Review of:
Wonaschütz, A., et al., Hygroscopic properties of organic aerosol particles emitted in the marine atmosphere, *Atmospheric Chemistry and Physics Discussions*, 13, 11919-11969 (2013).

This paper presents results from a truly novel "in situ experiment," where a plume of primary organic aerosol (mostly hydrocarbon like) was generated in the marine environment. Subsequent changes in the physical and chemical properties of the aerosol were quantified, with a focus on the aerosol hygroscopic properties and CCN. Different behaviors on consecutive days were observed and were likely due to dramatically different meteorology on the different days: one was sunny with lower RH while the other was foggy with much reduced solar radiation. The results are novel and will be of interest to many in the atmospheric science community. I recommend that the paper be accepted for publication after the following issues are addressed.

Overall Comments:

1. The writing overall is good, but there is a grammatical issue that needs to be addressed: the verb tense oscillates between present and past tense throughout the entire paper. The tense changes several times within paragraphs (and even at times within the same sentence), which is very distracting. Choose a tense and be consistent throughout.
2. I think that the central points of the paper would come through more clearly if the paper were shortened. As an example, Sections 3.1 and 3.5 could be shortened substantially without taking away from the overall findings of the paper. Other sections would also benefit from copy editing to reduce wordiness and extraneous discussion (one example: pg. 11932, lines 4-7).
3. A focus of the paper is the comparison of GFs for different particle sizes. In some cases, there are very small changes in GFs (for example: abstract, lines 17-20). A discussion of the uncertainty of the GF measurements is surely needed. Are GFs of 1.04 and 1.06 statistically different? (this comment pertains to Section 3.4 as well)
4. The lack of a sea salt measurement (as Na^+ and Cl^-) seems to hinder the analysis. Interpretations of changes in CCN/CN are largely based on AMS measurements, which (1) are limited to < 1 µm, and (2) cannot measure sea salt (even in the fine mode). How would changing sea salt concentrations impact any of the stated conclusions, and can the size distribution data (which extend up to 20 µm) be used to constrain the contribution of sea salt to total CCN?

Specific Comments:

1. The title is misleading – it implies that the organic particles were emitted naturally.
2. Since analysis of the plume is the focus of the paper, more detail is needed on the plume generation beyond simply citing the Russell et al. (2013) study: the paraffin-type oil chemical characteristics, aerosol/plume generation method…etc.
3. For clarity, consider labeling the July 18 plume encounters as B1 and B2.
4. Pg. 11931, line 13: that the background aerosol was dominated by sulfate and organics seems to be a function of the analysis methods. One would expect sea salt to be a prominent component of the background aerosol during this study. The AMS cannot measure sea salt (which is why the chloride concentration in Fig. 3 seems to have no

correlation with wind speed) – this is a limitation that should be explicitly stated. (see above comment as well)

5. Pg. 11935, lines 20-26: It is not clear why CCN concentrations would increase at the moderate *S* **before** they would increase at the higher *S*? This does not seem physically possible, and the stated explanation for this behavior is very confusing.
6. Sections 3.4, 3.5: when contrasting the CCN results with the GF results, care must be taken to put the respective measurements in perspective. The CCN measurements are not size specific in the way that the GF measurements are. Since the GF measurements are limited to < 300nm, these measurements are fundamentally characterizing different regimes in the aerosol distribution – this should be mentioned (e.g., Section 3.4, first sentence mentions this comparison).
7. From the FTIR spectra (Fig. 8), it appears that in plume A3, the strong alkane peaks are gone, or are at least dominated by the hydroxyl peaks – why then is the WSOM:Org ratio still relatively low during A3 (hard to tell from the scale on Fig 7, but it appears to be ~7-20%)?
8. From Figure 7, it appears as though the grey shaded region (labeled 'A3') represents time in the plume and significant 'dilute' (background) sampling as well? Was the entire shaded region depicted in Fig. 7 used for the A3 analysis? If so, how is this justified?
9. Figure 6: x-axis scale (i.e., horizontal scale of one hour) should be consistent for top and bottom panels.
10. Pg. 11937, line 9: is there a statistically significant difference between GFs of 1.05 and 1.09? (see above comment)
11. Pg. 11937, line 14: say "…those observed within the plume")…
12. Pg. 11939, line 13-14: what were the WSOM and AMS org. concentrations at points A1 and A2? These should be given for comparison.
13. Pg. 11942, line 17-18 "inorganic mass fractions were very low at all particle sizes" – Clarify that this only applies to sub-1 μm particles.
14. Pg. 11927, line 3: what fraction of scans were accepted/discarded? This should be stated.
15. Pg. 11927, line 27: is there an error in one of the diameters given: 266 nm (midpoint: 946 nm)?
16. Pg. 11936, line 25: delete 'ship'
17. Pg. 11944, line 16: change 'cloud' to either 'fog' or 'aqueous'
18. Pg. 11944, line 23: see comment above on the title – saying organic particles "emitted" in the marine atmosphere is misleading and suggests they are naturally occurring.