Aerosol and Air Quality Research, 16: 632–639, 2016

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2015.04.0256

Water-Soluble Organic Nitrogen in High Mountain Snow Samples from Central Japan

Tomoki Mochizuki[1], Kimitaka Kawamura[1*], Kazuma Aoki[2]

[1] *Institute of Low Temperature Science, Hokkaido University, N19 W8, Kita-ku Sapporo 060-0819, Japan*
[2] *Department of Earth Science, Faculty of Science, University of Toyama, 3190, Gofuku, Toyama 930-8555, Japan*

ABSTRACT

We measured water-soluble organic nitrogen (WSON) in snow pit samples, which were collected at the Murodo-Daira snowfield near the summit of Mt. Tateyama, central Japan in 2008, 2009, and 2011. The concentrations of WSON ranged from 12.8 to 96.7 ng g^{-1}, which were significantly lower than those reported in continental wet deposition samples from the Asian continent. WSON may be largely diluted in the snow samples during snowing processes over the high mountains. We found that WSON significantly correlated with nss-Ca^{2+} and water-soluble organic carbon (WSOC). Concentrations of WSON are likely controlled by the intensity of Asian dust events. Contributions of WSON to water soluble total nitrogen (WSTN) in snow pit sequence was found to be 15 ± 10%, which is lower than those (63–91%) of reference dust materials collected in China. Mass concentration ratio of WSOC/WSON was on average 23.7, which is significantly higher than the C/N weight ratio (5.6) calculated from the Redfield ratio. This result suggests that WSOC is largely produced by secondary photochemical oxidation of anthropogenic volatile organic compounds during the transport from East Asia to the high mountain areas in Japan. On the other hand, WSON may be contributed from Asian dusts from arid areas in China whereas water-soluble inorganic nitrogen may be derived from pollution sources.

Keywords: Water-soluble organic nitrogen; High mountain snow; Central Japan; Long-range atmospheric transport; East Asia.

INTRODUCTION

The annual global emissions of water-soluble inorganic nitrogen (WSIN) and water-soluble organic nitrogen (WSON) in particulate phase were estimated to be 34 and 3.9 Tg-N, respectively (Neff *et al*., 2002). WSON accounts for 10–40% of the water-soluble total nitrogen (WSTN) on a global scale (Cornell, 2011). Cape *et al*. (2011) reported that WSON concentrations in wet deposition are increasing recently; relatively high concentrations of WSON in wet deposition are observed in China due to the enhanced anthropogenic sources (Zhang *et al*., 2008). Increased emissions of nitrogen species to the air not only control the nitrogen saturation in terrestrial and aquatic ecosystems (e.g., Aber *et al*., 1989; Cornell *et al*., 1995), but also affect human health by decreasing the quality of air and water (e.g., Harrison and Yin, 2000; Zhang and Anastasio, 2001). Global nitrogen cycle should also be influenced by enhanced human activity. Water-soluble components including WSON in the atmosphere contribute to the formation of cloud condensation nuclei (CCN) and ice nuclei (IN) and thus have positive or negative radiative forcing, affecting global climate change (e.g., Kanakidou *et al*., 2005).

WSON is composed of various chemical species including amines, amino acids, urea, and nitrophenols (e.g., Cape *et al*., 2011). The WSON species can be emitted from marine organisms (Miyazaki *et al*., 2011), combustion system (Nojima *et al*., 1983), fertilizer (Ham and Tamiya, 2006), forest ecosystem (Miyazaki *et al*., 2014), and soil dust (Wang *et al*., 2013). It is known that removal process and/or chemical loss of water-soluble inorganic nitrogen (WSIN) (NO_3^- and NH_4^+) during long-range transport largely depend on the atmospheric processing time (e.g., Cornell, 2011) and meteorological conditions such as precipitation and fog (Sasakawa *et al*., 2002). However, the long-range transport of WSON and its fate in the atmosphere have been poorly understood.

Mt. Tateyama is located in central Japan, facing to the Sea of Japan (Fig. 1). Murodo-Daira locates near the summit of Mt. Tateyama and is covered with thick snow (> 6 m) in winter to spring. In our previous study, low molecular weight (LMW) monocarboxylic acids in the Murodo-Daira snowpit sequences have been reported (Kawamura *et al*., 2012). We found that higher concentrations of LMW

* Corresponding author.
Tel.: +81-11-706-5457; Fax: +81-11-706-7142
E-mail address: kawamura@lowtem.hokudai.ac.jp

