Study on the Origin and Migration of the Pollutants in Precipitation and Surface Water in Seoul, Korea

Chungwan Lim[1] · Insung Lee[1]

1. Introduction

Variations in abundance of stable isotopes of sulfur, hydrogen, and oxygen are studied on snow and rainwater samples from Seoul area. Their suitability as a tool for characterizing the seasonality and source of regions near Korea, which contribute to the pollution in Seoul Area was investigated. Also stable isotopic and chemical evidence has been used to trace the sources of sulfate in precipitation at study area and to distinguish sulfate between natural and anthropogenic origins.

The contribution of different sources to a specific receptor site depend on emission patterns and meteorological conditions. Measured isotope ratios reflect the actual superposition of the distinct source types based on the assumption that no major isotope fractionation occurs during the transportation process between source region and sampling site. Furthermore, the isotope ratios of hydrogen and oxygen in the snow and rain water samples were used to support the meteorological information. Seasonal variations of the $\delta^{34}S$ values and their relation to chemical composition of the atmospheric pollutants are discussed taking into account the $\delta^{34}S$ values of sulfur originating from seasalt, oil and coal combustion, and biogenic activity. Also, the $\delta^{34}S$ values of sulfate in the atmospheric pollutants show a remarkable seasonal variation depending on the wind directions. The relationship between wind direction and seasonal variation can be explained by contribution of sulfur from coal combustion in northwestern Asia.

2. Analytical techniques

2.1. Preparation and Chemical analysis of sample

Analyses of the major chemical composition of water samples were performed using

Key words: sulfur isotope, oxygen and hydrogen isotope, air pollutants, environmental tracers, precipitation, sulfur compound, chemical analysis, anthropogenic origins, surface water, ground water

1) School of Earth and Environmental Sciences, Seoul National University
(lcwan1@snu.ac.kr, insung@snu.ac.kr)

ICP-AES (SHIMADZU / ICPS-1000IV) for S, Na^+, K^+, Mg^{2+}, Ca^{2+}, Ion-Chromatography (Center for Mineral Resources Research, Korea University : DIONEX-120 Automated Dual Column IC.) for Cl^-, SO_4^{2-}, NO_3^-, and NH_4^+. Aerosol sample was analysed by X-ray diffraction (XRD). A computerized Rigaku Geigerflex RAD3-C with a scintillation counter was used for XRD. Analysis of the similar aerosol sample as fused beads was also made using X-Ray Fluorescence Spectrometry (XRF) in National Center for inter-University Research Facilities, Seoul National University.

2..2 Oxygen, Hydrogen and sulfur isotope analysis

Oxygen, Hydrogen and sulfur isotope processings were carried out separately. Samples from snow and rainwaters were collected for oxygen, hydrogen and sulfur isotope measurements. Samples for oxygen isotopic analysis were prepared by H_2O-CO_2 equilibration. Sulfur in the SO_4^{2-} form in order to measure sulfur isotopes was used by barium sulfate precipitation method. The isotopic compositions of the samples were determined using a substantially modified Mass Spectrometry-VG Isotech Prism II at KBSI (Korea Basic Science Institute).

3. Result and discussion

3.1. Oxygen and Hydrogen isotope

Oxygen and hydrogen isotopic study of precipitation was carried out at Seoul, Gwanaksan. The isotopic composition of precipitation is quite variable, with $\delta^{18}O$ raging from -15.5 to -3.2‰ and δD from -115 to -31‰ (Fig.1.a).

Seasonal variation reflects in the oxygen and hydrogen isotopic data. The values of deuterium excess (d-values) for winter precipitation (d >10%) are clearly distinct from those for summer precipitation (d <10%) (Fig.1.b).

These data indicates the nature of the isotopic variation for different seasons. With the view of present data, the slope and intercept of the regression line for summer precipitation are virtually identical to the global meteoric water line (GMWL) defined by Craig (1961).

