We would like to thank Dr. Andrius Garbaras for his comments that improved the clarity and readability of the manuscript. Our point-by-point responses are found below in blue ink. The revised content is in yellow highlighted.

1. I would like to see more details on the measurement of the isotope ratio in the samples itself. This is actually a research that requires a lot of mastery because of the small amounts of analyte encountered. I would like the authors to provide more details in the supplementary material: what was the linearity of the spectrometer, what smallest samples did the authors measure with sufficient accuracy, or was the linearity tested with international standards of various sizes? All of these details will be useful to readers who apply similar analysis in the future.

A: The following paragraph is added to the supplementary material to provide the required information for readers:

All $\delta^{15}N$ and $\delta^{18}O$ have been analyzed at Ren's lab at Department of Geosciences, National Taiwan University, using 'denitrifier method'. We use denitrifying bacteria strains *Pseudomonas aureofaciens* for $\delta^{15}N$ and $\delta^{18}O$ analyses on nitrate samples, and *Pseudomonas chlororaphis* for $\delta^{15}N$ analyses on total N samples after oxidizing reduced N forms to nitrate. The analytical errors for $\delta^{15}N$ and $\delta^{18}O$ of nitrate samples are generally smaller than 0.1‰ using the 'denitrifier method' for samples containing 5 nmol N or more (Fig. S9). The errors become slightly bigger with smaller samples, e.g., at 2 nmol N. As a result, we have only analyzed samples with 5 nmol or more N. The linearity on the current setup is within 0.2‰ between 5nmol and 20 nmol of N. But this does not affect our analyses, since we can correct for the linearity effect by analyzing samples and standards with constant N levels. Prior to isotopic analyses, we measure N concentration in each sample, so we could estimate the volume of samples needed to yield constant N amount (i.e., 5 nmol N). In addition, these samples are analyzed with standards at the same N level, such that any linearity effect will be sufficiently corrected. In addition, samples with very low nitrate concentration (less than 0.5 µmol/L in the dissolved solution) have greater errors for $\delta^{18}O$ analyses due to oxygen exchange effect with water during nitrate conversion. As a result, we only analyze samples that can yield greater than 1 µmol/L nitrate in the final dissolved solution. Samples or sample sizes will be binned if there is not sufficient N on each filter. Furthermore, we analyze samples with standards of similar concentration range. For example, samples with 7 µmol/L nitrate are analyzed with standards of 5 and 10 µmol/L nitrate, so the data correction using the nitrate standards also excludes uncertainties with different nitrate concentrations among samples. The above procedures are applied to all samples, which intend to address most if not all the uncertainties associated with isotopic analyses on nitrate samples. For total dissolved nitrogen, we use potassium persulfate reagent (3 g of Persulfate potassium and 5 g of Sodium hydroxide in 100 ml of Milli-Q water) to oxidize reduced N to nitrate prior to isotopic analyses. The main source of uncertainty in this oxidation step is associated with the blank of potassium persulfate reagent. We account for this uncertainty by using purified potassium persulfate after 3 times of recrystallization, which typically yield blank size of 0.4 µmol/L N, and account for 6% of the total oxidized sample on average. In addition, we also process 5 blanks and 3 to 4 oxidation standards using international standards USGS 40 ($\delta^{15}N$= -4.52 ‰) along with each run (typically containing less than 30 samples). The oxidation standards cover the range of blank/sample ratio in the samples, so we could also correct for blanks. Although we did not perform duplicates for the oxidation plus isotopic analyses on our samples, the 1 standard deviation for oxidation standards is less than 0.21 ‰, which represents the uncertainty for isotopic analyses for oxidized TN samples.

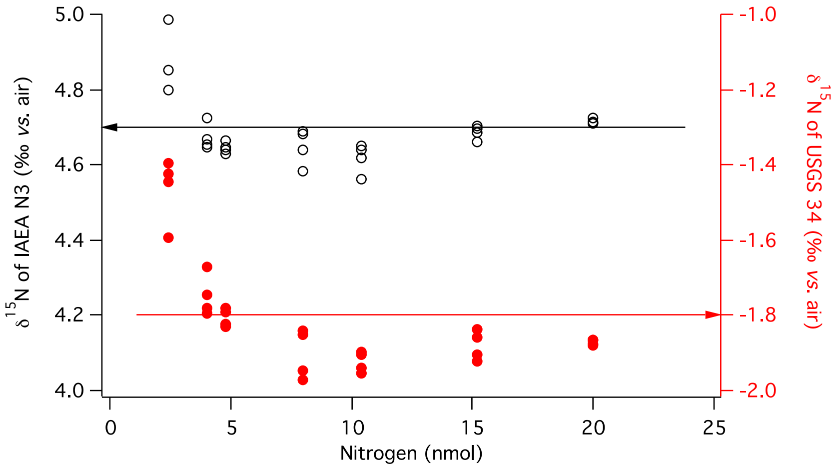

Figure S9. Measured δ^{15}N of IAEA N3 (open black circles) and USGS34 (closed red circles) at different nitrogen levels. The black and red lines indicate the true values of the two standards. 1std of δ^{15}N at each nitrogen level is ~0.1 ‰. The changes in the measured δ^{15}N at different nitrogen levels reflect the current linearity of the system, which would be corrected with standards.

