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Interactive comment on “Quantifying aerosol mixing state with entropy and diversity measures” by N. Riemer and M. West

Anonymous Referee #1

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Riemer and West both introduce and demonstrate a viable mathematical definition of aerosol mixing state derived from consideration of species distribution among particles. For this purpose the authors bring the useful concepts of “diversity” and “mixing state index” to the vocabulary of aerosol science. The paper is especially important because it quantifies mixing state beyond the textbook, extreme-case-limited definitions: external and internal mixing. The fact that the new quantities can actually be tracked in models and/or inferred from measurements is of course essential and the authors have made this case. For these reasons alone the paper is suitable for publication ACP

The authors weave their treatment with referenced historical perspectives on the mathematical application of diversity to other fields, specifically ecology, economics, and genetics. Finally the authors present an interesting extended definition of their metrics

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to the recently popular q -type generalizations of entropy (e.g. Tsallis entropy). These generalizations, non-intuitive to this reviewer, do reduce continuously to the conventional Shannon entropy for $q = 1$ and have analytical properties that could well be of value to definition of aerosol mixing state – but the jury is still out on this last point. As reviewer I find no weaknesses in the author's mathematical analysis, establishing the properties of their new metrics, and the presentation style to be excellent. Nonetheless, there are serious problems with the physics – the authors have achieved a mathematical definition of mixing state not a physical one. If discussions of physical entropy and the second law are omitted, the paper is fine.

Physics: My chief reservation with the paper derives from confusion of the mathematical definition of entropy, nicely developed here, and the thermodynamic one. The mathematical quantity defined in the paper is not extensive (i.e. it does not scale linearly with particle number or mass) and so cannot be the thermodynamic entropy. One of the pitfalls that can arise when this distinction is not followed is unfortunately illustrated at the end of Section 2 where it's stated: "The case of a coagulating population represents a closed system as the total mass is conserved. Hence according to the second law of thermodynamics, the total entropy of the system has to increase". There are two mistakes here: When particles irreversibly stick together during coagulation, some kinetic energy is converted to heat and dissipated to the environment, so the system is not closed. One might add that it is even intuitively plausible for a coagulation process in which several particles aggregate to one entropy is reduced. Precisely, if during its formation the aggregate releases heat Q to the environment, the entropy is reduced by Q/T , even though total entropy (system + environment) increases. On the other hand, the authors establish that their expression for the entropy, which pays no heed to particle number, increases for this process. The contradiction arises from trying to equate the mathematical and thermodynamic definitions of entropy.

In summary, this paper is a provocative start that should motivate others to take up the quest of defining general states of mixing to include not only species distribution among

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particles, as the authors have successfully done here, but also to include particle number, size, shape, and composition coordinates. With the all subtleties and pitfalls in this business of working with entropy this won't be an easy task.

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