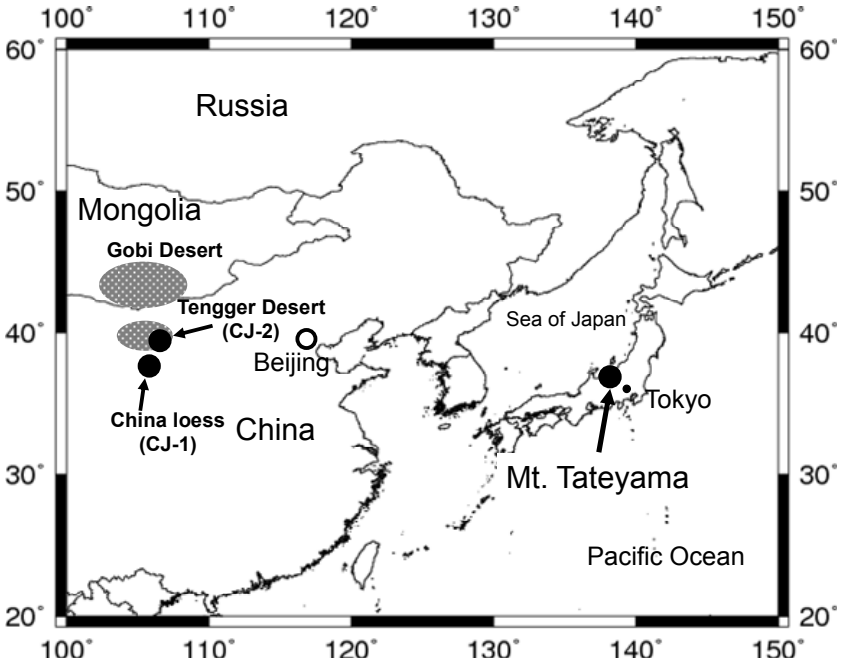

Fig. 1. Location of the snow pit sampling site near Mt. Tateyama, central Japan.

monocarboxylic acids in the snow pit sequences are associated with the presence of Asian dust layers (Kawamura *et al*., 2012; Mochizuki *et al.*, 2015). However, quantitative analysis of WSON has never been conducted in the snow pit sequences from the Japanese high mountain site. In this study, we measured WSON in 22 snowpack samples collected from the Murodo-Daira snowfield site near the summit of Mt. Tateyama, central Japan in 2008, 2009, and 2011. Here, we report WSON concentrations in the snow pit sequences and discuss long-range atmospheric transport of WSON from East Asia to the Japanese Islands during the season of Asian dust events.

MATERIALS AND METHODS

Snow accumulation during one winter season in 2008, 2009, and 2011 were 6.6 m, 6.0 m, and 6.5 m, respectively in the Murodo-Daira site (36.58ºN, 137.36ºE; elevation, 2450 m) near Mt. Tateyama (elevation, 3015 m), central Japan. Snowpack samples were collected in 2008 (n = 7), 2009 (5), and 2011 (10) from the Murodo-Daira site (Table 1). Because several dirty layers due to the occurrence of Asian dusts were recognized by visual observation, we have sampled the snow pit samples with dust layers to understand relationship between WSON and Asian outflow during the Asian dust season. After the collection of snow samples from the different layers of pit sequences, snow samples were placed in a pre-cleaned glass jar (8 L) and then mercuric chloride ($HgCl_2$) was added immediately to the samples to prevent microbial activity. The sample jar was sealed with a Teflon-lined screw cap, transported to the laboratory in Sapporo, and stored in the dark refrigerator room at 4°C prior to analysis. Kattner (1999) reported that the poisoning with $HgCl_2$ can make it possible to store inorganic nitrogen for a long period. Similarly, degradation of water-soluble organic nitrogen and water soluble organic carbon can be minimized by poisoning with $HgCl_2$ and storing the samples in cool and dark conditions.

After removing the particles from the melt snow sample using a disk filter (0.22 μm, Millipore Millex-GV, Merck, USA), water sample was analyzed for water-soluble organic carbon (WSOC) and water-soluble total nitrogen (WSTN) using a total organic carbon (TOC)/total nitrogen (TN) analyzer (Model TOC-Vcsh, Shimadzu). Before the WSOC and WSTN analysis, the extracts were treated with 1.2 M HCl and purged with a pure air to remove inorganic carbon and volatile organic compounds. We calculated WSON by subtracting WSIN (NH_4^+-N and NO_3^--N measured by ion chromatography) from WSTN (WSON = WSTN – WSIN) (Miyazaki *et al*., 2011). The uncertainties based on the combined analytical errors in the measurements of WSIN and WSTN were within 18% (Miyazaki *et al*., 2011). The concentrations of selected ionic species (NO_3^-, NH_4^+, and Ca^{2+}) in 2008, 2009, and 2011 are used from previous publications (Kawamura *et al*., 2012; Mochizuki *et al*., 2015). Concentrations of non-sea salt Ca^{2+} in melt snow sample is calculated by the following equation:

$$[\text{nss-}Ca^{2+}] = [Ca^{2+}] - 0.038 \times [Na^+] \quad (1)$$

where $[\text{nss-}Ca^{2+}]$ is concentrations of non-sea-salt (nss) Ca^{2+} and $[Ca^{2+}]$ and $[Na^+]$ are concentrations of total Ca^{2+} and Na^+, respectively (Berg and Winchester, 1978; Duce *et al*., 2003).

To evaluate the WSTN (WSIN and WSON) and WSOC of reference dust materials (Kosa); Chinese loess (CJ-1), Tengger desert (CJ-2), and Gobi desert (Gobi) provided by National Institute for Environmental Studies (Nishikawa *et al*., 2000, 2013), were analyzed for WSIN, WSTN, and WSOC. A dust sample (0.5 g) was extracted with 50 mL ultrapure water and analyzed by the method described above.

Table 1. List of snowpack samples collected from a pit at Morodo-Daira Mt. Tateyama, Japan in 2008, 2009, and 2011.