Some specific comments:

2. Line 75 It's not clear where samples were collected. It's written that in Xitou experimental forest, but is not clear the location is up in the hill or in valley.

A: Xitou experimental forest is located in a valley as shown in Figure R1. The content is revised as: "A field campaign was conducted over Xitou experimental forest (23°40'12" N, 120°47'54'' E, 1,179 m a.s.l.) in a valley from 1st to 24th December 2018 to investigate the interaction between air quality, local circulation, and human activities in central Taiwan."

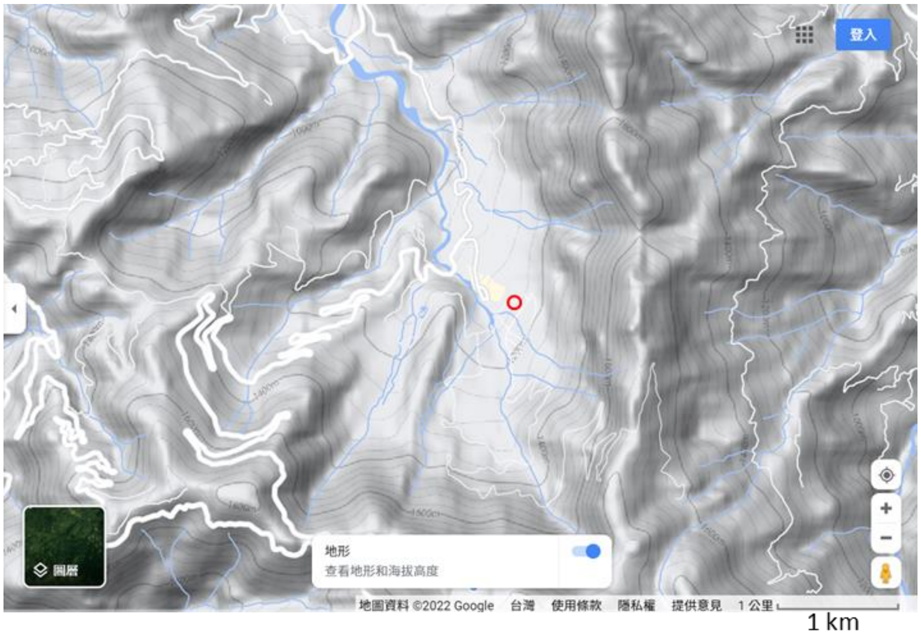

Figure R1. The topographic map nearby Xitou Experimental Forest (adapted from Google Maps). The red circle is the sampling location.

3. Line 105. There is no description how BC was measured with FTIR-ATR analysis. Does it is comparable with the measurements with other BC techniques, for example aethalometer?

A: Because BC absorbs broad radiation, the absorbance of BC was determined by the average absolute absorbance in the region of 3950 ± 5 $\mathrm{cm^{-1}}$ where the interference by other chemical species is negligible, as shown in Fig. S1 (the whole baseline shifted up). The calibration of BC absorbance at 3950 ± 5 $\mathrm{cm^{-1}}$ was performed in the earlier study (Huang, 2016), with the elemental carbon concentration determined using a DRI2001A carbonaceous aerosol analyzer, following the IMPROVE thermo-optical reflectance (TOR) protocol (Chow et al., 2001), as detailed in Chou et al. (2010). The BC measurement is clarified with the following statement added to the end of section 2.2: "As to black carbon (BC) concentration, the absolute absorbance at 3950 ± 5 $\mathrm{cm^{-1}}$ is applied to quantify the BC concentration based on the calibration done by Huang (2016) with the elemental carbon concentration determined using a DRI2001A carbonaceous aerosol analyzer, following the IMPROVE thermo-optical reflectance (TOR) protocol (Chow et al., 2001), as detailed in Chou et al. (2010).".

References:

Huang, R.-T.: A study of aerosol hygroscopicity in Kinmen, Graduate Institute of Atmospheric Sciences, National Taiwan University, Taipei, Taiwan, 10.6342/NTU201603559, 2016.

