

Interactive comment on “Modelling the formation of organic particles in the atmosphere” by T. Anttila et al.

Anonymous Referee #2

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The paper makes appropriate use of an aerosol dynamics model to investigate the evolution of freshly nucleated stable clusters in a varying source field of gaseous sulphuric acid and organic vapour. The model makes use of theory describing the activation of the clusters in a supersaturated vapour. It is based on well-established Kohler theory; the supersaturation in this case being of an undefined organic oxidation product (scaled to OH concentration) determined by its chemical source function rather than water vapour determined by the air parcel vertical displacement in traditional applications of the theory. As found in the original paper describing the application of the theory, the clusters initially grow by condensation in the available acid and by coagulation until they are of sufficient size to activate (around 2 nm). Thereafter, the evolution of the gas and aerosol is followed by the model. Sensitivity of the model to input parameters, to varying RH and to temperature was explored and detailed, then the discussion

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applied to address seasonal fluctuations in aerosol formation at a continental site. The paper applies the model well, and describes the processes affecting the formation of observable atmospheric aerosols. The work is built on the foundation of many years of observation, process description and careful model application by the authors and is a welcome contribution to atmospheric aerosol research. It provides a framework for detailed investigation into the specific precursor formation and application of more refined thermodynamic theory; but one which stands in its own right as a useful descriptive tool.

As mentioned above, the paper is worthy of publication, but certain points warrant consideration: i) It would be useful to have more numerical detail of the aerosol dynamics model in sections 3.2 and 3.3, particularly concerning the method of treatment of coagulation (was it treated explicitly, and if so, how was it split such that self-coagulation could be neglected). Also, were simulations allowing the development of the pre-existing size distribution carried out and how did this compare with the cases where CS was held constant? ii) The application should be contrasted to the behaviour of cloud droplet activation in the conventional application of Kohler theory: the rapidly-produced organic supersaturation of 1000s to 10000s of % with lifetimes of 20 minutes or so, contrasting with the modest sub 0.1% water supersaturation being easily and rapidly depleted by the activating droplet population is not stressed. iii) The gas phase chemistry scales the organic vapour to the OH production. Whilst primary OH production scales to solar flux, many studies have shown that oxidising capacity (and OH concentration) is very dependent on radical recycling and that the OH time evolution is skewed with respect to the solar flux. This will affect both sulphuric acid and organic vapour concentration so is not so important. What is more important is that the most condensable and water soluble organics are likely to be the most oxidised. These are not the primary oxidation products and will more likely be secondary or tertiary products (see e.g. publications on MCM by Saunders, Jenkin, Pilling etc...). Sensitivity of the aerosol evolution to organic vapour maxima formed significantly later than the sulphuric acid maximum should be explored. It is accepted that other oxidants may

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contribute to the oxidation process, but the secondary and tertiary products can only be produced later than the primary ones.

The most important point which should be addressed is that the Kohler theory adequately covers a range of applications. To use a new nomenclature merely to describe the application of the theory to equilibration in a non-water vapour at a higher than usual supersaturation and hence activation of smaller particles is not appropriate. The references to "nano-Kohler", and to a "new" or "newly-developed" theory (used throughout the manuscript), should be corrected to reflect that this is a new application of well-established theory. The work is good enough to stand in its own right considering the dynamic evolution of aerosols including nucleation, coagulation, activation and condensation, without using a new term to describe the activation.

One final minor point which should be addressed is that on line 21 of p 6163 it is stated that "...CS>10³ s⁻¹...". If this were true, the steady state organic vapour concentration would be 100 molecules / cc, far below supersaturation. If the value is of the order of 10⁻³ as stated on line 15 of p 6154, it is not so much greater than the last term of equation 5 as stated.

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