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Geochemical and stable isotope characteristics of urban heavy rain in the downtown of Tokyo, Japan

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ABSTRACT

In order to make clear the impact of air pollution on the formation of sudden and locally-distributed heavy rain in urban area (hereafter Urban Heavy Rain: UHR), we analyzed inorganic ion concentration and stable isotope ratio of water (δD and $\delta^{18}O$) in rainwater. Rainwater samples were collected in Shinjuku, which is a representative downtown of Tokyo, Japan, during four years from October 2012 to December 2015. The concentration and wet deposition fluxes of acidic components (H^+, NH_4^+, NO_3^-, and nss-SO_4^{2-}) in UHR were especially higher than those in other types of rain events, i.e. normal rain, typhoon heavy rain, and frontal heavy rain. UHR had distinctly lower stable isotope ratios than those in other urban rains with same rainfall amount and summer precipitation systems. There was a high negative correlation between $\delta^{18}O$ and the distances from the sampling point to the formation area of UHR within 10 km, while there were high positive correlations between $\delta^{18}O$ and the concentration of acidic components in UHR. These findings indicate that UHR could effectively scavenge acidic substances within cloud and suggest the use of stable isotope ratios as tracers of an urban heavy rain's water and in-cloud scavenging process.

1. Introduction

Increase of summer heavy rain events has been caused in urban area all over the world (Changnon, 1968; Atkinson, 1971; Jauregui and Romales, 1996; Mikami et al., 2005; Goswami et al., 2006; Dou et al., 2015) from the latter part of the twentieth century. This event is a kind of convective heavy rains and occurs suddenly and locally in urban area, which we here call “Urban Heavy Rain (UHR)”. UHR has much rainfall amount with short rainfall time, and causes severe flood disaster when the amounts of rainfall exceed the capacity of the sewage system and draining canals (Kawasaki and Meguro, 2011), for example, the UHR in Tokyo in 1999, which had the rainfall intensity over 130 mm/h, caused urban flooding because the capacity of the sewage system was 50 mm/h in the city, and killed a man who was in a basement storage (Mikami et al., 2005). Urban flooding occurred not only in Japan, but also in urban areas all over the world. For example, in Turin, Italy, plenty of car drivers were forced to stay in their car because water reached up half of car doors in 2014 (Bertoldo et al., 2016). Since UHR is a global problem, clarification of UHR formation process has been an urgent atmospheric environmental issue.

The effect of urban heat island phenomenon (UHI) has been considered as a cause of increasing summer heavy rain events (Shiraki et al., 2009; Mikami et al., 2005; Fowler and Hennessy, 1995; Rajeevan et al., 2008). Urban areas are 2 to 4 °C warmer than surrounding areas due to more buildings and pavements, which have occupied the original place of trees and vegetation (Chandler, 1965; Bornstein, 1968; Oke and East, 1971; Kim, 1992; Wang et al., 1990; Robaa, 2003; Yamato et al., 2011). UHI causes the difference between the upper and the lower air temperature, and could produce ascending air current and accelerate the formation of cumulonimbus cloud, which leads to local heavy rainfall (Fujibe, 2004; Mikami et al., 2005). Numerical simulations indicated that changes of the distribution of anthropogenic heat in urban area greatly affected on the positions and amounts of rainfall (Ito et al., 2006; Lei et al., 2008; Miao et al., 2011). During two decades heavy rain events over 50 mm/h have been also frequently observed during summer in the most populous parts of Tokyo in Japan (Sato and Takahashi, 2000). These convective precipitations occurred mainly in leeward urban area and tended to occur at afternoon (Shepherd et al., 2002; Ito et al., 2006). In order to minimize the urban flooding damage caused by extreme rainfall, monitoring techniques of real time rainfall events have been developed. For example, Bertoldo et al. (2016) developed a simple X-band mini

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weather radar with high resolution both in time and in space, which could identify and track the rain cells. However, the formation mechanism of UHR is poorly understood.

UHI could locally increase O_3 concentration, which could enhance the oxidation of NO_x and SO_2 in urban areas (Khiem et al., 2010; Ooka et al., 2011). HNO_3 and H_2SO_4 formed by the oxidation of their precursors could react with NH_3 to form hygroscopic aerosols such as NH_4NO_3, NH_4HSO_4, and $(NH_4)_2SO_4$. Hygroscopic aerosols are transported above by strong updraft caused by temperature differences, and then, damp air from sea inflows in urban area (Fujibe et al., 2002). Under these conditions, hygroscopic aerosols act as cloud condensation nuclei (CCN) and form convective clouds locally and continuously (Bently et al., 2010). Wang et al. (2016) simulated the impacts of anthropogenic aerosols on summer precipitation over the Beijing–Tianjin–Hebei urban agglomeration in China. They revealed that anthropogenic aerosols enhanced summer rainfall and ascending motion, triggering more cloud formation and convective systems in the urban areas. Lacke et al. (2009) found that instability is greater and mixing layers are shallower on the days with high aerosol concentrations, resulting in greater precipitation amounts in most parts of the region in a study carried out in metropolitan Atlanta, using observed PM2.5 and precipitation amounts data from 2003 to 2004. UHI could cause a dust dome phenomenon and increases the concentration of air pollutants (Ooka, 2001). These studies indicated that anthropogenic aerosols formed in urban areas could contribute to the formation of convective clouds and extreme heavy rain, but the formation mechanism of UHR has been still unknown.

Stable isotope ratio of hydrogen and oxygen in rainwater has information about the sources of water vapor and precipitation formation process (Dansgaard, 1964; Risi et al., 2008). Combination of chemical and stable isotopic analysis of UHR is expected to give useful information of the formation process of UHR. We here make clear geochemical characteristics and wet deposition fluxes of air pollutants by UHR. Using combination of chemical and stable isotopic analyses, we clarify the impact of air pollutants on the formation of UHR.

