5A5) Temporal Variations of Total Mercury Concentration in Precipitation: Difference in Source Characteristics between China and Korea

<u>Duc Luong Nguyen</u>^{1),2)} · Shang Gyoo Shim¹⁾ · Jin Young Kim¹⁾ Hyoun Cher Jin¹⁾ · Xiao-shan Zhang³⁾

1. Introduction

Total mercury (THg) in precipitation is resulted from wet scavenging of both atmospheric reactive gaseous mercury (RGM) and total particulate mercury (TPM). East Asia is the largest mercury source region in the world. China, at present, is the largest single country polluter regarding to mercury which contributes approximately 30% to the global emissions of mercury and 50% of the total emissions of mercury in Asia (Pacyna et al., 2006). The objectives of this study were to: (1) compare the levels and temporal variations of atmospheric TPM concentration and THg concentration in precipitation between China and Korea; and (2) elucidate the difference in source characteristics influencing temporal variations of THg concentration in precipitation between China and Korea.

2. Method

Measurements of atmospheric TPM concentration were conducted at two background sites including Chengshantou (China), Deokjeok island (Korea), three urban sites including Beijing, Ningbo (both in China), and Seoul (Korea) during 2007–2008 as shown in Fig. 1 using high-volume air sampler (SIBATA HV-1000F). Precipitation samples were collected on a weekly basis at Chinese sites and on an event basis at Korean sites during 2007–2008 using automatic wet-only precipitation collectors. Analytical procedures for determination of TPM and THg in precipitation were carried out using a DMA-80 Direct Mercury Analyzer (Milestone, Shelton, CT) according to the US-EPA Method 7473.

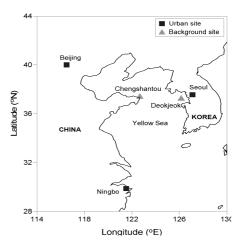


Fig. 1. Location of sampling sites for total particulate mercury and mercury wet deposition.

3. Results and Discussion

3.1 Characterization of atmospheric TPM concentrations and THg concentrations in precipitation

Atmospheric TPM mean concentrations measured at sampling sites during 2007–2008 were depicted in Fig. 2. TPM mean concentrations measured at Beijing and Ningbo sites were significantly high while those measured at Chengshantou, Deokjeok, and Seoul sites were within or close to the range of typical background levels of TPM (1–86 pg m⁻³, Keeler et al., 1995). Highly

¹⁾Global Environment Center, Korea Institute of Science and Technology (KIST),

²⁾University of Science and Technology (UST),

³⁾Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences

elevated TPM concentrations at Chinese urban sites are evidences for the strong impact of local anthropogenic mercury sources, especially coal combustion. TPM mean concentrations in the cold periods were higher than those in the warm periods at all sites (Fig. 3), probably due to the increased coal combustion for domestic heating and/or lower ambient temperatures in the cold periods increase the adsorption or condensation of the vapor mercury onto the atmospheric particles and decrease mercury volatilization from particles.

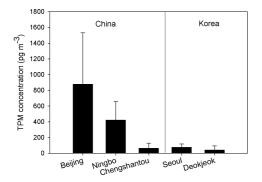


Fig. 2. TPM mean concentrations measured in China and Korea during 2007–2008. Error bars represent the standard deviations.

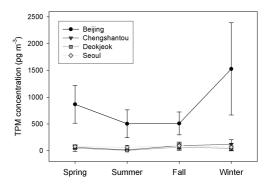


Fig. 3. Temporal variations of TPM mean concentrations measured at Beijing, Chengshantou, Deokjeok, and Seoul sites. Error bars represent the standard deviations.

Volume-weighted mean THg concentrations in precipitation measured during 2007-2008 were 36.86, 30.39, 23.66, 19.44, and 18.36 ng L⁻¹ for Beijing, Ningbo, Chengshantou, Seoul, and Deokjeok respectively. Higher THg concentrations at urban sites than those at background site in China reflects the importance of local anthropogenic sources in Chinese urban areas while the difference in THg concentrations (for both entire study period and seasonal basics) between urban and background sites in Korea was not so significant implying that local anthropogenic source emissions were possibly not the dominant process influencing THg concentrations in precipitation in Korean urban area. Precipitation amount and THg concentration in precipitation measured at sampling sites were shown in Fig. 4 and 5. It was seen that the temporal variations of THg concentration in precipitation were similar to those for atmospheric TPM concentration at all sites.

