the Saitama and Gunma Prefectures showed high concentration. The *Tokura Spring* and the *Hotaka Spring* in the Gunma Prefecture are located around the *Ashio Copper Mine* known as mineral poison. Similarly, there is the *Chichibu Mine* with a skarn deposit near the *Bisyamon Spring* and the *Bukousan Spring* in the Saitama Prefecture. We could find good agreement between distributions of these three elements of water samples and concentrations in river sediments reported as the Geochemical Map by AIST. It is thus expected that these trace elements will be useful parameters for regional characterization of the natural water. Concentrated Al, Mn, Zn and rare earth elements were detected from a water sampled from the *Detsubo Spring* in the Akita Prefecture. The pH value of this water was 4.4, although averages of spring waters and river waters analyzed in this study are 7.2 and 7.8, respectively. Previous studies pointed out that the pH value of the spring water and the groundwater could decrease with an increase in organic acids by microbial activity under the specific environment such as a swamp. As the result, Al and rare earth elements would dissolve into the water from soil under the acid condition. In conclusion, it was revealed that trace element composition of the spring and river waters obviously reflects their geological background. In the future, we aim to region characterization with a goal of application for the origin estimation of foods.

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