

Interactive comment on “New insights into atmospherically relevant reaction systems using direct analysis in real time-mass spectrometry (DART-MS)” by Yue Zhao et al.

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Received and published: 21 February 2017

We thank the reviewer for the helpful comments on the manuscript. Our point-to-point responses to each comment are as follows (reviewer comments are in black, and author responses are in blue). This is a well written manuscript reporting on the application of DART-MS for in-situ analysis aerosolized particles of C3-C7 dicarboxylic acids reacted with gas-phase amines and laboratory SOA generated by ozonolysis of alpha-cedrene – all are model systems relevant to atmospheric aerosols. The results show that DART-MS has a good potential for molecular-level analysis of aerosol mixtures with enhanced sensitivity to shallow surface (~ 30 nm) layer of particles. Complemented by more traditional AMS detection of the whole particle volumes, DART-MS

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can provide unique information on the surface chemistry of particles. The presented results are thoroughly evaluated in a context of available literature reports and are convincing. Overall, this is an accomplished work in all its aspects including scientific impact, original measurements, methodology development, and presentation quality. I recommend this paper for publication. Below, I list a few minor notes to consider in the revised manuscript. Response: We thank the reviewer for the positive evaluation of the manuscript. I think that quantitative estimate of the surface layer (~ 30 nm) probed by DART is a very important result that needs to be included in the abstract. Response: We have included a sentence “Results show that DART-MS probes ~ 30 nm of the surface layer” in the abstract. Line 24: ‘particles’ after (SOA) can be removed Response: We have removed it. Line 33: ‘However’ is not needed. Response: We have removed it. Eq (1): it needs to be noted that eq 1 assumes the same effective ionization efficiency for gas-phase and particle phase amines that likely won’t be always correct. Response: We have added a sentence “assuming the same ionization efficiency for the gas-phase and particle-phase amines” before the sentence “the fraction of amine taken up by the particles (fp) can be derived from Eq. (1)”. Line 205, eq 2, and then throughout the text: it is more common to use ‘normalized surface area’ rather than ‘surface area normalized’. Response: In this paper, the particle-phase fraction of amines is normalized to the surface area of the particles. We think the expression “surface area normalized ...” is more appropriate than “normalized surface area ...”. For clarity we have modified the sentence on lines 206-207 to read “Thus, the particle-phase fraction of amine taken up by the diacid, normalized by surface area, (Fp) is given by:...” Line 293: consider change of ‘explanation is the relative: : :’ to something like ‘a possible explanation can be suggested based on differences in the relative saturation vapor pressures’ Response: We have changed “a possible explanation is...” to “a possible explanation can be suggested based on differences in...”.

We have also made some minor editorial changes for clarification in several places in the manuscript: Page 7 line 145-146: The following was added “The SMPS was operated with a sheath flow of 3 LPM and an aerosol flow of 0.3 LPM.” Page 7 lines

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170-171: Details of the sheath and aerosol flows were added so that it reads “Size distributions of SOA particles formed in the flow reactor at the two residence times (44 or 27 s) were also measured using SMPS (sheath and aerosol flows were 15 LPM and 1.5 LPM, respectively). Page 7 lines 171-173: For clarification, the sentence was changed to read: “Typical surface weighted geometric mean diameters ($D_{\text{g,S}}$) were measured to be 28 nm and 21 nm, and number weighted diameters were ($D_{\text{g,N}}$) of 24 and 16 nm, respectively.” Page 8 line 185: “is” was replaced with “was” so that it reads “The DART probe was placed at the entrance of the MS...” Page 12 lines 303-305: “data” was changed to “value” and “suggest” was changed to “suggests” so that it reads “the F_p value (Fig. 2) for the BA reaction suggests that...” Page 13 lines 328-330: The unit of “ g cm^{-3} ” was removed from line 328 and was added to each density value so that it reads “amine-reacted diacid particles, which is assumed to be the same as the solid diacid samples (i.e., 1.619 g cm^{-3} for malonic acid, 1.429 g cm^{-3} for glutaric acid, and 1.329 g cm^{-3} for pimelic acid...)” Figure 2 caption: “fraction” was added so that it reads “Surface area normalized fraction...” Figure 3 caption: “Surface area normalized fraction” replaced “Particle phase fraction” so that it reads “Surface area normalized fraction, F_p , of...”

We have also made some minor editorial changes for clarification in the Supporting Information: Page 2 line 41: “ F_p ” has been replaced with “ f_p ” Figure S2a: Reformatted the y-axis. Page 6 line 78: “and” was replaced with “or” so that it reads “particle stream or standard solutions” Page 6 line 78 added after “standard solutions,” “but the ammonium adduct of the diacid was observed due to the ubiquitous presence of NH₃ in room air.” Figure S4: Peak labelling was reformatted and the ammonium adduct label was added to Fig. S4a.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-357, 2016.