Author's response to Referee #1

General Comments to the Author

The author reports two years of NHx and NH_4^+ data collected in Nagoya, Japan, and uses it to infer local sources of NH_3, such as traffic, plant stomata, soil pore water, and bird droppings. Observations of NH_3 are consistent with other studies (e.g., daytime maximum, strong seasonal variations), and data are presented in a fairly clear manner. The most novel part of the manuscript is the finding that bird droppings could be a relevant local source of NH_3 in urban areas. Although the manuscript is fairly well written, phrasing and grammar could be improved throughout the manuscript. Portions of the data analysis and discussion could be expanded to improve the manuscript, as noted below. Nonetheless, this measurement report contains valuable insight for understanding urban ammonia sources, and publication is recommended once the comments below are addressed.

General Comments:

Most of the data analysis only considers parts of the data set (e.g., July/December 2018, and RH < 70% when wind speed < 3 m/s). A more holistic look at the entire data set might give additional insight on various sources.

For example, the morning NH_3 peak ~2-4 hours after sunrise is decoupled from the maximum ambient temperature which is inconsistent with bi-directional exchange (i.e., stomata and soil) driving NH_3 emissions, since these emissions should peak with temperature. Is it possible the lack of a coincident peak of NH_3 and temperature is caused by enhanced vertical mixing (i.e., dilution) later in the day?

Furthermore, examining days with presumed surface wetness (i.e., RH > 70%) might provide insight on whether or not the morning peak in Fig. 4 (top left) is related to evaporation of surface wetness. In other words, the different peak times for NH_3 and temperature, as well as the impact of surface wetness evaporation should be explored further.

Response:

I thank anonymous Referee #1 for constructive comments exploring the cause of the morning NH_3 peak described in the manuscript. I asked a native-English proofreader to review the revised manuscript, particularly for clarity in preference to subjective style. Modified words and sentences in the text have been highlighted as yellow in the manuscript.

To cover a longer span and details of seasons, I modified Fig. 4 including the monthly average of diurnal variations for every 3 months from December 2017 through September 2019. I also added a new Fig. 5 to discuss the effects of direct sunshine on the morning peak. Related to the new figures, I have added additional discussion of the boundary layer height (BLH). Unfortunately, data related to BLH were not available for this study. I agree that dilution effects

during daytime reduce the NH_3 concentration. Diurnal variation of NH_3 concentrations for cloudy days shows a coincident peak with air temperature. In addition, strong effects of the inversion layer on NH_3 concentration must be limited for days of calm winds in winter, when the morning peak was absent.

Specific comments

Comment #1:

Line 108 – what was the measurement height above the ground?

Response:

The inlet height was ca. 26 m above the ground. I have added this explanation to the text and to the caption of Figure 1d.

Comment #2:

Lines 108 to 124 – what is the approximate residence time of the air sample, and distance it travels from the inlet, before it comes into contact with the water droplets (i.e., is dissolved)? Is it possible that some relevant fraction of NH_3 partitions to the surface of the sampling inlet, which could desorb later at high temperatures and/or lower NH_3 concentrations? In other words, has collection efficiency of the system been tested?

Response:

The distance from the end of the denuder glass tube to the mixing point of water droplets is approximately 10 cm. Because the sample air is transported through the PTFE tube (id: 3 mm) at a flow rate of 1 L min^{-1}, the residence time from the end of the denuder to the mixer is ca. 0.04 s, which means that water droplets immediately contact with the sample air after leaving the denuder glass tube. The collection efficiency of the system was higher than 95% for the condition employed in this study. I added these points to the revised text.

Comment #3:

Lines 144 to 145 – presumably diurnal variation in wind speed is not as clear in winter time due to the lack of sea breeze circulations, although the current phrasing implies a direct link between sunlight and wind speed. Recommend rephrasing to clarify that it's not sunlight that's directly impacting wind speed.

Response:

I revised this part of the text as suggested.

Comment #4:

Section 3.1 – the analysis focuses on only two months (July and December 2018). Is there a reason that more months weren't considered (e.g., Dec 2017, July 2019, Nov/Jan, June/Aug) when trying to interpret seasonal differences? Considering these additional months would likely make the analysis more representative of the winter/summer seasons.

Response:
To present more data, I renewed Fig. 4 to include the monthly average of diurnal variations for every 3 months from December 2017 through September 2019.

Comment #5:
Section 3.2 – there is a lot of discussion about mist/droplet pH; however, the impact of pH on NH_3 release from evaporating mist/droplets is not made clear. It would be helpful to provide a few sentences explicitly stating how NH_3 emissions from droplets are impacted by pH.

Response:
Additional explanations of chemical composition and pH were added to the manuscript, highlighting effects on NH_3 emission after the evaporation of mist droplets.

Comment #6:
Lines 226 to 227 – the assumption is that the air being sampled before sunrise under low wind conditions reflects local sources. However, is it possible that the nocturnal boundary layer is sufficiently shallow during these times, such that the sampling inlet on the 7th floor is above the nocturnal boundary and is decoupled from surface sources?

Response:
Although a detailed time evolution of the nocturnal boundary layer was not known for the site, the sampling inlet (26 m above the ground) is presumed to be located well within the boundary layer. I added discussion specifically related to the controlling factors of NH_3 concentration related to the boundary layer height, dry deposition of NH_3, and local emissions.

Comment #7:
Lines 303 to 306 – the description of crow abundance and behavior is very anecdotal. A more detailed description on what is meant by terms like "visual impression" and "rarely observed" would be useful.

Response:
I modified the sentence related to appendix photograph 1 and deleted the sentence that used

“rarely observed” from the revised manuscript.

Comment #8:
Line 322 – is this a unit conversion error (3.4 mol m^{-2} day^{-1} to 100 mmol m^{-2} $month^{-1}$)?

Response:
I corrected the unit of deposition (3.4 mmol m^{-2} day^{-1}).

Comment #9:
Figure 1b – please add a scale for distance.

Response:
I added a scale for distance in Figure 1b to the revised manuscript.