**Interactive comment on "External and Internal CCN Mixtures: Controlled Laboratory Studies of Varying Mixing States" by Diep Vu et al.**
**Anonymous Referee #2**

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 authors to conduct intensive literature search carefully again.

→ We have added the suggested references.

→ We have modified the sentence to read "However, dynamic changes in particle mixing states and subsequent treatment of CCN measurement and analysis have not been readily observed and studied in depth **under controlled laboratory conditions".**

→ We agree with the reviewer that there have been many studies (particularly field studies) that have explored changes in aerosol mixing state and CCN. Kuwata and Kondo 2008 measured ambient air in Tokyo and infer the mixing state properties of the ambient aerosol with subsequent behaviour on CCN. Su et al. 2010 brilliantly develop theoretical models to describe the CCN activation of mixed population and then validate the models with aerosol measurements taken in Beijing, China. A thorough literature review will show that the majority of these papers (like the ones suggested) apply mixing state assumptions to ambient data sets. To our knowledge, we are the only manuscript to show changes in laboratory CCN with controlled *dynamic* changes in mixing state aerosol population (*i)*from external to internal mixtures and *ii)* not to be confused with aerosol-phase states or changes in aerosol coatings). This is the novelty of the presented technique. Previous papers (dating back to the early 70's) have provided appropriate mixing, size-resolved composition theory and have shown the activation of laboratory externally mixed aerosol and internally mixed systems, separately. We have yet to find a CCN paper that has controlled and measured the dynamic change from external to internal mixtures in the laboratory. We apply the technique to CCN but the method can also be extended to understand optical, sub-saturated, and other physicochemical characteristics. We regret that this uniqueness did not come through in the first revision of the paper and thus we have spent considerable effort emphasizing the importance of the new technique presented in the revised manuscript.

→ The following text has been revised to include additional references as follows.
Knowledge of the mixing state and the chemical composition can greatly improve CCN predictions and has been the focus of several studies (e.g., but not limited to (Bilde and Svenningsson 2004; Abbatt et al. 2005; Henning et al. 2005; Svenningsson et al. 2006; King et al. 2007; Cubison et al. 2008; Kuwata and Kondo 2008; Zaveri et al. 2010; Su et al. 2010; Wang et al. 2010; Spracklen et al. 2011; Ervens et al. 2010; Asa-Awuku et al. 2011; Liu et al. 2013; Jurányi et al. 2013; Paramonov et al. 2013; Padró et al. 2012; Moore et al. 2012; Meng et al. 2014; Bhattu and Tripathi 2015; Almeida et al. 2014; Schill et al. 2015; Crosbie et al. 2015; Che et al. 2016; Ching et al. 2016; Mallet et al. 2017; Sánchez Gácita et al. 2017; Cai et al. 2018; Schmale et al. 2018; Mahish et al. 2018; Kim et al. 2018; Chen et al. 2019; Stevens and Dastoor 2019)

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).

→ We have updated the references and provided the following text.
→Previous work investigating the contribution of BC to aerosol-cloud interactions at various mixing states has been studied (e.g., but not limited to Bond et al., 2013 and references therein, (Lammel and Novakov 1995; Novakov and Corrigan 1996; Weingartner et al. 1997; Dusek et al. 2006; Kuwata and Kondo 2008; Koehler et al. 2009; Bond et al. 2013; McMeeking et al. 2011; Liu et al. 2013; Rojas et al. 2015)

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.
→ The title has been changed to Cloud Condensation Nuclei (CCN).
→ ‘CCN mixtures’ is simply a mixture of CCN. It refers to some of the earliest work in the field by Winkler 1973 that accounts for multiple chemical compositions in aerosol. Specifically, this definition of aerosol mixture refers to a property of the overall particle population not to the property of an individual particle (Riemer et al., 2019; Winkler, 1973). Recent studies have modified this definition to include changes in aerosol physical phase-state and also changes in individual particles. We have provided more detail in the revised manuscript and have also added the recent and comprehensive review of Riemer et al 2019 to clarify in the text.

**References added**
→ Winkler, Peter. "The growth of atmospheric aerosol particles as a function of the relative humidity—II. An improved concept of mixed nuclei." Journal of Aerosol Science 4.5 (1973): 373-387.
→ Riemer, N., Ault, A. P., West, M., Craig, R. L., & Curtis, J. H. ( 2019). Aerosol Mixing State: Measurements, Modeling, and Impacts. Reviews of Geophysics, 57. **https://doi.org/10.1029/2018RG000615**

L60 ‘the CCN mobility diameter data sets’ What does this word mean? Please clarify it.
→ The text has been revised to:
To help minimize the complexity in characterizing aerosol hygroscopic and CCN activation properties, CCN data analysis has traditionally been simplified by assuming that *i)* the aerosols share a similar or uniform hygroscopicity over a particle size distribution, *ii)* the CCN particle size can be described by the electrical mobility diameter, *iii)* CCN consists of few multiply charged aerosols and *iv)* all CCN active aerosols readily dissolve at activation.