Year	Sample ID	Snow depth (cm)	Description
2007–2008	No. 1	10–20	Clean snow layer.
	No. 2	49–51	Snow with ice plate.
	No. 3	90–110	Dusty snow layer. Asian dust event were observed on March 3, 2008 by a lidar over Toyama.
	No. 4	120–130	Weak dust layer. Asian dust event were observed on February 11, 2008 by a lidar over Toyama.
	No. 5	350–360	Clean snow layer.
	No. 6	370–380	Dust snow layer with ice plate. Asian event were observed on January 11, 2008.
	No. 7	640–650	Clean snow layer.
2008–2009	No. 8	325–335	Weak dust layer. Asian dust event were observed on February 2, 2009 by a lidar over Toyama.
	No. 9	410–420	Clean snow layer.
	No. 10	425–435	Dusty snow layer. Asian dust event were observed on January 1, 2009 by a lidar over Toyama.
	No. 11	520–530	Dusty snow layer. Asian dust event were observed on December 10, 2008 by a lidar over Toyama.
	No. 12	530–540	Granular snow.
2010–2011	No. 13	115–125	Granular snow with ice plate.
	No. 14	169–178	Dusty and granular snow Asian dust event were observed on February 22–24, 20011 by a lidar over Toyama.
	No. 15	290–300	Compacted snow layer.
	No. 16	390–400	Compacted snow layer.
	No. 17	400–410	Dusty and compacted snow. Asian dust event were observed on December 31, 2010 by a lidar over Toyama.
	No. 18	430–440	Compacted snow layer.
	No. 19	460–466	Dusty and compacted snow. Asian dust event were observed on December 25–26, 2010 by a lidar over Toyama.
	No. 20	507–527	Compacted snow with ice plate.
	No. 21	542–548	Dusty and compacted snow. Asian dust event were observed on December 6, 2010 by a lidar over Toyama.
	No. 22	630–635	Granular snow.

Asian dust events over Imizu, Toyama prefecture, Japan near Mt. Tateyama during December to March in each year were recorded by the lidar observations (http://www-lidar.nies.go.jp/Toyama/). This result is supported by the air mass back trajectories at height of 3000 m (Figs. 2(a)–2(c)).

We calculated seven-day air mass back trajectories at height of 3000 m above sea level using the Meteorological Data Explorer (METEX) provided by the National Institute for Environmental Studies, Tsukuba, Japan (http://db.cger.nies.go.jp/metex/index.html).

RESULTS AND DISCUSSION

Air Mass Back Trajectory

The seven day air mass back trajectories at the Murodo-Daira site during the snowfall period (November–April) showed that most of the air masses passed through the Asian continent including China, Mongolia, and Korea and the Sea of Japan (Figs. 2(a)–2(c)), suggesting that snow pit samples are affected by the outflows from East Asia. In particular, the air masses associated with dust layers in 2008, 2009, and 2011 were mainly delivered from Taklamakan Desert, Gobi Desert, and Loess Plateau, which are characterized by heavy Asian dust events (see bold lines in Figs. 2(a)–2(c) and Table 1). In addition, air mass heights over the Asian continent and Sea of Japan were between 2500 m and 6000 m.

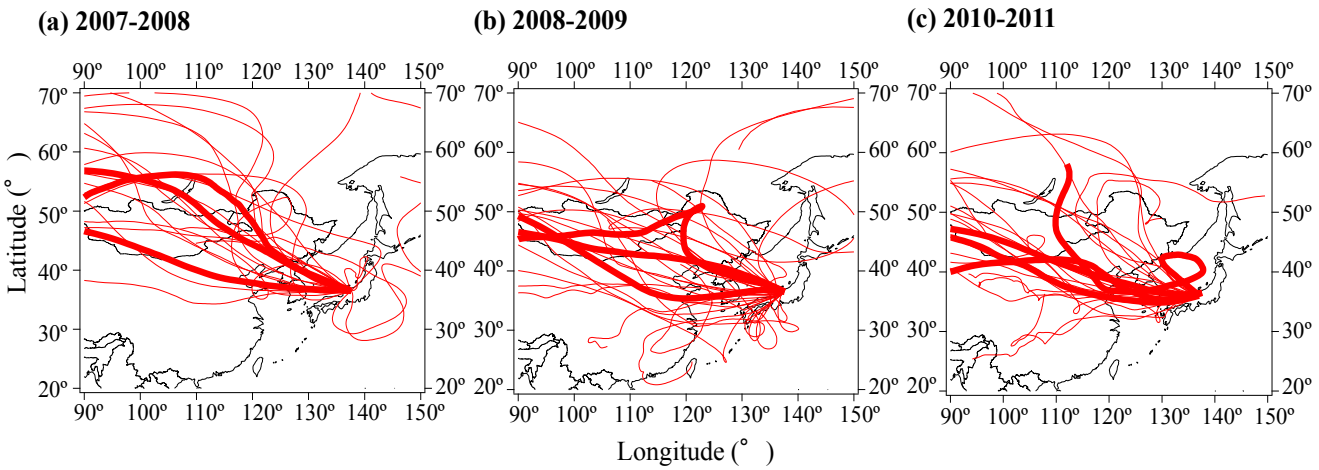

Fig. 2. Seven-day air mass back trajectories at a level of 3000 m a.s.l. during main snow fall period (January–March) in (a) 2008, (b) 2009, and (c) 2011. Bold lines show the trajectories associated with dust layers as observed by a lidar.

The monthly average of precipitation recorded in Urumqi (China), Beijing (China), and Seoul (Korea) were less than 20 mm, 6.0 mm, and 33 mm, respectively (Japan Meteorological Agency website: http://www.data.jma.go.jp/gmd/cpd/monitor/mainstn/obslist.php). Such low precipitations are important for the intensified uplift of dusts and subsequent long-range atmospheric transport from arid regions.

