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Rapid isotope analysis of the deuterium/hydrogen ratio of hydrogen in atmosphere at several hundreds ppm levels

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Molecular hydrogen (H_2) is the second most abundant trace gas in the atmosphere (after methane, CH_4), with a mixing ratio of ca. 500 ppb (e.g., Novelli et al., 1999). In the near future, there is a possibility for a change in a balance of anthropogenic emissions of H_2 and an increase overall because it is considered to be an important future alternative to fossil fuels. The increase in atmospheric H2 levels leads to an important change in air quality, such as an increase of the lifetimes of other important trace gases such as CH_4 and carbon monoxide (CO) in the troposphere or a change in the water budget in the stratosphere. To identify the cause of the increase of the atmospheric H_2 levels and its influence on atmospheric chemistry, it is important to constrain the current budget of atmospheric H_2.

Measurements of stable isotope ratio (i.e., D/H ratio) in atmospheric H_2 and individual H_2 source and sink processes have the potential to help constrain the local, regional and global H_2 budget. However, the measurement of the D/H ratio in atmospheric H_2 and its sources has so far been less developed, because of difficulties in the preparation of samples that is required before analysis by a dual-inlet isotope ratio mass spectrometer (IRMS) used by previous studies (e.g., Ehhalt, 1966; Friedman and Scholz, 1974; Gerst and Quay, 2000).

More recently, an off-line H_2 gas separation from the air matrix followed by introduction to an IRMS (Rahn et al., 2002) and an on-line H_2 gas separation and concentration coupled with a gas chromatograph (GC) - IRMS (Rhee et al., 2004) have been developed. These techniques not only reduced the amount of air required for isotopic analysis of atmospheric H_2 by a order of three compared to the previous studies, but also obviate the need for catalytic conversion of H_2. However, these techniques require the vacuum line system equipped with a cold-trap for concentration of H_2 gas operated at very low temperature (30 - ca.60 Kelvin).

In this study we tried a preliminary development of a simple method that avoids the use of the cold-trap for concentration of H_2 gas. The method used GC-IRMS system (Finnigan Delta plus-XL coupled with HP6890 GC via GC-III interface, ThermoFinnigan). The GC was equipped with a split/splitless injector and capillary column (CARBOXEN^{TM} 1010PLOT, 30m, Supelco). Top part of the capillary column (ca.20 cm) immersed in 77K liquid N_2. Up to 1 mL of air sample is injected into the injector of the GC with splitless mode by a gas-tight syringe. Using this method, the D/H ratio of atmospheric H_2 at several hundreds ppm levels was determined with precision of 2 permil. The method is useful for atmospheric samples with relatively high H_2 concentrations, such as biomass burning source.