

We thank the reviewers for careful reading and helpful comments that improve the quality of the manuscript. Reviewer comments have been copied followed by our responses in bold.

Anonymous Referee #1)

In the manuscript, a hypothesis was proposed based on the intensive field measurements and validated by a smog chamber experiment. In addition, the authors discussed on the validity of the policy direction to reduce NO_x emission.

The manuscript is well structured and the logical development of the hypothesis is reasonable. Also, the implications of the research results look interesting. However, there are some parts that should be addressed in more detail and that can be improved. Thus, I recommend the manuscript be published in the ACP with some revisions. These points are:

R1C1) In Abstract, it can be written more clearly. For example, the sentences in lines 15-17 can be re-written.

Response)

Now it reads:

“... was sampled and analyzed. Due to the synoptic meteorological conditions PM at Deokjeok Island can be served as a surrogate of Chinese PM transported into Seoul. We hypothesize ...”

R1C2) The hypothesis is a reasonable one. Still, in most areas in Northeast Asia, ammonia levels are rather high and it would affect the nitrate uptake into the particulate phase. The authors should discuss in more detail, especially, in section 3.3.2 on the effects of ammonia in the ambient conditions.

Response)

We agree that ammonia plays an important role in particle formation. To investigate the effect of ammonia, we use ISORROPIA-II with the forward mode, which simulates the partitioning from the gas phase. We first simulate nitrate uptake when only NO₂ is present (i.e., [NO₂] = [HNO₃] = 68 ppb) (Fig. S8A). Then we simulate nitrate and ammonia uptake when both NO₂ and ammonia are present (Fig. S8B). We assume the ammonia concentration at Seoul during winter is 10 ppb (Lee et al., 1999). Although these simulation results still underestimate the measurements of Seoul haze particles, it clearly demonstrates that the presence of ammonia increases not only ammoniums, but also nitrates in particles. And it also lowers the acidity of particles a little bit from pH 2.7 to pH 3.2. It should be noted that our equilibrium model works for HNO₃ uptake; however, it simulates unrealistically high concentrations for both ammonia and HNO₃ uptake since in the equilibrium model ALW is assumed to be infinite and the concentrations of NH₃ and HNO₃ are constants (i.e., it is an “open” system) so ammonium and nitrate reach unrealistically high saturation points. It should be also noted that simulations by ISORROPIA-II are underestimated since the system is assumed to be “closed”, where total concentrations in gas and particle phase are fixed and equal to initial concentrations (“batch” system).

We include Fig. S8A and B, and modify the text (Line 23-25, p 9):

“Ammonia emission, which is mainly from agriculture, is as low as SO₂ emission (~ 10 ppb) (Lee et al., 1999), yet it may affect the chemical aging of particles. To investigate the effect of ammonia, we use ISORROPIA-II with the forward mode, which simulates the partitioning from the gas phase. We first simulate nitrate uptake when only NO₂ is present (i.e., [NO₂] = [HNO₃] = 68 ppb) (Fig. S8A). Then we simulate nitrate and ammonia uptake when both NO₂ and ammonia are present (Fig. S8B). Although these simulation results still underestimate the measurements of Seoul haze particles, it clearly demonstrates that the presence of ammonia increases not only ammoniums, but also nitrates in particles. And it also lowers the acidity of particles a little bit from pH 2.7 to pH 3.2. It should be noted that our equilibrium model works

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R1C3) Line 18, p. 9: How the pH was measured?

Response)

We did not measure pH. We estimated pH by using *E-AIM*. Please see our response to R2C2, which includes simulation results from ISORROPIA-II.

R1C4) The discussion in section 4.2 needs further discussion or clearer suggestion since it is against most policy directions used by the countries in Northeast Asia.

Response)

NO_x reduction is generally considered the strategy for PM reduction; however, to be effective, according to our simulation results the significant reduction needs to be achieved. Although HNO₃ uptake into particles is a local process, hygroscopic particles at high concentrations (> 100 µg/m³ at Deokjeok Island) are transported into Korea and these wet particles significantly increase the Henry’s law constant for HNO₃ at Seoul. Therefore, local NO_x reduction at Seoul alone is likely to be ineffective. PM reduction in China is also important; therefore, reduction of VOCs, SO₂, and NO_x in major emitting cities at China also needs to be accompanied. Particularly, inorganic reduction (SO₂ and NO_x) is important because inorganic species (sulfates and nitrates) makes particles hygroscopic, and in turn ALW increases PM mass concentrations substantially by taking up more inorganics and facilitating aqueous chemistry. Therefore, PM reduction strategy needs to be considered in a transboundary perspective (i.e., air qualities in Korea and China should be considered in one regional basis) requiring both Korean and Chinese efforts.

Now the last sentence in section 4.2 (Line 25-27, Page 10) reads:

“... (vehicle exhaust). NO_x reduction is generally considered the strategy for PM reduction; however, to be effective, according to our simulation results the significant reduction needs to be achieved since even a few ppb of NO_x (NO₂) concentration acidifies particles to pH ~ 2 and forms ~ 2 M of nitrates (Fig. 12). Although HNO₃ uptake into particles is a local process, hygroscopic particles at high concentrations (> 100 µg/m³ at Deokjeok Island) are transported into Korea and these wet particles significantly increase the Henry’s law constant for HNO₃ at Seoul. Therefore, local NO_x reduction at Seoul alone is likely to be an ineffective solution. PM reduction in China is also important; therefore, reduction of VOCs, SO₂, and NO_x in major emitting cities at China is also required. Particularly, inorganic reduction (SO₂ and NO_x) is important because inorganic species (sulfates and nitrates) makes particles hygroscopic, and in turn ALW increases PM mass concentrations substantially by taking up more inorganics and facilitating aqueous chemistry. Therefore, PM reduction strategy needs to be considered in a transboundary perspective (i.e., air qualities in Korea and China should be considered in one regional basis), requiring both Korean and Chinese efforts.”

R1C5) Section 4.3 is missing.

Response) It was a typo. Section 4.4 is corrected to Section 4.3.

References

Lee, H. S., Kang, C.-M., Kang, B.-W., and Kim, H.-K.: Seasonal variations of acidic air pollutants in Seoul, South Korea, *Atmos. Environ.*, 33, 3143-3152, [https://doi.org/10.1016/S1352-2310\(98\)00382-3](https://doi.org/10.1016/S1352-2310(98)00382-3), 1999.