2. Method

2.1. Sampling site and method

Rainwater samples were collected on the rooftop of a building (ca. 65 m) in the Nishi-Waseda campus (35.7°N, 139.7°E), Waseda University, which is located in Shinjuku, Japan (Fig.1). Shinjuku is a central ward of Tokyo and over 300,000 people including 40,000 foreigners live there. Area around Shinjuku station, which is a center of Shinjuku, is a huge business, commercial, and entertainment area. The sampling site is located along Meiji street (36,000 cars/day, Toshima city office, 2013) and about 2 km north of Sinjuku station, which is located at about 6 km northwest away from the Tokyo bay. The Keihin industrial zone, which has developed into the largest industrial area in Japan, is located there. Southerly winds bring various air pollutants emitted from business, commercial, and industrial areas along with humid sea air.

The 72 rainwater samples were collected on an event basis during 4 years from October 2012, to December 2015, using a polyethylene bottle (10 L) with a polyethylene funnel (diameter: 300 mm). Prior to use, the sampler was carefully washed with Milli-Q water and dried. The collector was placed manually just before each event (approximately 30–60 min before rainfall) and collected immediately after the event (on the day or the next day). The rainfalls beginning times were checked using radar charts of every 5 min obtained from Bureau of Sewerage Tokyo Metropolitan Government, Tokyo Amesh (2016). Events of precipitation less than 0.5 mm were not collected.

2.2. Analytical method

The rainwater samples were filtered with a membrane filter (pore size: 0.45 μm) to remove any particulate matter. The filtrates were stored in cleaned polypropylene bottles in a refrigerator at 4 °C until chemical and stable isotopic analyses. The pH was measured by a pH meter (MA325, METTLER TOLEDO). Calibration was performed before measurement using standards buffer solutions of pH 6.86 and 4.01 (Wako Pure Chemical, Osaka, Japan). Major cations (NH_4^+, Na^+, K^+, Mg^{2+}, and Ca^{2+}) and anions (Cl^-, NO_3^-, and SO_4^{2-}) were measured by ion chromatography. DIONEX ICS-1000 (separation column; Dionex Ion Pac® CG12A RFIC™ 4 × 250 mm, guard column; Dionex Ion Pac® CG12A RFIC™ 4 × 50 mm, eluent: 18 mM Methanesulfonic acid, suppressor; Thermo CERS) was used for analysis of major cations, while DIONEX DX-320 (separation column; Dionex Ion Pac™ AG4A-SC RFIC™ 4 × 250 mm, guard column; Dionex Ion Pac™ AG4A-SC RFICTM 4 × 50 mm, eluent: 1.8 mM Na_2CO_3 + 1.7 mM $NaHCO_3$, suppressor; DIONEX ASRS™) was used for analysis of major anions. The standards used for calibration were prepared from stock solutions of each ion (1000 mg/L, Wako Pure Chemical, Osaka, Japan). Dissolved inorganic carbon (DIC) was measured by a TOC meter (TOC-V_{CSH}, SHIMADZU). The calibration standards were prepared from stock solutions of $C_6H_4(COOK)(COOH)$ (Nacalai tesque, Kyoto, Japan). All solutions were prepared using Milli-Q water. The ratios of the sum of anions to the sum of cations in 72 rain samples were 0.95 ± 0.20, suggesting that almost all major inorganic ions were measured. Stable isotope ratio of rainwater ($\delta^{18}O$ and δD) was measured by a water stable isotope analyzer (L2130-i, PICCARO, measurement accuracy: 0.025‰). The water stable isotope standards were SMOW (standard mean ocean water, GL Sciences, Tokyo, Japan). We analyzed major inorganic ions for 72 samples and water stable isotopes for 51 samples.

2.3. Classification of heavy rain

According to the Japan Meteorological Agency, (n.d.) (JMA), heavy rain is defined as the hourly rainfall amount over 30 mm. However, UHR usually stops within 1 h, therefore, we defined heavy rain as a rain event with rainfall intensity more than 5 mm/10 min using data of 10-min rainfall amount from JMA to classify rain events. We also defined a normal rain as a rain event with 10-min rainfall amount lower than 5 mm during each event. In addition, we classified heavy rain events into 3 types by their formation processes; 1) Frontal heavy rain (FHR): heavy rains caused by a front, 2) typhoon heavy rain (THR): heavy rains caused by a typhoon, 3) UHR: sudden and locally-distributed heavy rains caused by neither a front nor a typhoon. Heavy rains were distinguished according to meteorological charts and weather cameras obtained from Japan Weather Association (2016). We divided 72 rainwater samples into 55 normal rain samples, 12 UHR samples, and 5 THR samples. Table 1 and Table 2 show the date and time of heavy rain occurrence (rainfall intensity exceeds 5 mm/10 min) during warm season (April–October) in studied period extracted from JMA rainfall amount data. We consulted the nearest two JMA observatories: Nerima station (35.7°N, 139.6°E, ca. 51 m) and Tokyo station (35.7°N, 139.8°E, ca. 25 m). Tables 1 and 2 shows UHR mainly occurs in summer. As for water stable isotopes of FHR samples, we only analyzed 2 of them, so we don't discuss them in this study.

3. Results and discussion

3.1. Geochemical characteristics of UHR

Table 3 shows the mean concentration of major inorganic ions in normal rain, UHR, and typhoon heavy rain (THR), which were collected at Shinjuku, Tokyo on an event basis, respectively. HCO_3^- concentration was calculated by the equilibrium calculation using rainwater pH and the concentration of dissolved total inorganic carbon. UHR had the