

## Spike-like increases of He/Ar and CH<sub>4</sub>/Ar ratios of groundwater gas bubbles possibly correlating with crustal strain changes

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**Introduction:** Geochemical earthquake prediction concerns with intimate correlations between seismic events and anomalous changes of chemical or isotopic compositions of gases associated with groundwater. The geochemistry laboratory, Nagoya Univ. has reported preseismic anomalies of gas bubbles issuing from mineral spring waters by automated gas monitoring. Here is reported continual gas monitoring results at a 500m borehole-well of Haruno Observation station, RSVD-NU<sup>^</sup> from Dec. 8, 2006 to Mar. 29, 2007. Gas bubbles issuing from the groundwater of the well have been analyzed with one-hour interval by a newly designed automated gas monitoring system utilizing a micro-gas chromatograph (GC).

**Experimental methods: 1) Monitoring site** Haruno Station is located at Haruno-cho, Hamamatsu city, Shizuoka Pref. Three boreholes 300m, 400m, and 500m deep are used for crustal strain observations. The bedrock of borehole-wells is the sandstone-shale alternation belonging to Shimanto Belt. A small quantity of groundwater overflows from only the 500m deep borehole-well, and a small amount of gas bubbles are issuing from it.

**2) A new automated gas monitoring system** The GC with a micro-thermal conductivity detector and a capillary column (0.32 mm ID, 10m long) packed with Molecular Sieve 5A is used in the new system. The volume concentrations of He, H<sub>2</sub>, Ar, N<sub>2</sub>, and CH<sub>4</sub> can be determined precisely and simultaneously, because oxygen is used as the carrier gas. Gas bubbles collected with a plastic funnel placed upside down in the well water are introduced into GC system through Teflon and stainless tubes. The sample gas is pumped into the GC, and is analyzed every one hour, but the standard gas analysis is made every ten hours for calibration.

**Results:** The chemical compositions of gas bubbles are fluctuating considerably with time. In particular, spike-like increases of gas concentration ratios (values normalized by Ar) were repeatedly observed in late Dec., 2006, late Jan. and middle Feb., 2007. For example, the CH<sub>4</sub>/Ar ratio abruptly increased from 25 to 250 in late Dec., 2006. The He/Ar, H<sub>2</sub>/Ar, and N<sub>2</sub>/Ar ratios also abruptly increased at the same time. The crustal strain records at Haruno show that the cubic dilatation was exhibiting a steadily increasing trend until the end of Jan., 2007, but that the trend terminated after that time. The 'breaking' point of the cubic dilatation corresponds to the large abrupt increases of gas ratios. In addition, according to NIED<sup>^</sup>, the non-volcanic deep tremor activity in Tokai area also indicated a major peak in two or three days after the large spike-like gas anomalies (Fig. 1).

**Discussion and conclusions:** A good positive linear correlation between N<sub>2</sub>/Ar and CH<sub>4</sub>/Ar ratios indicates a mixing line of two end-members ( $N_2/Ar = 1.37 \times CH_4/Ar + 85.1$ ). Because the mixing line has the N<sub>2</sub>/Ar-intercept in agreement with the atmospheric N<sub>2</sub>/Ar ratio, one of the end-members is the atmospheric air, and the other is to be the subsurface gas. Assuming the end-member value of N<sub>2</sub>/Ar to be 450, we can estimate the chemical composition of the subsurface gas. The calculated results are N<sub>2</sub> (63 %), CH<sub>4</sub> (37 %), Ar (0.14 %), He (24 ppm), and H<sub>2</sub> (41 ppm), representing the crustal gas associated with the sedimentary rocks of the Shimanto Belt enriched in N<sub>2</sub> and CH<sub>4</sub>. We interpret that the observed abrupt increases of gas ratios are the unsteady transports of subsurface gas from underground to the surface. Our observation results in late January and early February, 2007 (Fig. 1) may suggest that there is an intimate correlation among the unsteady subsurface gas transport to the surface, the crustal strain changes, and the peak activity of non-volcanic deep tremors.

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Fig. 1

Time series of the tremor activities in the Tokai area, He/Ar ratio and cubic dilatation at Haruno Observatory. Arrows below the cubic dilatation indicate trend changes of cubic dilatation, which is the "breaking" point.