Water Soluble Organic Nitrogen

Fig. 3(a) presents concentrations of WSTN and WSON in the snowpack samples from the Murodo-Daira site at Mt. Tateyama, Japan in 2008, 2009, and 2011. The WSTN and WSON concentrations across all snow pack samples in 2008, 2009, and 2011 ranged from 94.7 to 1760 ng g^{-1} (425 ± 455 ng g^{-1}) and 12.8 to 96.7 ng g^{-1} (35.6 ± 22.5 ng g^{-1}), respectively. The average concentration of WSON in 2008 (44.4 ng g^{-1}) and 2009 (48.2 ng g^{-1}) are higher than that in 2011 (24.3 ng g^{-1}). Overall, WSON concentrations in the snow samples are significantly lower than those reported in rainwater samples from multiple sites (180 ng g^{-1}) in the world (Cornell, 2011) and from Shengsi Island, China in March (ca. 420 ng g^{-1}) (Zhang *et al*., 2011). However, WSON levels reported in the Murodo-Daira site are similar to those of the ice floe samples from the Fram Strait near the Arctic Ocean (ca. 50 ng g^{-1}) (Thomas *et al*., 1995), suggesting that WSON concentrations in the Japanese high mountain snow samples are near the background level on the Earth. Interestingly, the WSON levels in the Murodo-Daira site are very low, in spite that this site is affected by the outflows of air masses from the Asian continent (Figs. 2(a)–2(c)). WSON is emitted from marine biota (e.g., Miyazaki *et al*., 2011), but biological activity is low during winter. Our results suggest that WSON is less supplied from the Sea of Japan through long-range transport by the winter Asian Monsoon. The details of sources, decomposition, and transport of WSON will be discussed in later section.

Sources and Long-Range Transport of WSON

Several types of nitrogen compounds contribute to WSON. For example, amino acids are emitted not only from marine biological sources such as bacteria and degraded algal proteins but also from terrestrial sources such as plant debris and livestock farming. Organic nitrates and nitrophenols can be produced by the reaction of anthropogenic and biogenic hydrocarbons with nitrate radicals. Amines are derived from biogenic sources. These potential sources have been discussed in Cape *et al*. (2011). In addition, urea is emitted not only from primary sources such as microbial modification of agricultural fertilizers (Cape *et al*., 2011) but also from secondary formation via atmospheric reactions (Fu *et al*., 2014). Unfortunately, we are not able to identify these WSON components in this study.

Relatively high abundances of WSON were observed in snow layers with dusts such as snow samples Nos. 3 and 10. We found a positive correlation (r = 0.88) between WSON and nss-Ca^{2+}, an Asian dust tracer, using all the data points (Fig. 4(a)). This result suggests that Asian dust is an important controlling factor of WSON concentrations in snowpack samples. Co-variation was reported for nss-Ca^{2+} and LMW monocarboxylic acids at the Mt. Tateyama snow site (Kawamura *et al*., 2012; Mochizuki *et al*., 2015). On the other hand, relatively weak correlation was found between WSON and WSIN concentrations (r = 0.46) (Fig. 4(b)). In contrast, there is no correlation between WSIN and nss-Ca^{2+} (r = 0.22, not shown as a figure). We suggest that Asian dust can control the long-range transport of WSON and the production process in the atmosphere and that adsorption processes on the dust particles are similar between WSON and LMW monocarboxylic acids but these processes are different between WSON and WSIN.

Fig. 5 plots WSON concentrations as a function of WSOC concentrations. WSON shows a good correlation with WSOC (r = 0.81). It is known that WSOC is derived from biomass burning and secondary production via the photooxidation of gaseous hydrocarbons and other non-polar organic precursors (Snyder *et al*., 2009; Matsumoto *et al*., 2014). Our result indicates that WSON is biomass burning and secondarily produced by photochemical nitrification of organic gasses.

The average WSOC/WSON ratio at the Murodo-Daira site is 23.7, which is higher than those of reference dust

