FROM: Dong, K., Woo, C., Yamamoto, N., authors of acp-2019-487 "Plant assemblages in atmospheric deposition"
RE: Response to Reviewer #2
DATE: August 2, 2019

The authors thank high-quality comments, especially regarding our sampling strategy, by the Reviewer #2. Please find our responses to the reviewers' comments. The page numbers in our responses refer to those in our revised manuscript.

Reviewer #2:

Comment #1: General Comments:
In the paper the authors examine via genetic techniques the deposition flux of plant derived material in Korea. The topic is relevant and interesting, especially considering the potential climatic interactions of primary biogenic aerosols (PBAPs). Any further insight on PBAPs emissions and deposition is for sure a much needed information. The paper is well written and clear, however the reviewer would like some more clarification about the sampling strategy used to compute deposition fluxes and deposition velocities in the paper.

Response #1: We appreciate the positive appraisal by the Reviewer #2. Regarding our sampling strategy, please see our responses to the Reviewer #2's comments below.

Comment #2: Specific Comments:
Page 3, Lines 5-7: How did the building height compares with the forested area around? Was the sampler located significantly above the treetops? A figure showing the samplers and the sampling location would greatly help.

Response #2: It is hard to generalize since the sampling point is situated in a mountainous area, with considerable elevational variation. In general, however, the building height is below upland parts but above lowland parts of the area. The sentences were revised and added to provide the following local topographical information:

"Briefly, air and deposition samples were collected on the roof (approximately 20 m above ground level) of a building at an altitude of 105 m above sea level in a mountainous, forested area of Seoul in South Korea (37°27'55.0"N; 126°57'17.7"E). The highest peak (632 m) at which sparse trees exist was situated in the south-southeast of the sampling site at a horizontal distance of ca. 2.3 km." Page 3 Lines 5-8

The positional information (37°27'55.0"N; 126°57'17.7"E) was provided to check the topographical information using internet-based tools, e.g., Google Maps. We wish that it is found by such internet-based tools rather than by providing a new figure in order to minimize the space of the paper. The information is available, for example, by accessing to the following link:

URL
https://www.google.com/maps/place/37°27'55.0"N+126°57'17.7"E/

Comment #3: Page 3, Lines 10-11: These lines implies that both deposition and concentration samples were taken monthly. Was the Andersen sampler operated continuously for the month? Were there any issue in saturation of the substrates due to overcollection?

Response #3: The Reviewer #2 is correct. Each sampling continued for a period of 1 month. To prevent from particle overloading, the substrate was rotated once every week for particles to be collected as evenly as possible on the substrate. To clarify, the following sentence has been added:

"The substrate placed onto each stage of the Andersen sampler was rotated once every week to prevent from particle overloading at the same spot under each impactor nozzle." Page 3 Lines 19-21

Comment #4: Page 3, Line 12: What are the specifications of such custom-made sampler? The geometry of the collector do impact the deposition process, so how was this custom made sampler validated? In the reviewer's view these are needed information that are lacking also in the referenced Woo et al., 2018 and Han et al., 2016 papers (referred in Woo et al., 2018 regarding the custom made sampler).

Response #4: Photos of the samplers are available in Supplementary Fig. S1 in Woo et al. (2018). The configuration of the dry deposition sampler is identical to that reported by Yi et al. (1996), while the configuration of the wet deposition sampler is similar to that reported by Landis and Keeler (1997). The following sentence has been added for clarification.

"The configuration of the dry deposition sampler is identical to that reported by Yi et al. (1997), while the configuration of the wet deposition sampler is similar to that reported by Landis and Keeler (1997)." Page 3 Lines 17-19

References

Landis, M. S., and Keeler, G. J.: Critical evaluation of a modified automatic wet-only precipitation collector for mercury and trace element determinations, Environ. Sci. Technol., 31, 2610–2615, https://doi.org/10.1021/es9700055, 1997.

Woo, C., An, C., Xu, S., Yi, S.-M., and Yamamoto, N.: Taxonomic diversity of fungi deposited from the atmosphere, ISME J., 12, 2051–2060, https://doi.org/10.1038/s41396-018-0160-7, 2018.

