Review of Wan et al., 2020:

This paper contains reports on the multitude of organic compounds present in size segregated aerosol samples at a coastal location where frequent new particle formation (NPF) was observed, as measured by a powerful off-line mass spectrometric technique. Iodine-NPF events are identified (I-NPF) and these are linked to aerosol chemical composition during periods with three days of consecutive I-NPF days. These are compared with similar data for C-NPF days. The potential sources are discussed, and volatilities of these compounds as calculated by a group contribution method are presented and discussed with a view to linking these compounds to new particle growth. The paper is well within the remit of ACP and contains a very valuable dataset in a field where not much is yet known, however, I find issues with the authors' definition of I-NPF, and the discussion of calculated vapour pressures could do with some extension as there are many uncertainties with these calculations not discussed. I recommend publication after the following issues are addressed.

General comments:

- In **section 3.1** the authors present 10-56 nm particle phase composition in 72 hour aerosol samples through 3 consecutive NPF days of two different categories (iodine-induced NPF, or *I-NPF*, & continental regional NPF, or *C-NPF*), identified from size-distribution measurements. It is evident from the size distributions that these NPF days *are* very dissimilar, and the evolution of the size distribution is markedly similar to prior reports of iodine-nucleation (such as Sipilä et al., 2016), however, the authors state that an elevation to the mass concentration of iodine in 10-56 nm aerosol samples during the I-NPF days is clear indication that NPF was linked to iodine nucleation. I would strongly argue this is not a clear indication a nucleation process involving iodine vapours, as aerosol mass between 10-56 nm is not an indicator of what process produces particles at ~1.7 nm.

  I would urge the authors to back this claim up with reference to similar reports of iodine driven nucleation under conditions that produce either similar particle composition or similar size distributions. HSO_4^- concentrations are still an order of magnitude higher than I^- concentrations in 10-18 nm particles on I-NPF days. If there is there any evidence within the data that nucleation processes are not dominated, for example, by sulphuric acid processes, this would strengthen this section greatly.

- It would be very useful to mention the magnitude of the effect of these NPF events to aerosol mass in the relevant size ranges – coming up with an exact number is a rather uncertain process, but a simple alternative would be a plot showing the time evolution of size-segregated aerosol mass as currently there is no indication of how much of the aerosol mass is actually arising from NPF.

- Recent chamber results utilising EESI-ToF-MS would indicate that oligermisation processes in the particle phase can produce a diverse range of compounds (Pospisilova et al., 2020), as well as particle phase processes producing organosulphates (Mutzel et al., 2015), formation of ester hydroperoxides (Zhao et al., 2018) etc. I would consider how such mechanisms may affect your proposed formation mechanisms, and further your estimations of volatility - if some of these compounds arise from particle-phase oligomerisation, does this change the conclusions of the paper? I would like to echo reviewer #1 here and suggest an extension of the discussion so the results can be more easily extrapolated to other coastal regions.

- The estimation of volatilities through SIMPOL has some associated uncertainties, and, at least in the case of HOMs, estimations of volatility can vary by several orders of magnitude when compared with other methods (see Figure 8 of Peräkylä et al., 2019). This should be a point of discussion as it may affect the outcomes of the paper.

Specific comments:

Line 22 and throughout: Change "organics" to "organic compounds".

Line 43: "Highly oxygenated (multifunctional) organic molecules" is a more correct term for these than "highly-oxidised" (see Bianchi et al., 2019)

Line 44: If you're using Extremely Low Volatility here to refer to molecules that fall into the volatility class C*(300K) $3\cdot10^{-9} < C^*(300K) < 3\cdot10^{-5}$ μg m^{-3}, then it is worth note that by current classification of HOMs as discussed in Bianchi et al., 2019, HOMs fall across many of these volatility classes. As you refer to nucleation on line 47, I would consider revising this statement as it is currently thought that only ULVOC (as defined by Schervish & Donahue, 2019; Simon et al., 2020) are capable of undergoing pure biogenic nucleation. I would include this also in your discussion of section 3.3.3, as many of your compounds fall into this volatility class and this could make a valuable addition to this discussion.

Line 46-47: Given the nature of the paper, it may also be worth mentioning recent work discussing formation of HOMs from chlorine oxidation also (Wang et al., 2020).

Line 89: It would be nice if you provided more detail here for those unfamiliar with I-NPF. How does the evolution of the size distribution differ from conventional "banana-plot" NPF? This definition feels very arbitrary. Is there also any air-mass data that can support the assignment of "I-NPF"? It would be very beneficial to have a steadfast criteria that separates the two.

Table 1: Consider using the same units (μg m^{-3}) for all quantities.

Line 208-209: "The formula number of the least common CHN^+ group is only 46". Do you mean CHN^+ is the least common group?

Line 214-216: Is there any evidence for the role of amines in iodine nucleation? A reference to this would be informative here, otherwise it is of little relevance given the section discusses iodine nucleation

Line 301-303: It is not evident to me that these spectra indicate oxidation products of continental terpene emissions, the peaks at 9-10, 13-15 and 18-20 carbon numbers that would typically be expected of these products are not present. This point could do with expanding.

