

Interactive comment on “Confirming anthropogenic influences on the major organic and inorganic constituents of rainwater in an urban area” by K. Chon et al.

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Response to the Reviewer Comments

Comment #1: This is a prospective study that investigates the seasonal variation of rainwater chemical composition, and to identify possible sources of inorganic and organic compounds in rainwater. The authors present valuable data that are useful for future environmental studies. This manuscript therefore can be accepted after minor revision as below: Response: We appreciate your comments and thank you for your positive recommendation.

Comment #2: In abstract, It would be better if includes some statement of the problem,
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objectives and a summary of employed methods in the research approach. Response: According to your comment, we have added the some statement of the problem, aims in the abstract. Main experimental methods already have been mentioned in the abstract, abstract section has been modified as follows:

“Recently, rainwater composition affected by atmospheric pollutants has been the topic of intense study in East Asia because of its adverse environmental and human health effects. In the present study, the chemical composition and organic compounds of rainwater were investigated from June to December 2012 at Gwangju in Korea. The aim of this study is to determine the seasonal variation of rainwater chemical composition, and to identify possible sources of inorganic and organic compounds. The volume weighted mean of pH ranged from 3.83 to 8.90 with an average of 5.78. 50% of rainwater samples had pH values below 5.6. The volume-weighted mean concentration (VWMC) of major ions followed the order: $\text{Cl}^- > \text{SO}_4^{2-} > \text{NH}_4^+ > \text{Na}^+ > \text{NO}_3^- > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$. The VWMC of trace metals decreased in the order as follows $\text{Zn} > \text{Al} > \text{Fe} > \text{Mn} > \text{Pb} > \text{Cu} > \text{Ni} > \text{Cd} > \text{Cr}$. The VWMCs of major ions and trace metals were higher in winter than in summer. The high enrichment factors indicate that Zn, Pb, Cu, and Cd originated predominantly from anthropogenic sources. Factor analysis (principal component analysis) indicates the influence of anthropogenic pollutants, sea salt, and crustal materials on the chemical compositions of rainwater. Benzoic acids, 1H-Isoindole-1,3(2H)-dione, phthalic anhydride, benzene, acetic acids, 1,2-benzenedicarboxylic acids, benzonitrile, acetaldehyde, and acetamide were the most prominent pyrolysis fragments for rainwater organic compounds identified by pyrolysis gas chromatography/mass spectrometry (Py-GC/MS). The results indicate that anthropogenic sources are the most important factors affecting the organic composition of rainwater in urban area.”

Comment #3: As the objective of this study is to investigate the seasonal variation of rainwater chemical composition, why did you collect the samples from June to December and not for one year? It is highly unusual to collect the samples during 6months for

seasonal study. Response: Thank you for your critical comment. We agree with your comment. This is our pre-study to investigate seasonal variation of rainwater chemical composition. Firstly, we wanted to compare chemical composition of rainwater between summer and winter. Even though our study period was quite short compared to other studies, we tried to present valuable analysis data related to the chemical composition and organic compounds of rainwater. We are planning to investigate long term seasonal variation of rainwater chemical composition for the further studies.

Comment #4: In introduction section there is no relevant literature related to anthropogenic influence in rainwater. Response: We thank the reviewer for this comment. We have already included the literature review about anthropogenic influence in rainwater in the introduction section. We have highlighted the sentences with red color as follows: "Rain is an efficient scavenging process for pollutants in the air and is becoming a source of pollution to the environment (Santos et al., 2011). The emission of SO₂ and NO_x from fossil fuel combustion and industrial processes has rapidly increased in East Asia due to its fast growing economy. These gases are converted into sulfuric and nitric acids before precipitating as acid rain (Lee et al., 2000; Báez et al., 2006). Consequently, there have been adverse environmental effects on aquatic, biological, and terrestrial systems (Bard, 1999; Başak and Alagha, 2004). Thus, the chemical composition of precipitation has been investigated all around the world during the last decade (Lara et al., 2001; Mouli et al., 2010; Santos et al., 2011). Coal combustion, automobile exhaust, and industrial emissions represent the dominant anthropogenic sources of heavy metals in rainwater (Kaya and Tuncel, 1997; Hu and Balasubramanian, 2003; Cheng et al., 2011). Heavy metals from precipitation accumulate in the biosphere and may cause adverse human health and environmental effects (Barrie et al., 1987; Báez et al., 2007). Thus, the studies of heavy metals in rainwater have increased in many countries (Pike and Moran, 2001; Al-Momani, 2003; Báez et al., 2007)."

Comment #5: In the Table 2, major ions in rainwater at Gwangju were compared to those reported in other urban areas around the world. There is no analyzing for these

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data, for example why Cl⁻ in Gwangju and Istanbul is much higher than those reported for other sites? Response: We thank the reviewer for this insightful comment. We have already included the analysis and discussion for the Table 2 in the manuscript. The Cl⁻ value is comparable to that in Istanbul and much higher than those reported for other sites. The high concentration of marine elements was likely due to typhoons during summer seasons. Please, refer to Page 83, line 12-26 as follows: "The VWMC of major ions in rainwater at Gwangju were compared to those reported in other urban areas around the world (Table 2). The pH value measured in our study is lower than that in Tirupati, India, but higher than those reported for other sites. The concentration of Na⁺ is comparable to that in Shanghai, but lower than Istanbul and higher than other areas. The Cl⁻ value is comparable to that in Istanbul and much higher than those reported for other sites. The high concentration of marine elements was likely due to typhoons during summer seasons. The value of NO₃⁻ is higher than that in Southeast Brazil and close to that in other sites. Regarding SO₄²⁻, its concentration is higher than that in Seoul, Mexico and Brazil and lower than in other areas. Shanghai shows the highest value of NO₃⁻ and SO₄²⁻ indicating severe air pollution problems in China. Both of these ions were mainly derived from the high coal/fuel consumption and mobile sources. The concentration of K⁺, Ca²⁺, Mg²⁺ are higher than those in Seoul, Mexico, Brazil and lower than ones in other sites. The concentrations of these ions are comparatively higher in Istanbul than in other areas. The value of NH₄⁺ is comparable to that in Seoul and higher than that in India, Turkey, and Brazil."

Comment #6: In the section 3.7, what is the level of anthropogenic origin? Also providing some comparisons with previous studies are needed.

Response: We thank the reviewer for this comment. However, we are not sure what you exactly want to ask about the anthropogenic origin. We have already provided some comparisons with previous studies. We have highlighted the sentences with red color as follows: "Table 6 lists the matrix correlation between ions and trace metals (correlation coefficients greater than 0.5 are marked in bold letters). Moderate correlation

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was found among the trace metals and NH₄⁺, NO₃⁻, and SO₄²⁻, with the exception of Cr and Zn, suggesting anthropogenic origin of the species. These correlations were also observed in previous studies (Garcia et al., 2006; Jung et al., 2011). Significant correlations between SO₄²⁻ and trace metals in rainwater were observed in Clarke and Radojevic (1987)."

Comment #7: Please double check the errors bar in the most figures. Response: According to your comment, we have double checked the error bars in the figures.

Please also note the supplement to this comment:

<http://www.drink-water-eng-sci-discuss.net/8/C52/2015/dwesd-8-C52-2015-supplement.pdf>

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