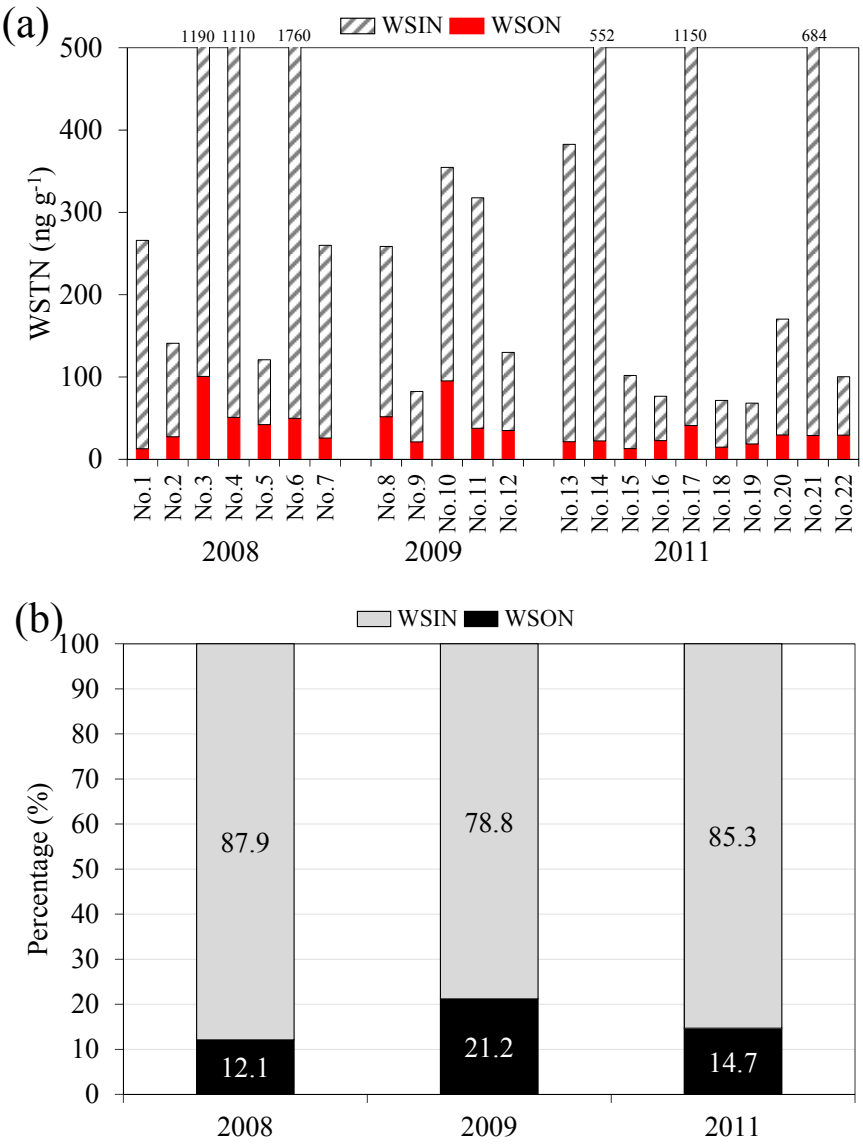

Fig. 3. (a) Concentrations of WSTN, WSON, and WSIN, and (b) % distributions of WSON and WSIN.

materials such as CJ-1 (4.9), CJ-2 (6.5) and Gobi (16.0) and those reported in urban (21.6) and rural aerosols (7.6) (e.g., Rastogi *et al*., 2011; Montero-Martinez *et al*., 2014), dust-influenced particles (0.84) in Xi'an, China (Wang *et al*., 2013), and fog water (7.6) from Italy (Montero-Martinez *et al*., 2014). In addition, C/N ratios of bacterial mass (5–10), amino acids (3–16) and organic nitrates (2–12) (Neff *et al*., 2002) are lower than the present snowpack samples (WSOC/WSON: 23.7). Interestingly, the average WSOC/WSON ratio in the present study was similar to that (24.0) reported for the ice floe samples from the Fram Strait near the Arctic Ocean (Thomas *et al*., 1995). For example, amino acids and 2- and 4-nitrophenol can be decomposed by photochemical processing in the atmosphere (e.g., Mikne and Zika, 1993; Vione *et al*., 2009). In addition, because air masses over industrial regions in China and Korea passed through a height of more than 2500 m, WSON may be largely decomposed in the atmosphere. The ratio of WSON/nss-Ca^{2+} (0.020) at the Murodo-Daira site was determined from the slope of WSON and nss-Ca^{2+} (Fig. 4(a)). This value is lower than that in the reference materials for dust particles including CJ-1 (0.055), CJ-2 (0.133) and Gobi (0.076). Therefore, the low WSON/nss-Ca^{2+} ratios suggest that WSON may be decomposed due to photochemical processes involved with ozonolysis and OH radical attack during long-range atmospheric transport of Asian dusts.

The fraction of WSON in WSTN ranged from 3.1 to 35.0% in the Murodo-Daira site snow pit sequences. Average WSON/WSTN ratio in 2009 (21.2%) is higher than in 2008 (12.1%) and 2011 (14.7%) (Fig. 3(b)). This result is consistent with the intensified Asian dust events in 2009 over the Japanese Islands (Mochizuki *et al*., 2015). However, because sources and production/removal processes are different between WSON and WSIN, WSON/WSTN ratios did not show a correlation with nss-Ca^{2+} for all the data points (r = 0.04). The overall average of WSON/WSTN ratios is 15.2 ± 10.2% in snowpack samples for three years at Mt. Tateyama, which is lower than that in global mean rainwater

