

The authors have addressed most of my previous comments. There are still a couple major issues that needs to be clarified. The first one left me more confused than before. In addition, some minor comments should be addressed, too.

Major comments

1) The authors state in their response and also ion the revised manuscript (p. 4, l. 5) “During the cloud event, the collected fine aerosols were most 5 of the interstitial aerosols, together with some residual particles of smaller droplets < 3 μm .”

By definition, interstitial particles are those particles that are not activated into cloud droplets, cf, for example in standard textbooks, e.g. (Seinfeld and Pandis, 2006)

p. 790: ... The maximum supersaturation reached inside a cloud/fog is an important parameter. Particles with critical supersaturations lower than this value will become activated and become cloud droplets. The rest remain close to equilibrium but never grow enough to be considered droplets and are called interstitial aerosol.

p. 794: First, a fraction of the aerosol distribution is activated and becomes cloud droplets while the rest remains as interstitial particles.

Thus, interstitial aerosol particles are usually too small and/or of too low hygroscopicity to act as nuclei for droplets. Thus, they do not become modified by chemical processes in cloud droplets. Interstitial aerosol usually does not make up a major mass fraction but a major particle fraction. However, only collecting drop residues from drops with a size of 3 μm will miss a major fraction of the droplets, i.e. those particles that are likely most significantly altered by chemical cloud processing. It has to be clarified what exactly you collected and how it was done. As it is written now, i.e., discussing significant sulfate and DOM formation in interstitial aerosol resulting in droplet mode particles is not consistent with the concept of chemical cloud processing which occurs in droplets.

2) The authors claim that the subsaturation of the small monoaldehydes is likely a consequence of their fast oxidation in the aqueous phase. However, comparing the rate constants with OH in the aqueous phase for the compounds listed in Table S4 (all data taken from CAPRAM (Herrmann et al., 2005), I do not see any significant difference that could explain the trends in partitioning.

	$k_{\text{OH(aq)}} / \text{M}^{-1} \text{s}^{-1}$
Glyoxal	1.1e9
Methylglyoxal	7.9e8
Formaldehyde	1e9
Acetaldehyde	3.6e9
Acetone	1.7e8
Propanal	2.2e9

Thus, I suggest removing the rather vague statement regarding oxidation reactions as a reason for subsaturation. This would be only true if for a given compound the transport from the gas phase is slower than the consumption reaction in the aqueous phase. However, this effect is more important for highly soluble compounds rather than for less soluble ones (Ervens et al., 2014) so that this cannot explain either the observed trend in partitioning.

Minor comments

p., 1, l. 14 (and later in the manuscript): It should be ‘acetone’ – unless you also measured its substituted forms.

p. 7, l. 16: This is much closer to a factor of 6 not 5 (0.08 vs 0.47).

p. 7, l. 25: ‘tens’ should be ‘ten’

p. 10, l. 26: ‘than 1.0’ should be removed here

p. 12, l. 10: The study by Waxman et al was also performed for ambient particles. I suggest rewording this sentence to

‘...although the salting-in/out effect is of particular importance in the effective uptake of carbonyl compounds by ambient particles (Shen et al., 2018; Waxman et al., 2013) where solute concentrations are high.’

p. 13, l. 3: *“In contrast, the less-soluble monocarbonyls are still supersaturated.”* This sentence seems redundant as the same is discussed on the previous page.

p. 15, l. 3”’performed’ should be replaced by ‘shows’ or ‘exhibits’

p. 15, l. 17: *“Cloud processing can efficiently remove aerosol particles from the air by nucleation scavenging and impaction scavenging, especially at the initial stage of cloud events.”* This text is not correct. The scavenged particles are still in the air and not all scavenging processes will result in precipitation.

p. 15, l. 18/19: *“At the same time, chemical cloud processing greatly favors the in-cloud formation of sulfate.”* This sentence is not clear either. ‘At the same time’ implies that you are still talking about the initial stages of cloud events; however, it is more likely that sulfate formation occurs more efficiently in grown droplets, i.e. in later stages. Do you simply want to say that sulfate formation in clouds is globally more significant than in the gas phase?

p. 17, l. 4/5: While you explained in the response that and why the sulfate and DOM loss rates are not additive, this should be also added here as otherwise it is not clear why the sum of the individual loss rates (0.47 ug/m³h + 0.28 ug/m³/h) exceeds the total PM loss rate (0.46 ug/m³h).

p. 17, l. 19: Usually sulfate formation in clouds occurs under oxidant (i.e. H₂O₂)-limited conditions, e.g. (Ervens, 2015). Even in the cited study, Shen et al. (2012) state that “The measured aqueous cloud water H₂O₂ concentration was not considered appropriate to use in these calculations because it can be rapidly consumed by reaction with S(IV)...”

A better reasoning of the lack of a significant competition for H₂O₂ by carbonyls vs SO₂ is the study by Schöne and Herrmann (2014) that showed that the reactions of carbonyls with H₂O₂ are likely negligible under cloud conditions.

p. 21, l. 10: The contributions by H. Herrmann should be added to the list.

References

Ervens, B.: Modeling the Processing of Aerosol and Trace Gases in Clouds and Fogs, *Chem. Rev.*, 115(10), 4157–4198, doi:10.1021/cr5005887, 2015.

Ervens, B., Sorooshian, A., Lim, Y. B. and Turpin, B. J.: Key parameters controlling OH-initiated formation of secondary organic aerosol in the aqueous phase (aqSOA), *J. Geophys. Res. - Atmos.*, 119(7), 3997–4016, doi:10.1002/2013JD021021, 2014.

Herrmann, H., A. Tilgner, P. Barzagli, Z. Majdik, S. Gligorovski, L. Poulain and A. Monod: Towards a more detailed description of tropospheric aqueous phase organic chemistry: CAPRAM 3.0, *Atmos. Environ.*, 39, 4351–4363, 2005.

Schöne, L. and Herrmann, H.: Kinetic measurements of the reactivity of hydrogen peroxide and ozone towards small atmospherically relevant aldehydes, ketones and organic acids in aqueous solutions, *Atmos. Chem. Phys.*, 14(9), 4503–4514, doi:10.5194/acp-14-4503-2014, 2014.

Seinfeld, J. H. and Pandis, S. N.: *Atmospheric Chemistry and Physics - From air pollution to climate change*, 2nd ed., edited by John Wiley, John Wiley & Sons, Inc., Hoboken, New Jersey., 2006.

Shen, H., Chen, Z., Li, H., Qian, X., Qin, X. and Shi, W.: Gas-Particle Partitioning of Carbonyl Compounds in the Ambient Atmosphere, *Environ. Sci. Technol.*, 52(19), 10997–11006, doi:10.1021/acs.est.8b01882, 2018.

Shen, X., Lee, T., Guo, J., Wang, X., Li, P., Xu, P., Wang, Y., Ren, Y., Wang, W., Wang, T., Li, Y., Carn, S. A. and Collett Jr, J. L.: Aqueous phase sulfate production in clouds in eastern China, *Atmos. Environ.*, 62(0), 502–511, doi:http://dx.doi.org/10.1016/j.atmosenv.2012.07.079, 2012.

Waxman, E. M., Dzepina, K., Ervens, B., Lee-Taylor, J., Aumont, B., Jimenez, J. L., Madronich, S. and Volkamer, R.: Secondary organic aerosol formation from semi- and intermediate-volatility organic compounds and glyoxal: Relevance of O/C as a tracer for aqueous multiphase chemistry, *Geophys. Res. Lett.*, 40, 1–5, doi:10.1002/grl.50203, 2013.