Yi, S.-M., Holsen, T. M., and Noll, K. E.: Comparison of dry deposition predicted from models and measured with a water surface sampler, Environ. Sci. Technol., 31, 272–278, https://doi.org/10.1021/es960410g, 1997.

Comment #5: Page 3, Line 13: How far were the deposition sampler and the Andersen one? If they were co-located too close to each other, the active air sampling of the Andersen could affect the deposition on the custom made sampler. Again a figure of the sampling setup would greatly help instead of referring to Woo et al., 2018 (in which the figure of the sampler is in the supplementary materials).

Response #5: The deposition and Andersen samplers were placed distant enough to avoid the interference, with approximate horizontal distance of 2.5 m and vertical distance of 2.3 m (Fig. AC2-1 below, which is also included as Fig. S1 in the revised Supplement). The deposition sampler was placed on a wooden raised floor with approximately 2.3 m height from the rooftop, while the Andersen sampler was placed on the rooftop under the raised floor to protect from precipitation, with an additional rain shield.

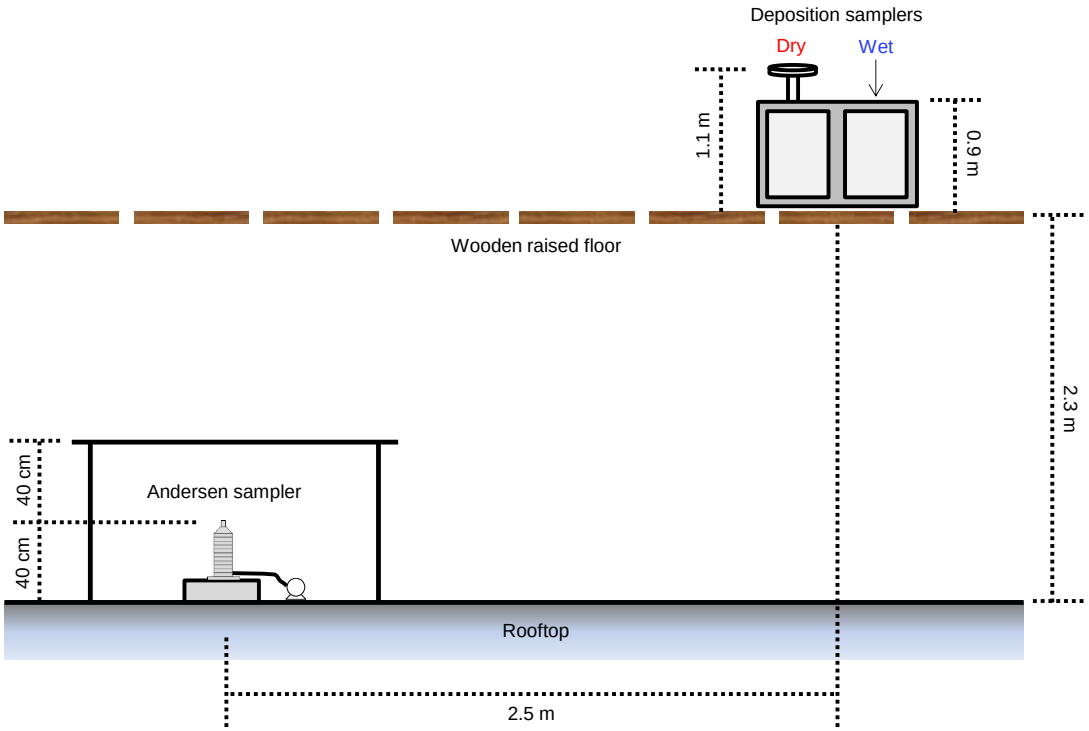

Figure AC2-1: Schematic diagram showing the sampling setup.

In our revised manuscript, the following sentence has been added to clarify our sampling setup:

"The deposition and Andersen samplers were placed distant enough to avoid the interference, with the approximate horizontal distance of 2.5 m and vertical distance of 2.3 m (Fig. S1 in the Supplement)." Page 3 Lines 16-17

Comment #6: Page 4, Lines 11-12: There is a mismatch in the units for flux and concentration. The flux is stated to be reported in CN cm^{-2} month^{-1}, but the concentration is stated to be measured in CN m^{-3}.

