

Interactive comment on “Single-particle experiments measuring humidity and inorganic salt effects on gas-particle partitioning of butenedial” by Adam W. Birdsall et al.

Anonymous Referee #1

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This manuscript reports a study on the evaporation kinetics of butenedial using single particle levitation. The hypothesis is that the hydration of butenedial results in a low vapor pressure form that will evaporate more slowly than the non-hydrated form. This effect has been studied for other compounds (glyoxal, methylglyoxal etc.) but larger dicarbonyls, such as butenedial, have received little attention despite its observed presence in the atmosphere. Hydration will lead to much larger Henry's law uptake coefficients, with implications for gas-particle partitioning in the atmosphere.

This is particularly interesting and well-motivated research and the single particle MS technique reported has a lot of potential. However, I believe this is also one of the

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flaws of the study – the technique itself is poorly suited to the measurements being made. That does not discredit the observations or invalidate the findings, but the large uncertainties are primarily a consequence of the experimental strategy, and similar methods could have produced data with clearer trends.

Overall, the paper is very well written and the work addresses the hypothesis, concluding that even under dry condition, the butenedial exists in the hydrated form. I recommend this for publication, although some minor points should be addressed as detailed below:

- 1) Please clarify the term “effective vapor pressure” – as far as I can tell, it is the vapor pressure that you'd calculate if you assume the activity coefficient to be unity. However, the description on Page 2 lines 15-20 is confusing.
- 2) Other single particle MS methods have been reported (Jacobs et al. 2018, for example) and should be cited.
- 3) The height of the pulse is used to quantify the abundance – what happens when you use the area of the pulse? Peak height is much more susceptible to peak shape effects (as evidence by the change in going from 1 Hz to 3 Hz) but peak area may be more robust.
- 4) What is the precision in the spring point method? Asked another way, how reliably can two droplets of a similar size be segregated by size?
- 5) The PEG is very hygroscopic and will drive the uptake of water at higher RH. It may also effectively hold on to water at low RH, possibly influencing the observations. Were measurements made with pure butenedial to verify if a faster rate of evaporation was observed under dry conditions?
- 6) The size of the particle is really key for these measurements given that only one component is evaporating. Typically, vapor pressure measurements are made from looking at the evolving size and then fitting these data to a model. It is unclear in

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this study why MS was used as the sole measure of the amount of volatile material, when simple light scattering measurements would allow the size change to be resolved with high precision and accuracy. Clearly, for multi-component evaporation, the MS technique would be beneficial.

7) Figures 6 and 7 reveal a troubling amount of scatter in the measurement, demonstrating that even using an internal standard, a consistent measure of the composition is not possible. While vague trends are apparent, the uncertainty in the fit must be large. It is not clear if this is accounted for in the reported vapor pressure value. Please clarify. The MS response is not discussed as a source of uncertainty in 3.3, but clearly this is one of the more major sources. Droplets of different radii should exhibit the same relative intensity using the internal standard, so the variation seen in Figures 6 and 7 is an additional factor relating to the response of the instrument.

8) The evaporation rate is proportional to radius-squared, so an uncertainty of up to 50% could lead to an error in vapor pressure by a factor of 4.

9) I would like to see measurements reported for pure butenedial under dry and high RH conditions. Even if the data is crude due to lack of internal standard, this seems like a key measurement to aid in interpretation of the other data.

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