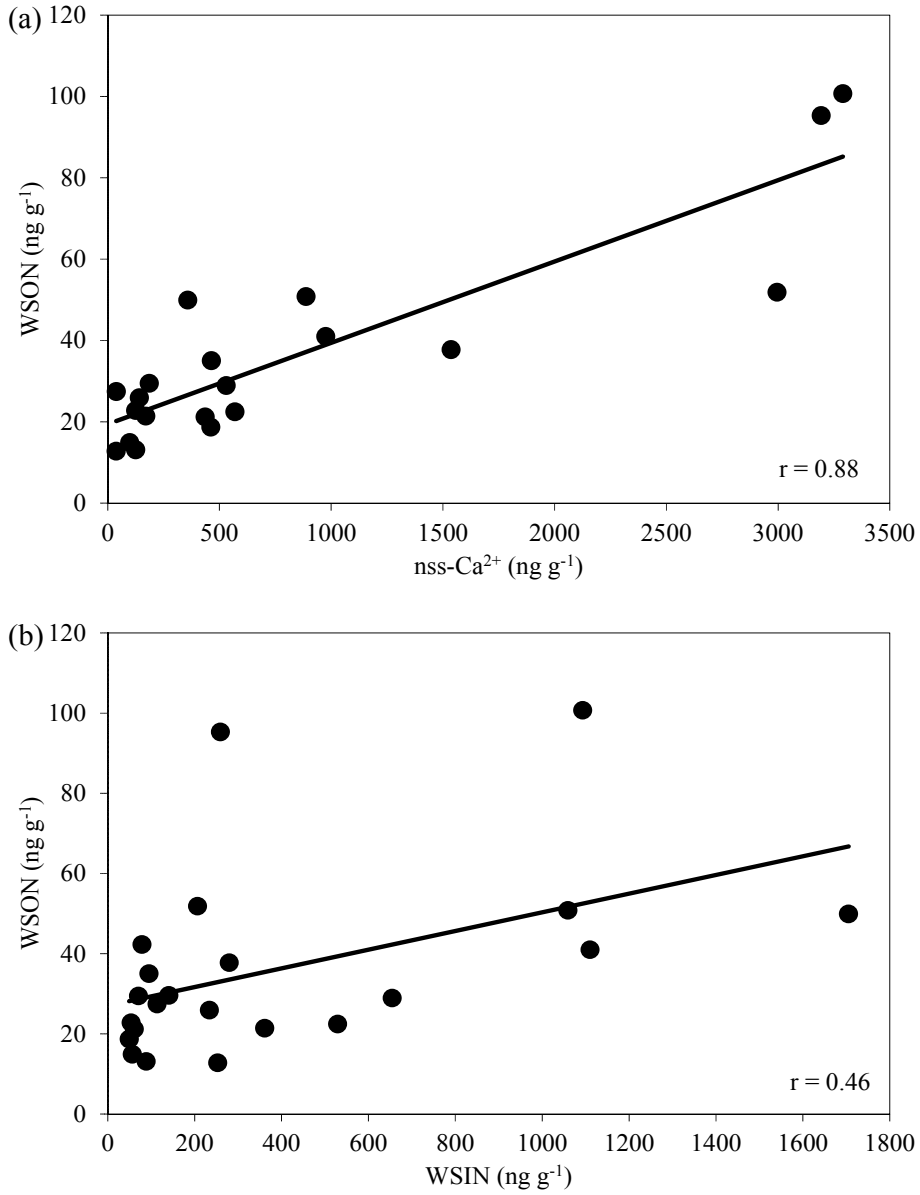

Fig. 4. Relations between (a) WSON and nss-Ca^{2+}, and (b) WSON and WSIN in Mt. Tateyama snow samples.

(30%) (Cornell, 2011) and particulate matter (31%) (Cape *et al*., 2011). In this study, we found high WSON/ WSTN ratios in reference dust materials including CJ-1 (66%), CJ-2 (63%) and Gobi (91%). Wang *et al*. (2013) also reported high WSON/WSTN ratio of 76 % in dusty aerosol samples from a semi-arid region in central China (dust storm period). In contrast, WSON/WSTN ratio in aerosol samples from Okinawa Island, Japan, an outflow region of Asian dust, was about 14% (Kunwar and Kawamura, 2014). We found that WSON does not comprise of major fraction of WSTN in the Japanese high mountain snow samples.

In conclusion, WSON in the snowpack samples has less significant influence from the Asian continent over the Japanese Islands due to the photochemical decomposition of WSON during long-range transport. In the future, the potential chemical aging of WSON and its degradation rate in the atmosphere should be investigated.

CONCLUSIONS

Snowpack samples were collected in the Murodo-Daira snowfield site near the summit of Mt. Tateyama, central Japan in 2008, 2009, and 2011, and analyzed for the measurement of WSON. The concentrations of WSON in the high mountain snow samples ranged from 12.8 to 96.7 ng g^{-1}, which are near the background level in the ice floe samples from the Fram Strait near the Arctic Ocean. On average, the average contribution of WSON to WSTN is 15 ± 10%. WSON is not a major component of WSTN in the high mountain snow samples. We found that concentrations of WSON significantly correlate with nss-Ca^{2+} (Asian dust tracer) and WSOC (secondary formation). Our study suggests that WSON is mainly derived from long-range transport of Asian dust associated with Chinese air pollutants over the Japanese Islands during winter. In addition, the ratios of