Response #6: The dimension of flux density (F) is given by [quantity][area]$^{-1}$[time]$^{-1}$. ($ML^{-2}T^{-1}$), while the dimension of concentration (N) is given by [quantity][volume]$^{-1}$ (ML^{-3}). The dimension of velocity (V) is given by [length]$^{-1}$[time]$^{-1}$ (LT^{-1}). We believe that the physical dimensions in our manuscript are correctly given to provide a following relationship of:

$$V = \frac{F}{N} \quad \left(\because \frac{\mathrm{L}}{\mathrm{T}} = \frac{\mathrm{M/L^2T^1}}{\mathrm{M/L^3}} \right)$$

Comment #7: Page 6, Lines 8-10: That seems an extremely anomalous result, which is not further discussed. How do the authors explain that? Was the rain sampler from which precipitation data are taken sufficiently close to the deposition experiments or was it far away enough to justify local differences in rainfall amounts?

Response #7: The dry and wet samplers were deployed closely enough with the approximate distances of 25–55 cm (please see Supplementary Fig. S1 in Woo et al. (2018)). Therefore, we believe that it was not due to artifacts associated with the distance between the dry and wet deposition samplers.

We do not know why the peak contribution of wet deposition preceded the peak precipitation by one month (i.e., from July to June) (Fig. 3c), which was anomalous as the Reviewer #2 pointed out. We expect, however, that it was in part attributable to the uncertainty of taking monthly averages for the analyses. Within each month, there were both rainy and non-rainy days. It is possible that some species were released, because of its seasonality, more preferentially during a less rainy month of June than during a rainier month of July even though these species were released more preferentially in rainy days. For instance, we found that *Quercus* and *Juglans* were released in a less rainy month of June, but not in a rainier month of July (Fig. 2a), even though these genera were found abundantly in wet deposition (Fig. 5), indicating that these genera were released preferentially in rainy days of a less rainy month of June. To explain such a possibility, the following paragraph has been added:

"It should be noted, however, that several genera was detected exclusively from wet deposition (Fig. 4), and some allergenic genera were detected abundantly from wet deposition, e.g., 60% for *Juglans*, and 32% for *Quercus* (Fig. 5), indicating that these genera might be specifically involved in precipitation. Additionally, we observed that *Quercus* and *Juglans* were released in a less rainy month of June than in a rainier month of July (Fig. 2a), even though they were detected abundantly in precipitation (Fig. 5), indicating that these genera might be released preferentially in rainy days of a less rainy month of June. We expect that the taxon dependency of seasonal pollen dispersals in conjunction with the taxon dependency of rainfall involvement might partially explain our anomalous observation where the peak contribution of wet deposition preceded the peak precipitation by one month (i.e., from July to June) (Fig. 3c)." Page 9 Lines 7-14

References

Woo, C., An, C., Xu, S., Yi, S.-M., and Yamamoto, N.: Taxonomic diversity of fungi deposited from the atmosphere, ISME J., 12, 2051–2060, https://doi.org/10.1038/s41396-018-0160-7, 2018.

Comment #8: Page 6, Lines 27-28: Deposition velocities are computed as the ratio between the deposition sampler and the Andersen one. Given simultaneous measurement of the two it is reasonable to expect that, at least for dry deposition,

the mass collected on one sampler strongly correlates with the mass collected on the other one (Mohan, 2016). A "decoupling" between the samplers could also explain some issues in computed deposition velocities, were some kind of mass-comparison tests performed on the samplers?
Cited References: Mohan S. M. (2016) "An overview of particulate dry deposition: measuring methods, deposition velocity and controlling factors", Int. J. Environ. Sci. Technol., 13:387-402.

Response #8: The suggested analysis was made, with a good correlation (r=0.91) observed between the dry deposition and Andersen samplers (Fig. AC2-2 below).

