

## ***Interactive comment on “New particle formation and growth at a remote, sub-tropical coastal location” by R. L. Modini et al.***

**Anonymous Referee #2**

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The paper discusses formation and growth of nucleation mode particles inferred from measurements made at a coastal location south of the Great Barrier Reef. VHTDMA data were used to infer composition of the nucleation mode particles, and it was concluded that sulphate or organic vapours were responsible for growth to the observed sizes.

The measurements are useful and the methodology and the some of the text reasonably balanced. In the analysis, I found it particularly useful to differentiate between the coastal and non-coastal classification schemes of Buenrostro Mazon et al. (2009) and the coastal scheme Vana et al. (2008) for the classification of the events.

The measurements are very well made and quite complementary, but I have a major concern about their interpretation. For this reason, I recommend quite major modifica-

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tion of the manuscript.

The concern is as follows: from the AIS and SMPS data it is obvious that any apparent nucleation leading to the observed "nucleation" mode was non-local (nothing really seen sub-10 nm). It is stated that the particle number decreased before every nucleation event as a result of the daily shift in wind direction from continental to marine / coastal. It appears very likely that the condensable vapours responsible for the nucleation / growth had a continental origin and were simply photochemically processed to produce nucleation as the air mass returned to the coastal measurement site. Were there ever nucleation events that were not contaminated by this sea-breeze effect? Such effects have been well-known for decades for gaseous secondary pollutants such as ozone, when the precursors are advected out to sea by land breezes and ozone returned onshore after photochemical processing. It would be important to eliminate such effects by a null event where there was no land-sea breeze effect - the only other way to eliminate it is by an anthropogenic tracer such as CO or black carbon measurements which should both be at background MBL concentrations if the land-sea breeze is not the source of the particles. It is essential that the authors screen their data for such effects before drawing conclusions about the nucleation / growth precursor sources. In the absence of being able to screen for such an eventuality, this should be clearly stated as the likely source of the nucleation precursors - moreso than any remote marine sources. This interpretation is obvious from figures 6 and 7. It is, of course, possible that the clusters were advected and grew offshore rather than being nucleated from vapours offshore - however, these two options must be eliminated before anything can be said about coastal or marine sources. It was particularly interesting to note the difference in growth rates from the Barrier reef event and all other events, though of course this may simply be the result of different continental precursor strengths being advected offshore in the different wind direction. In any case, one would not expect to see a tidal signature in particles of a continental origin - only a land-sea breeze signature. Likewise, one would not see such local meteorological effects in trajectory data as these are based on mean geostrophic winds and will not capture

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such effects.

Because of these reasons, the source study and conclusions should be rewritten after further analysis. Given any amendments to the conclusions if the continental influence cannot be removed, reference to the tidal and open ocean productivity figure should be minimised - probably removing the figures entirely.

I have one further criticism of note: I feel it must be better distinguished between the growth to detectable and growth to observed sizes. As the authors state, it may correctly be inferred that sulphate and organic material was responsible for the mass determining the volatility and hygroscopic growth of 17 to 22.5 nm particles. However, since the particles are detectable at 3 nm, the mass of newly detectable particles would be (at constant density) a maximum of  $3^3 / 17^3 = 0.005$  times (or 0.5% of) the mass of the particles whose properties were being measured. It is therefore regrettably impossible to deduce the properties of the material contributing to the growth to detectable sizes of the particles. The authors must therefore remove the reference to INITIAL growth being driven by sulphate and organic vapours in the abstract and throughout the manuscript - it is merely the growth to sizes above 10 nm. There has been care to make this clear in many places of the manuscript, but this point cannot be too strongly made.

Minor comments: Intro It's stated that marine aerosols constitute a significant fraction of the aerosol mass. This seems an odd way to justify studies into marine aerosol (I'd probably normally expect a statement of the high contribution to radiative forcing); if the authors choose to phrase it this way, the fraction should be stated and referenced.

It is also stated that a relatively stable number implies MBL replenishment. Continuous entrainment from the FT and reasonable lifetimes with respect to deposition will also result in stable numbers. Entrainment will have continuous and sporadic components - variation in number can result from gentle fluctuations in the continuous entrainment rate and vigorous changes in sporadic entrainment and boundary layer ventilation un-

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der less stable conditions. Why is this discounted? In any case, entrainment should be mentioned.

The arguments surrounding the composition of the first step in MBL aerosol production seems very skewed towards sulphuric acid. Firstly, there is no unambiguous evidence for the participation of sulphuric acid playing a role in MBL nucleation. Admittedly there is no unambiguous direct evidence for any component of nucleation mode aerosol - but where there is evidence, it points to iodine, not sulphuric acid, ammonia or organics. Admittedly this is at the coast and is tidal, but non-coastal MBL nucleation (and not entrained from the FT) has not been unambiguously observed. This is mentioned, but less prominently than is necessary.

Reference to Whitehead et al. (2009) should be made when discussing the other studies of nucleation at a coastal site on page 5. (Whitehead, J. D., G. B. McFiggans, M. W. Gallagher and M.J. Flynn, Direct linkage between tidally driven coastal ozone deposition fluxes, particle emission fluxes and subsequent CCN formation, *Geophys. Res. Lett.*, 36, L04806, doi:10.1029/2008GL035969, 2009)

Referencing In-line citations should consistently be in correct ACP format e.g. O'Dowd and DeLeeuw (2007), not O'Dowd and DeLeeuw (O'Dowd and DeLeeuw, 2007)

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 9, 12101, 2009.

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