3.2 Difference in source characteristics influencing temporal variations of THg concentration in precipitation between China and Korea

In order to investigate the influence of source processes in temporal variations of THg concentration in precipitation, relationship between precipitation amount and THg concentration in precipitation was analyzed for both spring (cold period) and summer-fall (warm periods). As shown in Fig. 6a, there were strongly negative correlation between precipitation amount and THg concentration in precipitation, especially those at Chinese sites (except Chengshantou) during the cold period indicating that TPM was effectively scavenged from the atmosphere within the initial period. This implies that scavenging of TPM is one of dominant processes influencing the variations of THg concentration in precipitation. However, the variability within the data indicates that other processes are also influencing, possibly scavenging of RGM. Significantly higher TPM concentrations

in the cold period might result in higher THg concentrations than those in the warm periods despite that the removal process is less efficient in the cold period than that in the warm periods. Stronger relationship observed in China than that in Korea probably associated with significantly higher TPM concentration in China. There was an evidence of the difference in major source processes influencing THg concentrations in precipitation during the warm periods between China and Korea. Fig. 6b shows that there were still negative correlation between precipitation amount and THg concentration in precipitation at Chinese sites; however the relationship strengths were less than those during the cold period. This suggested that variations of THg concentration during the warm periods in China are still governed by scavenging processes of both TPM and RGM. In the contrast, slightly positive correlation between precipitation amount and THg concentration in precipitation was observed at Korean sites suggesting that a large and "unlimited" source term for THg rather than scavenging of TPM. It is reasonably inferred that scavenging of RGM is primary responsible for variations of THg concentration in precipitation during the warm periods in Korea. Since local anthropogenic emissions were not the dominant source of THg concentrations in precipitation as discussed earlier, then source of RGM is probably the in-situ gas-phase oxidation of Hg⁰ by atmospheric oxidants which is favored during warm periods with higher air temperature, solar radiation, and concentration of atmospheric oxidants.

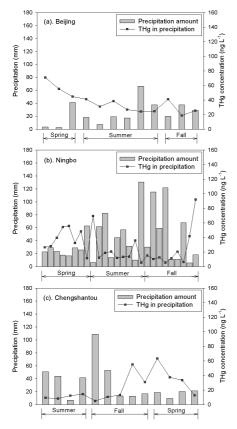


Fig. 4. Weekly-integrated precipitation amount and THg concentration in precipitation measured at Chinese sites.

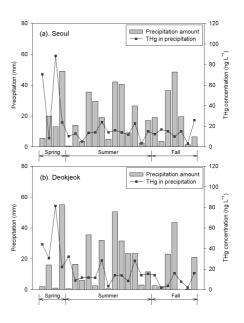


Fig. 5. Event-based precipitation amount and THg concentration in precipitation measured at Korean sites.

4. Summary

This study shows that atmospheric TPM concentrations and THg concentrations in precipitation measured in China were higher than those measured in Korea. TPM concentrations and THg concentrations in precipitation during the cold periods were generally higher than those during the warm periods in both China and Korea. In China, variations of THg concentration in precipitation during the cold and warm periods were influenced by scavenging of both TPM and RGM. Different from China, in Korea, variations of THg concentration in precipitation during the cold period were also influenced by scavenging of both TPM and RGM; however, those during the warm periods were dominantly governed by scavenging of RGM.

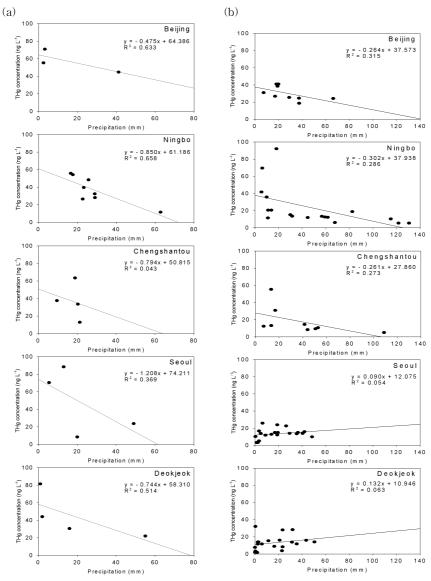


Fig. 6. Relationship between precipitation amount and THg concentration in precipitation measured at sampling sites during: (a). spring (cold period) and (b). summer-fall (warm periods).

References

- Keeler, G., G. Glinsorn, and N. Pirrone, N. (1995) Particulate mercury in the atmosphere: its significance, transport, transformation and sources. Water, Air, and Soil Pollution, 80, 159–168.
- Pacyna, E.G., J.M. Pacyna, F. Steenhuisen, and S. Wilson (2006) Global anthropogenic mercury emission inventory for 2000. Atmospheric Environment, 40, 4048–4063.