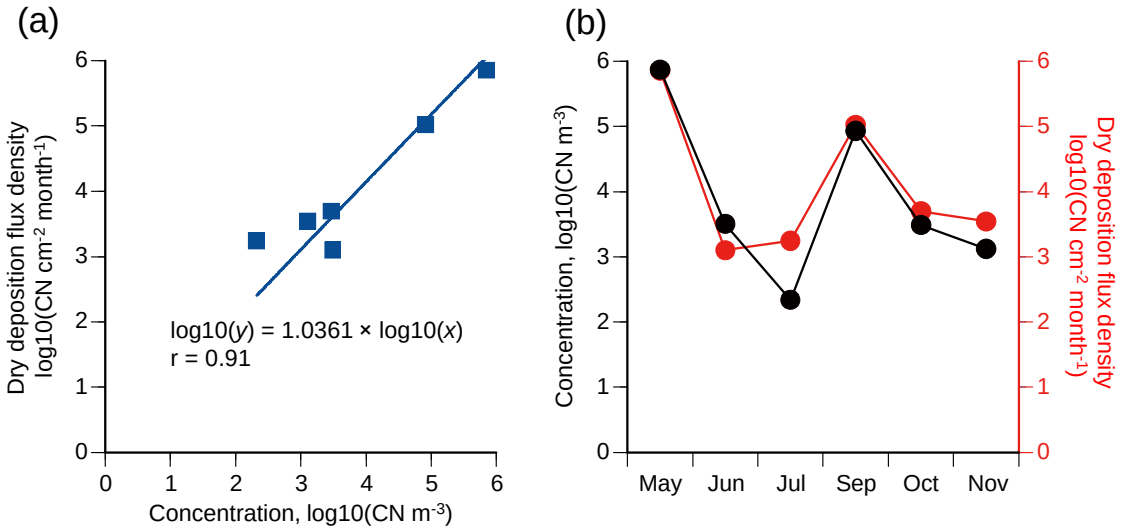

Figure AC2-2: Relationships between flux densities and concentrations of total plants measured by the dry deposition and Andersen samplers, respectively. (a) Scatter plot. (b) Time-series plot.

The result shows the largest between-method variability observed in July (Fig. AC2-2b above). We checked the taxon-specific results, but could not find any systematic tendencies. It is likely because of the intrinsic measurement uncertainty since the similar variabilities were observed for the duplicate measurements of deposition flux densities in low quantity regions (Fig. S2b and c in the Supplement).

In our revised manuscript, Mohan (2016) has been cited.

"The annual dry deposition velocity (V_d) was estimated for each plant taxon according to the following equation (Mariraj Mohan, 2016):" Page 5 Lines 18-20

Comment #9: Page 8, Lines 28-30: The reviewer does not really agree, there's no information in this study to support the actual existence of a taxon-dependent rainout for the sampled pollens, nor to support a prevalence of washout over rainout. The lack of differences between wet and dry deposition samples' structures might also be simply due the lack of any taxon-dependence to rainout,

rather than the more complicated assumption of washout prevalence over taxon-dependent rainout processes. The reviewer suggests rephrasing.

Response #9: We agree that this is our speculation. The sentence has been revised to clarify that it is just a possibility.

"The minimal differences in plant assemblage structures between dry and wet deposition (Fig. 3e) indicated a possibility that washout, which is possibly taxon-independent, predominated over rainout, which is possibly taxon-dependent, for wet deposition of atmospheric plant particles although it is also possible that there is no taxon dependency in rainout." Page 9 Lines 17-20

Comment #10: Page 9, Lines 28-29: Again this is a speculation (see previous comment). The reviewer suggests rephrasing.

Response #10: We agree. It is just a possibility. The sentence has been revised as follows:

"Plant assemblage structures did not differ significantly between dry and wet deposition, indicating a possibility that washout, which is possibly taxon-independent, predominated over rainout, which is possibly taxon-dependent, for wet deposition of atmospheric plant particles." Page 10 Lines 18-20

For the Reviewer #2's 9th and 10th comments above, we replaced the words "likely" with "possibly" since we do not know the likeliness (although we do know it is possible because the taxon dependency was observed at least for fungi). The sentence in the abstract section was also revised accordingly.

"Plant assemblage structures did not differ significantly between dry and wet deposition, indicating a possibility that washout, which is possibly taxon-independent, predominated rainout, which is possibly taxon-dependent, for wet deposition of atmospheric plant particles." Page 1 Lines 16-18