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?

→ We do not have a comment. Our data shows that the calculated molar solutions agree with predicted values to within 10% of their kohler prediction. Any potential evaporation that could occur is not reflected in the CCN values obtained from the internal mixture studies.

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.

→ The residence time, temperature, pressure, and relative humidity were conducted at ambient room conditions. Typical conditions were dry relative to any experiments conducted with aerosol water. Pressure and temperature were expected to be consistent from test to test (air conditioned laboratory with little temperature fluctuations, and minimal changes in barometric pressure). The above has been inserted and clarified in the text.

L170 'For this study the first curve is similarly defined as the hygroscopic externally mixed fraction (EMF) with an asymptote, or plateau of _. The dependence of _ varies with the presence of mixed components and their respective hygroscopicities. Thus we evaluate _ 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.

→ Please refer to figure 2. In the previous revision, we include this concept figure to explain the difference in plateaus of externally and internally mixed data.

L241 'After one hour in the flow tube' Does it mean that particles stayed in the flow tube for one hour? Please clarify.

→ this is in reference to 1 hr after initial injection. Text has been modified from 'After one hour in the flow tube' to 'One hour after initial injection into the flow tube' to clarify this.

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.

→ Unfortunately, this was beyond the scope of this study; independent aerosol mass measurement was not available. However, with known chemical composition the mass distribution was converted to a number distribution using literature density values to yield a number value to conduct a CCN analysis. Although there is some error attributed to this method, it is consistently done from test to test.

L282 ' Combustion Aerosol, hereon also referred to as Black Carbon (BC)' Combustion aerosol is not equivalent as black carbon.

→ Yes, combustion aerosol is not all Black Carbon (BC). We have clarified that BC is this paper is short for black carbon mixtures or BC containing particles and have revised the text in multiple places as follows:
→ Combustion aerosol or soot can form external and internal complex aerosol mixtures. Soot is considered insoluble but wettable (Lammel and Novakov 1995; Moore et al. 2014) and the contributions of Black Carbon containing particles to aerosol-cloud interactions at varied mixing states is not well known or understood (Lammel and Novakov 1995; Novakov and Corrigan 1996; Weingartner et al. 1997; Dusek et al. 2006; Kuwata and Kondo 2008; Koehler et al. 2009; Bond et al. 2013; McMeeking et al. 2011; Liu et al. 2013; Rojas et al. 2015). Thus, the ability of black carbon to mix with inorganic and organic compounds and to observe the extent of mixing as they activate as CCN is of great interest. Prior to investigating the impact of mixing fresh combustion emissions with inorganic and organic aerosols, the CCN activation spectra of soot was measured using combustion aerosol generated from the APG. The aerosol is likely composed of black carbon and oxidized carbonaceous material (Moore et al. 2014; Durdina et al. 2016). Thus we also refer to carbonaceous aerosol as black carbon mixtures (simply, BC mixtures).

L283 'BC is considered insoluble but wettable' Please show experimental data/cite literature to support the idea.

→ Previous work (e.g. but not limited to, Lammel and Novakov 1995; Moore et al. 2014) has indicated BC as hydrophobic, but still wettable.
→ It is understood that CCN can be soluble or insoluble but wettable particles - a water film can cover the surface of the aerosol and the activation of the particle is more dependent of the curvature and less of the composition. This is further supported by this study which showed a low activation diameter (fig. 7) for combustion aerosol and thus a low kappa value of approximately 0.001, which is close to 0 - particles that are insoluble but wettable have been characterized with a kapps value of close to 0.

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.

→ A homogeneous mixture similar chemical compositional fractions across different particle sizes. A homogenous mixture is an internal mixture. The data presented for known compounds illustrated that homogenous (internal) mixtures exhibit singular curves. That is the chemical composition across CCN active sizes is constant and thus the activation is smooth (with no plateau). Externally mixed CCN exhibit plateaus and multiple points of activation. The CCN activation of the soot is a singular curve and thus indicates that the aerosol are internally (homogeneously) mixed.
→ We have removed the word brown carbon and replaced the wording to reflect the more oxidized soot components found in aerosol generated from flame burners.
→ The text is now rewritten as :There are no plateaus in the activation curve and the single sigmoid fit indicates that the aerosol generated is a homogenous internal mixture.

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

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

→ The apparent hygroscopicity is calculated from the electrical mobility diameter and thus assumes all particles as spherical. Electrical mobility diameter measurement does not take into account non-spherical fractal particles.
→ The text has been changed as follows: It is noted that the apparent hygroscopicity is defined by the electrical mobility diameter that assumes particles are spherical.

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