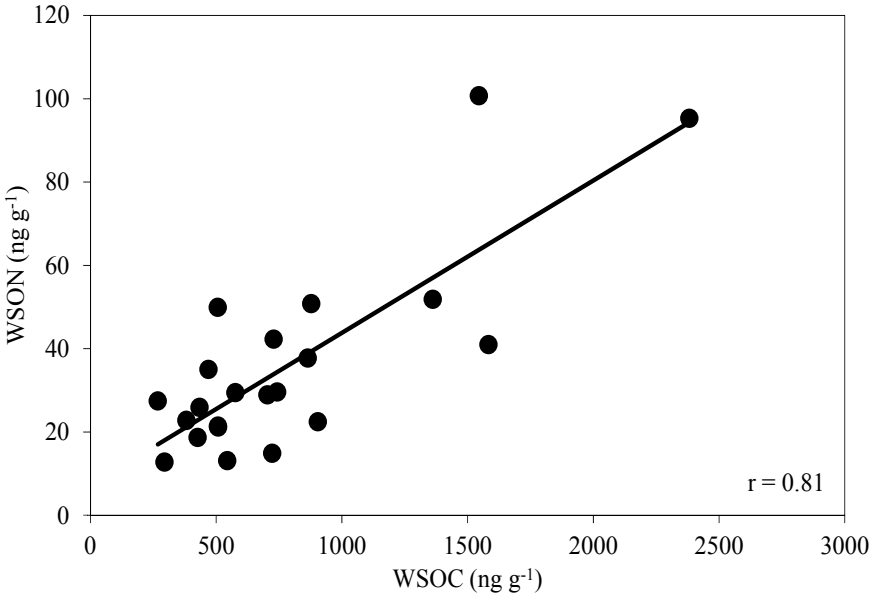

Fig. 5. Relation between WSON and WSOC in Mt Tateyama snow samples.

WSOC to WSON ranged from 9.8 to 48.5 (av. 23.7 ± 10.2), suggesting that WSON is significantly lower than WSOC in the high mountain snowpack samples in central Japan due to the possible decomposition of WSON and/or photochemical production of WSOC during long-range atmospheric transport.

ACKNOWLEDGEMENTS

This study was supported by the Japan Society for the Promotion of Science (Grant-in-Aid Nos. 1920405 and 24221001). We thank the students and researchers of University of Toyama for their help during the snow sampling. The data of this paper are available upon request to K. Kawamura (kawamura@lowtem.hokudai.ac.jp). We acknowledge Dr. N. Sugimoto for the courtesy of lidar data (http://www-lidar.nies.go.jp/Toyama/).

REFERENCES

Aber, J.D., Nadelhoffer, K.J., Steudler, P. and Melillo, J.M. (1989). Nitrogen Saturation in Northern Forest Ecosystems. *BioScience* 39: 378–386.

Berg, W.W., Jr. and Winchester, J.W. (1978). Aerosol Chemistry of the Marine Atmosphere. In *Chemical Oceanography*, 2nd Ed. 7. eds. Riley, J.P. and Chester, R. (Eds.), Academic Press, London, pp. 173–231.

Cape, J.D., Cornell, S.E., Jickells, T.D. and Nemitz, E. (2011). Organic Nitrogen in the Atmosphere – Where Does it Come from? A Review of Sources and Methods. *Atmos. Res.* 102: 30–48.

Cornell, S., Rendell, A. and Jickells, T. (1995). Atmospheric Inputs of Dissolved Organic Nitrogen to the Oceans. *Nature* 376: 243–246.

Cornell, S.E. (2011). Atmospheric Nitrogen Deposition: Revisiting the Question of the Importance of the Organic Component. *Environ. Pollut.* 159: 2214–2222.

Duce, R.A., Arimoto, R., Ray, B.J., Unni, C.K. and Harder, P.J. (1983). Atmospheric Trace Elements at Enewetak Atoll: 1, Concentrations, Sources, and Temporal Variability. *J. Geophys. Res.* 88: 5321–5342.

Fu, P.Q., Kawamura, K., Cheng, Y.F., Hatakeyama, S., Takami, A., Li, H. and Wang, W. (2014). Aircraft Measurements of Polar Organic Tracer Compounds in Tropospheric Particles (PM_{10}) over Central China. *Atmos. Chem. Phys.* 14: 4185–4199.

Ham, Y.S. and Tamiya, S. (2006). Contribution of Dissolved Organic Nitrogen Deposition to Total Dissolved Nitrogen Deposition under Intensive Agricultural Activities. *Water Air Soil Pollut.* 178: 5–13.

Harrison, R.M. and Yin, J. (2000). Particulate Matter in the Atmosphere: Which Particle Properties are Important for its Effects on Health? *Sci. Total Environ*. 249: 85–101.

Kanakidou, M., Seinfeld, J.H., Pandis, S.N., Barnes, I., Dentener, F.J., Facchini, M.C., Van Dingenen, R., Ervens, B., Nenes, A., Nielsen, C.J., Swietlicki, E., Putaud, J.P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G.K., Winterhalter, R., Myhre, C.E.L., Tsigaridis, K., Vignati, E., Stephanou, E.G. and Wilson, J. (2005). Organic Aerosol and Global Climate Modelling: A Review. *Atmos. Chem. Phys*. 5: 1053–1123.

Kattner, G. (1999). Storage of Dissolved Inorganic Nutrients in Seawater: Poisoning with Mercuric Chloride. *Mar. Chem.* 67: 61–66.

Kawamura, K., Matsumoto, K., Tachibana, E. and Aoki, K. (2012). Low Molecular Weight (C_1-C_{10}) Monocarboxylic Acids, Dissolved Organic Carbon and Major Inorganic Ions in Alpine Snow Pit Sequence from a High Mountain Site, Central Japan. *Atmos. Environ.* 62: 272–280.

Kunwar, B. and Kawamura, K. (2014). One-year Observations of Carbonaceous and Nitrogenous Components and Major Ions in the Aerosols from Subtropical Okinawa Island, and Outflow Region of Asian Dusts. *Atmos. Chem. Phys.* 14: 1819–1836.

Matsumoto, K., Yamamoto, Y., Kobayashi, H., Kaneyasu, N. and Nakano, T. (2014). Water-soluble Organic Nitrogen in the Ambient Aerosols and its Contribution to the Dry Deposition of Fixed Nitrogen Species in Japan. *Atmos. Environ.* 95: 334–343.