Technical corrections:

Line 44: High O:C ratio**s** (rather than "ratio")

Line 72-73: Change "Studies investigating coastal organic aerosols have been rarely" to "...aerosols are rare"

Line 80: Missing hyphen in FT-ICR-MS

Line 86: Space between 200 and m

Figure S1: I would urge the authors to consider using a colour scale that does not "cap out" at 1,500x64 cm^{-3} as this disingenuously misrepresents the I-NPF event.

Line 149: Should this read "Nanometer *sized* new particles"?

Figure S2 and throughout: Dalton is not a unit of mass/charge.

Line 222: Should this read (free) troposphere?

Line 253: change to "**a** relatively high unsaturation degree"

References

Bianchi, F., Kurtén, T., Riva, M., Mohr, C., Rissanen, M. P., Roldin, P., Berndt, T., Crounse, J. D., Wennberg, P. O., Mentel, T. F., Wildt, J., Junninen, H., Jokinen, T., Kulmala, M., Worsnop, D. R., Thornton, J. A., Donahue, N., Kjaergaard, H. G., & Ehn, M. (2019). Highly Oxygenated Organic Molecules (HOM) from Gas-Phase Autoxidation Involving Peroxy Radicals: A Key Contributor to Atmospheric Aerosol. *Chemical Reviews*.

Mutzel, A., Poulain, L., Berndt, T., Iinuma, Y., Rodigast, M., Böge, O., Richters, S., Spindler, G., Sipila¨, M., Jokinen, T., Kulmala, M., & Herrmann, H. (2015). Highly Oxidized Multifunctional Organic Compounds

Observed in Tropospheric Particles: A Field and Laboratory Study. *Environmental Science and Technology*, *49*(13), 7754–7761. https://doi.org/10.1021/acs.est.5b00885

Peräkylä, O., Riva, M., Heikkinen, L., Quéléver, L., Roldin, P., & Ehn, M. (2019). Experimental investigation into the volatilities of highly oxygenated organic molecules (HOM). *Atmospheric Chemistry and Physics Discussions*, 1–28. https://doi.org/10.5194/acp-2019-620

Pospisilova, V., Lopez-Hilfiker, F. D., Bell, D. M., El Haddad, I., Mohr, C., Huang, W., Heikkinen, L., Xiao, M., Dommen, J., Prevot, A. S. H., Baltensperger, U., & Slowik, J. G. (2020). On the fate of oxygenated organic molecules in atmospheric aerosol particles. *Science Advances*, *6*(11), 1–12. https://doi.org/10.1126/sciadv.aax8922

Schervish, M., & Donahue, N. M. (2019). Peroxy Radical Chemistry and the Volatility Basis Set. *Atmos. Chem. Phys. Discuss*. https://doi.org/10.5194/acp-2019-509

Simon, M., Dada, L., Heinritzi, M., Scholz, W., Stolzenburg, D., Wagner, A. C., Kürten, A., Rörup, B., He, X., Almeida, J., Baccarini, A., Bauer, P. S., Beck, L., Bergen, A., Bianchi, F., Brilke, S., Caudillo, L., Chen, D., Chu, B., … Yan, C. (2020). Molecular understanding of new-particle formation from alpha-pinene between -50 °C and 25 °C. *Atmospheric Chemistry and Physics Discussions*, *January*, 1–42.

Sipilä, M., Sarnela, N., Jokinen, T., Henschel, H., Junninen, H., Kontkanen, J., Richters, S., Kangasluoma, J., Franchin, A., Peräkylä, O., Rissanen, M. P., Ehn, M., Vehkamäki, H., Kurten, T., Berndt, T., Petäjä, T., Worsnop, D., Ceburnis, D., Kerminen, V. M., … O'Dowd, C. (2016). Molecular-scale evidence of aerosol particle formation via sequential addition of HIO3. *Nature*, *537*(7621), 532–534. https://doi.org/10.1038/nature19314

Wang, Y., Riva, M., Xie, H., Heikkinen, L., Schallhart, S., Zha, Q., Yan, C., He, X., Peräkylä, O., & Ehn, M. (2020). Formation of highly oxygenated organic molecules from chlorine atom initiated oxidation of alpha-pinene. *Atmospheric Chemistry and Physics*, *20*, 5145–5155. https://doi.org/10.5194/acp-2019-807

Zhao, R., Kenseth, C. M., Huang, Y., Dalleska, N. F., Kuang, X. M., Chen, J., Paulson, S. E., & Seinfeld, J. H. (2018). Rapid Aqueous-Phase Hydrolysis of Ester Hydroperoxides Arising from Criegee Intermediates and Organic Acids. *Journal of Physical Chemistry A*, *122*(23), 5190–5201. https://doi.org/10.1021/acs.jpca.8b02195