

***Interactive comment on* “External and Internal CCN Mixtures: Controlled Laboratory Studies of Varying Mixing States” by Diep Vu et al.**

Anonymous Referee #2

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The manuscript by Vu et al. discusses about the experimental method and data analysis for CCN activity of externally mixed particles. Although the technical approach sounds reasonable, the reviewer is skeptical about the novelty. I suggest the authors to significantly improve the manuscript by appropriately referring other papers so that the readers will be able to understand the significance of the manuscript better.

Major comments:

L68 ‘However, dynamic changes in particle mixing states and subsequent treatment of CCN measurement and analysis have not been readily observed and studied in depth.’

I disagree with the statement. A number of papers has been published about the topic in the last decade (e.g., Kuwata and Kondo 2008; Su et al. 2010). I suggest the

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authors to conduct intensive literature search carefully again.

L78 'BC is renown for its direct radiative effects yet little is known experimentally about the contributions of BC to aerosol-cloud interactions at varied mixing states.'; L283' the contributions of BC to aerosol-cloud interactions at varied mixing states is not well known or understood'

Even though there might be only a limited number of studies, some researchers have conducted laboratory experiments/atmospheric observations on CCN activity of BC particles. I suggest the authors to conduct literature search carefully again. For example, there are some description about it in a review paper of BC particles (Bond et al. 2013).

Minor comments:

Title 'CCN Mixtures'

It is not clear to me how this word is defined. Please clarify it, or consider to use other expressions.

L60 'the CCN mobility diameter data sets'

What does this word mean? Please clarify it.

L107 'Five aqueous solutions of succinic acid and NaCl with molar ratios of 100:0, 87:13, 69:31, 43:57, 0:100 were aerosolized using a single atomizer'

I wondered if potential evaporation of succinic acid in the experimental setup could influence the final molar ratios of the compounds. Do the authors have any comments on it?

L112 'As conditions (e.g., but not limited to, residence time, temperature, pressure, relative humidity) change in a flow tube,'

I wondered how well these parameters were controlled during this study. Please clarify.

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L170 'For this study the first curve is similarly defined as the hygroscopic externally mixed fraction (EMF) with an asymptote, or plateau of η . The dependence of η varies with the presence of mixed components and their respective hygroscopicities. Thus we evaluate η for controlled compositions and compare how representative they are of the known fractions of mixtures.'

It may be a good idea to add a figure to explain about the concept.

L241 'After one hour in the flow tube'

Does it mean that particles stayed in the flow tube for one hour? Please clarify.

L257 'The mass size distribution was then converted to number size distribution'

How does the error/uncertainty for measuring mass-size distribution influence the estimated number size-distribution? It would be ideal to conduct sensitivity study about it.

L282 'Combustion Aerosol, hereon also referred to as Black Carbon (BC)'

Combustion aerosol is not equivalent as black carbon.

L283 'BC is considered insoluble but wettable'

Please show experimental data/cite literature to support the idea.

L288 'The single sigmoid fit suggests that the aerosol generated is a homogenous mixture of black and brown carbon.'

The activation curve is very broad. I do not think that such a broad activation curve could be observed if all the particles have exactly the same chemical composition. I wonder what the authors wanted to mean by 'homogeneous.' How can the authors make sure that brown carbon existed in the particles they measured? Please demonstrate the evidence for it.

L292 'It is noted that the apparent hygroscopicity does not account for non-spherical

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fractal particles.’

I do not understand what this sentence means. Do the authors want to clarify that these particles do not have spherical shape?

References: Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Karcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S. (2013). Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res.* 118:5380-5552.

Kuwata, M. and Kondo, Y. (2008). Dependence of size-resolved CCN spectra on the mixing state of nonvolatile cores observed in Tokyo. *J. Geophys. Res.* 113:D19202.

Su, H., Rose, D., Cheng, Y. F., Gunthe, S. S., Massling, A., Stock, M., Wiedensohler, A., Andreae, M. O., and Poschl, U. (2010). Hygroscopicity distribution concept for measurement data analysis and modeling of aerosol particle mixing state with regard to hygroscopic growth and CCN activation. *Atmos. Chem. Phys.* 10:7489-7503.

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