Authors' Response

In their revised manuscript, the authors acknowledged the limitations of their measurement technique, and provided additional details regarding the data analysis. However, as pointed out by both reviewers, many of the conclusions drawn by the authors in this work cannot be supported by the observations alone.

RE: Every instrument has advantages and disadvantages. We added this statement in the conclusion (Lines 363-372):

"Measuring ammonia and amines in the atmosphere is one of the most challenging areas in the development of atmospheric analytical instruments [*S.-H. Lee*, 2022; *Shan-Hu Lee et al.*, 2019]… Very importantly, our CIMS, despite its relatively low mass resolution, has measured ammonia and amines at various atmospheric conditions, ranging from the rural forests [*Kanawade et al.*, 2014; *You et al.*, 2014], a relatively less polluted site [*Mark E. Erupe et al.*, 2010; *You et al.*, 2014; *Yu and Lee*, 2012], to the extremely polluted urban environment (this study), with consistent instrument sensitivities over the decade; to our knowledge, this is the only instrument that demonstrated such consistency in the performance."

Previous research has demonstrated that NH3 and amines were often present in the same emission sources (Ge et al., AE 2011). Therefore, it is not surprising to see the good correlations between NH3 and amines in this work. While it is a practical idea to use NH3 as a proxy for amines to surrogate the missing emission inventory of amines, the NH3 proxy will only be meaningful if it represents a specific emission source, such as vehicular exhausts, biomass burning, and domestic waste management. The emission ratios of amines concerning NH3 reported by this work (see Line 314) were measured at a receptor site, which would be affected by the type of sources present, photochemical aging experienced by the air mass before it arrived at the site, and other physical processes (such as gas/particle partitioning, especially in the case of NH3). Therefore, the reviewers are of the opinion that the observations do not support the suggestion of using 0.1% of NH3 as a proxy of DMA in NPF modeling.

RE: We are not doing emissions studies, so whether it is near the sources or in the aged air is NOT relevant to the ambient concentration measurements, or to the proxy of the dimethylamine concentrations compared to ammonia. We added this paragraph in Atmospheric Implications section (Lines 324-333):

"From these observations made in very polluted Houston and less polluted Kent, we propose that at the polluted sites in the United States, dimethylamine concentrations can be estimated using the proxy, 0.1% ammonia concentrations. The caveat of this proxy is based on only two locations in the United States and did not consider different emission sectors. There has been so far only one

attempt to use quantify the aerosol nucleation processes using sulfuric acid and dimethylamine in the global model by Zhao et al. (Zhao et al., 2024) and they concluded that this nucleation process is dominant globally in polluted boundary layer, from China, India, Europe to United States. In this cited study the authors used the proxy of dimethylamine using ammonia concentrations, for example, dimethylamine/ammonia ratio of 0.0070, 0.0018, and 0.0100, for chemical industrial, other industrial, and residential sources; these proxies were derived by Mao et al. (Mao et al., 2018) based on the measurement made in Nanjing (Zheng et al., 2015). Our observations indicate that (Zhao et al., 2024) likely overestimated dimethylamine concentrations for polluted sites in the United States overall, and thus overpredicted nucleation rates as well. Thus, our results can provide more constrained proxy for polluted sites in the United States for future modeling studies."

We also revised the abstract (Lines 25-29):

"Our observations made in very polluted Houston, as well as a less polluted site (Kent, Ohio) from our previous study (You et al., 2014), indicate there is a consistent ratio of dimethylamine over ammonia at these two sites. Thus, our observations can provide a relatively constrained proxy of dimethylamine using 0.1% ammonia concentrations at polluted sites in the United States to model NPF processes."

Additionally, the reviewers are of the opinion that statements regarding the "pronounced diurnal cycles and temperature dependence of NH3 and amines" are not conclusive. All the provided explanations were possible, but no specific evidence was provided to support any of them. Particulate phase chemical compositions are needed to support the gas/particle partitioning assumption. At the very least, a thermodynamic simulation (such as an E-AIM model) should be carried out to support the statement of the "gas/particle partitioning process."

RE: AIM does not have amines, so this request is not reasonable. And it is not possible to make AIM calculations without other key species, such as organic and inorganic acids. We revised the first paragraph of the conclusion (Lines 341-349) as the following:

"Our observations in urban Houston show that ammonia and amines generally followed a clear diurnal cycle, peaking in the early afternoon when the ambient temperature was highest during the day. We found a correlation between ammonia/amines and ambient temperature. The diurnal cycles and temperature dependence of these compounds are consistent with [*You et al.*, 2014] which showed that the gas-to-particle conversion contributes to the temperature dependence. To verify this process quantitatively, the chemical composition of particle is needed, but particle measurements were not available during the present study. Additionally, the observed temperature dependence could be due to increased emissions of ammonia and amines from biogenic and anthropogenic sources. On the other hand, photochemical aging that occurs typically during the higher solar flux can also reduce the gas phase amines at the noontime; thus,

photochemical aging was unlikely the main driving factor to produce higher concentrations of amines around noon."

Therefore, I recommend that the authors further revise their manuscript, with a focus on not over-interpretating their observations given the limitations of their measurements.