3.2. Sulfur isotope

The sulfur isotopic composition of sulfate ($\delta^{34}S_{SO_4}$) shows the range of +3.0~+6.5‰ in the snows and rainwaters. The observed isotopic composition is consistent with that of atmospheric SO_4 at Seoul area, which is attributed to anthropogenic emissions with minor contributions from sea spray. Similar $\delta^{34}S$ values with the range +0.5 to +5.5‰

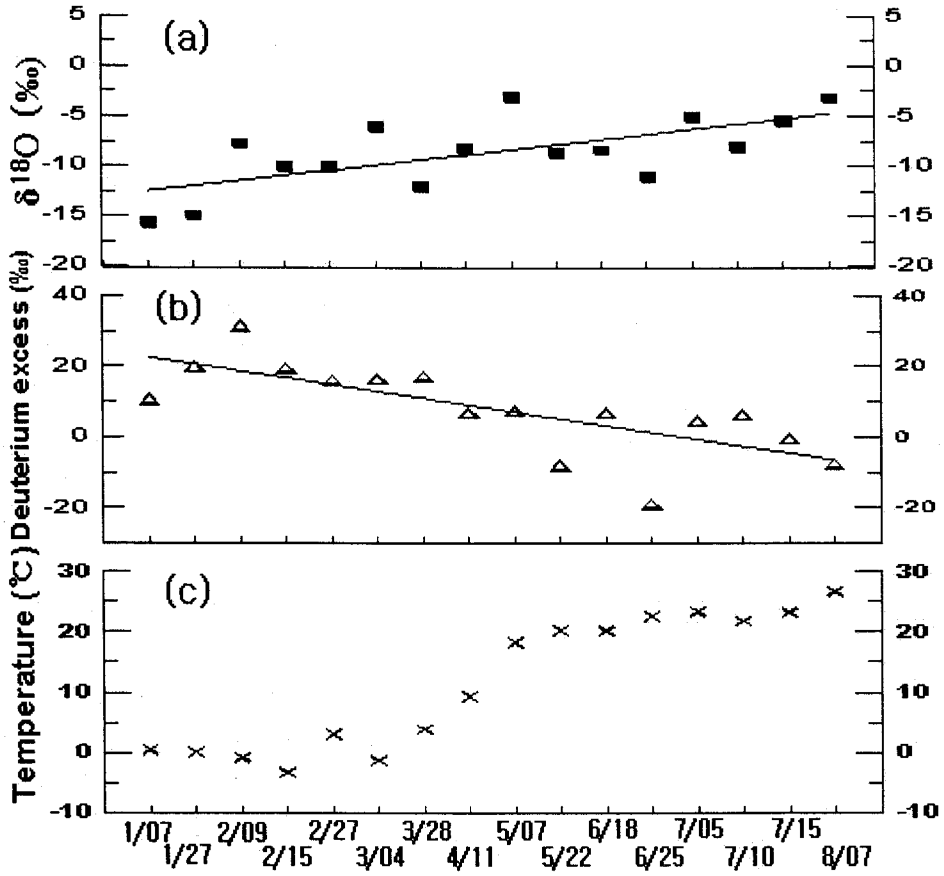

Fig. 1. Seasonal variations of oxygen isotopic composition(a), d-parameter(b), and air temperature(c) in Seoul area.

(mean +2.5 ±0.3‰) are also found by Nakai *et al.* (1991) in rainwater SO_4 from the heavily industrialized and urbanized Tokyo and Nagoya districts in Japan during 1978-1979. These observations highlight a major anthropogenic origin of SO_4 in precipitation at Seoul area. Additional sources, although minor, are sea spray and biological activity.

The present work shows the intriguing results that the isotopic values of the precipitation samples vary with season. $\delta^{34}S$ values of atmospheric samples are higher in winter and lower in summer. These results are in good accord with the Japanese counterparts studied for the five-year period by Maruyama (2000). The reason is that sulfur during the winter is mainly derived from space heating and industrial sources; but in summer the large emission of ^{34}S-depleted biogenic sulfur from soil, vegetation, marshes, and wetlands results in the lowering of $\delta^{34}S$ values of airborne sulfur.