Milne, P.J. and Zika, R.G. (1993). Amino Acid Nitrogen in Atmospheric Aerosols: Occurrence, Sources and Photochemical Modification. *J. Atmos. Chem.* 16: 361–398.

Miyazaki, Y., Kawamura, K., Jung, J., Furutani, H. and Uematsu, M. (2011). Latitudinal Distributions of Organic Nitrogen and Organic Carbon in Marine Aerosols over the Western North Pacific. *Atmos. Chem. Phys*. 11: 3037–3049.

Miyazaki, Y., Fu, P. Q., Ono, K., Tachibana, E. and Kawamura, K. (2014). Seasonal Cycles of Water-soluble Organic Nitrogen Aerosols in a Deciduous Broadleaf Forest in Northern Japan. *J. Geophys. Res.* 119: 1440–1454, doi: 10.1002/2013JD020713.

Mochizuki, T., Kawamura, K., Aoki, K. and Sugimoto, N. (2015). Long-range Atmospheric Transport of Volatile Monocarboxylic Acids with Asian Dust over High Mountain Snow Site, Central Japan. Submitted to *Geochim. Cosmochim. Acta*, April 1.

Montero-Martinez, G., Rinaldi, M., Gilardoni, S., Giulianelli, L., Paglione, M., Decesari, S., Fuzzi, S. and Facchini, M.C. (2014). On the Water-soluble Organic Nitrogen Concentration and Mass Size Distribution during the Fog Season in the Po Valley, Italy. *Sci. Total Environ.* 485–486: 103–109.

Neff, J.C., Holland, E.A., Dentener, F.J., Mcdowell, W.H. and Russell, K.M. (2002). The Origin, Composition and Rates of Organic Nitrogen Deposition: A Missing Piece of the Nitrogen Cycle? *Biogeochemistry* 57/58: 99–136.

Nishikawa, M., Hao, Q. and Morita, M. (2000). Preparation and Evaluation of Certified Reference Materials for Asian Mineral Dust. *Global Environ. Res.* 4: 103–113.

Nishikawa M., Dashdondog B., Ukachi M., Onishi K., Nagano K., Mori K., Matsuia I. and Sano T. (2013). Preparation and Chemical Characterization of an Asian Mineral Dust Certified Reference Material. *Anal. Methods* 5: 4088–4095.

Rastogi, N., Zhang, X., Edgerton, E.S., Ingall, E. and Weber, R.J. (2011). Filterable Water-soluble Organic Nitrogen in Fine Particles over the Southeastern USA during Summer. *Atmos. Environ.* 45: 6040–6047.

Sasakawa, M. and Uematsu, M. (2002). Chemical Composition of Aerosol, Sea Fog, and Rainwater in the Marine Boundary Layer of the Northwestern North Pacific and its Marginal Seas. *J. Geophys. Res.* 107, doi: 10.1029/2001JD001004.

Snyder, D.C., Rutter, A.P., Collins, R., Worley, C. and Schauer, J.J. (2009). Insights into the Origin of Water Soluble Organic Carbon in Atmospheric Fine Particulate Matter. *Aerosol Sci. Technol.* 43: 1099–1107.

Thomas, D.N., Lara, R.J., Eicken, H., Kattner, G. and Skoog, A. (1995). Dissolved Organic Matter in Arctic Multi-year Sea Ice during Winter: Major Components and Relationship to Ice Characteristics. *Polar Biol.* 15: 477–483.

Vione D., Maurino, V., Minero, C., Duncianu, M., Olariu, R.I., Arsene, C., Saralja, M. and Mailhot, G. (2009). Assessing the Transformation Kinetics of 2- and 4-Nitrophenol in the Atmospheric Aqueous Phase. Implications for the Distribution of Both Nitroisomers in the Atmosphere. *Atmos. Environ.* 43: 2321–2327.

Wang, G.H., Zhou, B.H., Cheng, C.L., Cao, J.J., Li, J.J., Meng, J.J., Tao, J., Zhang, R.J. and Fu, P.Q. (2013). Impact of Gobi Desert on Aerosol Chemistry of Xi'an, Inland China during Spring 2009: Differences in Composition and Size Distribution between the Urban Ground Surface and the Mountain Atmosphere. *Atmos. Chem. Phys.* 13: 819–835.

Zhang, Y., Zheng, L., Liu, X., Jickells, T., Cape, J.N., Goulding, K., Fangmeier, A. and Zhang, F. (2008). Evidence for Organic N Deposition and its Anthropogenic Sources in China. *Atmos. Environ.* 42: 1035–1041.

Zhang, J., Zhang, G.S., Bi, Y.F. and Liu, S.M. (2011). Nitrogen Species in Rainwater and Aerosols of the Yellow and East China Seas; Effects of the East Asian Monsoon and Anthropogenic Emissions and Relevance for the NW Pacific Ocean. *Global Biogeochem. Cycles* 25: GB3020, doi: 10.1029/2010GB003896.

Zhang, Q. and Anastasio, C. (2001). Chemistry of Fog Waters in California's Central Valley Part 3: Concentrations and Speciation of Organic and Inorganic Nitrogen. *Atmos. Environ*. 35: 5629–5643.

Received for review, April 24, 2015
Revised, August 16, 2015
Accepted, September 15, 2015