博　士（環境科学）　スンモヌ　ルクマン　アヤニー

学位論文題名

Characteristics of seasonal and short-term variation of near-surface water vapor D/H isotope ratio revealed by continuous in-situ measurement

（連続・現場測定から明らかとなった地表近くの水蒸気D/H同位体比の季節および短時間変動特性）

学位論文内容の要旨

Seasonal and short-term variation of stable isotope ratio of atmospheric water vapor (δD) was investigated in surface air in Sapporo, (43.1N, 141.3E), Hokkaido in Japan. The measurement was carried out on a continuous basis and on a high temporal resolution with a Water-Vapor Isotope Analyzer, model DLT-100 in four phases: (I) 28 April 2009 - 25 June 2009; (II) 15 Sep. 2009 - 29 Nov. 2009; (III) 1 April 2010 - 21 May 2010 and (IV) 17 Dec. 2010 - 30 May 2011. The data reported here is for a total period of ten months.

The Water Vapor Isotope Standard Source (WVISS) is used to automatically and comprehensively characterize WVIA's operation in real time. When used together, as we did during the last phase of our measurement, they provide reliable and real-time absolute measurements of δD and $\delta^{18}O$ in ambient water vapor. The data obtained from the WVIA, calibrated automatically and comprehensively by the WVISS, was corrected using two methods based on the fact that δD and $\delta^{18}O$ are dependent on the amount of water vapor mixing ratio. Overall calibration revealed that δD variation less than or equal to 20‰ can be said to be due to instrumental uncertainties while variation greater than this value can be said to be due to natural occurrence associated with weather phenomenon.

Equal day-to-day variability in the hydrogen isotope composition of the atmospheric water vapor is seen both in the cold and warm season. Monthly mean δD shows good correlation with monthly mean air temperature. However, the values of δD before summer

were lighter than those after summer. The correlation coefficient (r^2) between δD and Sea Surface Temperature (SST) of the Sea of Japan was greater than that between δD and air temperature in Sapporo. This result suggests that SST is a better predictor for δD in Sapporo, rather than ambient temperature. An Isotope-Global Spectral Model (IsoGSM) was used in simulating δD for the same period with that of the observation. The seasonal trend of δD was well simulated, except that the IsoGSM overestimates δD by about 20 ‰.

Extreme variation in δD was investigated using three-sigma analysis. Six case studies were identified based on this analysis with the values of δD lighter than -210‰ and with the period of variation ranging from 2 hours to about 15 hours. Results of simulation by using an Isotope-Regional Spectral Model (IsoRSM) for these cases suggest that mesoscale upper air intrusion associated with low pressure systems might be the main process to bring extremely light δD to the ground surface. It is also found that δD of northerly wind was very light when sea-ice developed over the Sea of Okhotsk .

Ship-borne measurement from the Sea of Okhotsk (45 °N) to Pacific Ocean (20°N) indicated that values of δD changed largely with SST. However, highest δD approached isotopic equilibrium with sea water during quiescent weather but large variations exist when there are atmospheric disturbances such as low pressure systems and tropical cyclones. This result indicates that equilibrium fractionation of stable isotopes of water vapor occurs over the ocean. However, they are largely affected by such atmospheric disturbances as low pressure systems and tropical cyclones.

Relationship between δD and $\delta^{18}O$ of falling snow particles was also investigated in Sapporo and Kanazawa (36.54N, 136.67E). These observations were made from 5th Jan. to 16th Feb., 2011 in Sapporo and from 3rd to 4th Feb., 2010 in Kanazawa. The δD and $\delta^{18}O$ values of snow particles were linearly correlated with a correlation coefficient (r^2) of 0.95. The value of deuterium excess of the δD-$\delta^{18}O$ line is 45 (much greater than 10) and the slope is 9.1. The short-term variation of δD of falling snow particles did not correlate with that of water vapor.

学位論文審査の要旨

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This study performed almost 10-months high-temporal measurements of the stable isotope ratio (δD) of near-surface atmospheric water vapor by an Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS)-type water vapor isotope analyzer (WVIA) in Sapporo. The measurement was carried out on a continuous basis and on a high temporal resolution with a Water-Vapor Isotope Analyzer in four phases: (I) 28 April 2009 - 25 June 2009; (II) 15 Sep. 2009 - 29 Nov. 2009; (III) 1 April 2010 - 21 May 2010 and (IV) 17 Dec. 2010 - 30 May 2011. The Water Vapor Isotope Standard Source (WVISS) was used to automatically and comprehensively characterize WVIA's operation in real time. Both the reliability and scientific value of the data are very high, because the data obtained from the WVIA was corrected by comparing the δD values with those obtained from the cold trap method and subsequent cavity ring-down spectroscopy isotopic water analysis.

Based on the data, this study presents following outstanding and quite new results. The δD values showed a marked seasonal cycle but showed a different seasonal cycle from that of the surface air temperature. It is found that the monthly mean SST_{mon} of the nearest sea was a better predictor for δD_{mon} than the monthly mean air temperature.

This study successfully simulated observed long-term and short-term variation of δD

by using an Isotope-Global & Regional Spectral Models for the first time in the world. The simulated δD values showed almost the same seasonal cycle as that of observed δD values, although simulated values showed about 10‰ difference from observed values in 10-month average. This study found extremely low δD cases (lower than -210‰). Results of simulation by using an Isotope-Regional Spectral Model for these cases suggest that mesoscale upper air intrusion associated with low pressure systems might be the main process to bring extremely light δD to the ground surface.

Based on the data measured by the WVIA deployed on the *T/S Oshoro-Maru* of Hokkaido University, from the Sea of Okhotsk (45°N) to Pacific Ocean (20°N), this study confirmed that δD of water vapor generally increased with increasing SST and the highest values of δD were almost the same as those of vapor in equilibrium with ocean liquid. This result indicates that equilibrium fractionation of stable isotopes of water vapor occurs over the ocean. However, they are largely affected by such atmospheric disturbances as low pressure systems and tropical cyclones.

This study certainly promotes and accelerates the modeling and observational studies of the evaluation of the atmospheric moisture and hydrological cycle.

All of review members highly evaluate the research products of the applicant. Considering high quality of the applicant as a researcher and high score of the units, we hereby decided that the applicant is a person who is qualified to be a Doctor of Environmental Science.