Chow, J. C., Watson, J. G., Crow, D., Lowenthal, D. H., and Merrifield, T.: Comparison of IMPROVE and NIOSH Carbon Measurements, Aerosol Science and Technology, 34, 23-34, 10.1080/02786820119073, 2001.

Chou, C. C.-K., Lee, C. T., Cheng, M. T., Yuan, C. S., Chen, S. J., Wu, Y. L., Hsu, W. C., Lung, S. C., Hsu, S. C., Lin, C. Y., and Liu, S. C.: Seasonal variation and spatial distribution of carbonaceous aerosols in Taiwan, Atmospheric Chemistry and Physics, 10, 9563-9578, 10.5194/acp-10-9563-2010, 2010.

4. Line 135. What stands for letter p in "p-NO3 -=…"

A: The letter p stands for particulate phase. However, in this study, only particulate NO_3^- is discussed. The letter "p-" is deleted in the revision.

5. Line 170. Fig. 2(c) and 2(d). NH4+ is not in the Fig. 2(d).

A: The reviewer is correct. Figures 2(c) and 2(d) are for NH_4^+ and NO_3^-, respectively. The content in Line 170 is revised as " The mass distribution seems to shift to a larger size bin (0.56-1.8μm) for NH_4^+ as shown in Fig. 2(c), while NO_3^- in Fig. 2(d) has a significantly high concentration for the 0.56-1.8μm size bin during the foggy period."

6. Line 170. It's not clear boundary level effect. Does it mean that the boundary level is always above the sampling station?

A: During daytime, the land was heated by solar radiation, causing boundary layer height to rise to a higher altitude (~1-2 km). The daytime boundary layer height mostly above the sampling site. The foggy period is likely associated with a stronger boundary layer inversion, which has a lower boundary height but is still above the sampling site. However, the sample site is likely below the nighttime boundary layer height as it was estimated to be less than 600 m a.s.l. based on the radio-sounding measurements at the foot of the hill nearby. We add the following sentence to the end of section 3.1.1 for clarification: "The sampling site is mostly below the boundary layer height during daytime and above the boundary layer height during nighttime."

7. Line 190. I look at Fig. 3a and I see on average lower $\delta^{15}N$ values in submicron range comparing to bigger particles. Authors say that the "trend of a higher NH_4^+ $\delta^{15}N$ in submicron aerosol was also observed in Beijing". I do not understand how Authors compare different size bins.

A: The statement "a higher NH_4^+ $\delta^{15}N$ in submicron aerosol" didn't provide an accurate description. For a given collection period data, $\delta^{15}N$ values generally show an increasing trend first and then a decreasing trend with particle size. The maximum $\delta^{15}N$ happens around the 0.56-1 µm size bin for most non-foggy daytime. The sentence is revised as "The increasing and then decreasing trend of NH_4^+ $\delta^{15}N$ with aerosol size was also observed in Beijing…" to provide a more accurate illustration.

8. Line 195. What is mean "daytime fractionation"?

A: We use "daytime fractionation" to describe the fractionation that happened during daytime. To avoid confusion, the content is modified as "As the mountain wind dominates after sunset, available NH_3 might be attributed to the daytime residual (having lower $\delta^{15}N$ due to the fractionation that happened during daytime) or the local biogenic sources having a lower $\delta^{15}N$."

9. Line 200. PM1-10 was higher … similar to 0.32-1 µm aerosol. So no difference in all size bins, as almost the whole range fall in the 0.32 – 10 µm. This kind of differentiation seems artificial.

A: Yes, the NH_4^+ $\delta^{15}N$ of foggy daytime is relatively flat at a diameter larger than 0.56 µm. However, $\delta^{15}N$ for PM_{1-10}-NH_4^+ at foggy daytime is higher than that at non-foggy conditions. It might be attributed to the growth of part of 0.56-1 µm aerosols under high RH. To improve the clarity, the content in this paragraph is revised as follows: " Fog varies the composition mass distribution among different size bins and can affect the isotopic ratio. Under foggy daytime conditions, the $\delta^{15}N$ value of larger size aerosols (PM_{1-10}-NH_4^+) was more like to be the extension of 0.56-1 µm with a value up to 21.39‰, higher than that of non-foggy days. As stated in section 3.1, high NH_3 concentration can promote the partition of HNO_3 during foggy conditions to enhance hygroscopicity. The observed flat trend of $\delta^{15}N$ at dimeter $\geq$ 0.56 µm might result from the hygroscopic particle growth of NH_4^+ from the 0.56-1 µm size bin aerosols. "

10. Fig. 2. The legend must be revised. I suggest adding a legend to the (b) and (d) for clarification.

A: Thanks for Dr. Garbaras' comment. We added a legend to Fig. 2(b) and (d) and adjusted the legend location for clarification. The updated figure is as follows: