Response: We thank the two reviewers for thoughtful suggestions and constructive criticism that have helped us improve our manuscript. Below we provide responses to reviewer concerns and suggestions in blue font.

Reviewer #1:

1- Without direct measurements of actinic flux, I am not sure how sturdy the authors' conclusion are about the role the OH radical plays in the nucleation in this layer but previous studies seem to support this conclusion.

Response: Just as the reviewer said, we are using the literature references to support this conclusion. As a result, we do not make any changes to the manuscript with regard to this comment.

2- Another point to raise is the actual aerosol type that is found in the vicinity of the EIL be it smoke, dust, or pollution. Can the authors comment on what the dominant type is and how each can affect or influence particle enhancement in the EIL? There was not much emphasis on this since aerosol type is important when quantifying aerosol cloud-climate interactions.

Response: This is an excellent point made by the reviewer. The main aerosol types in the EIL and free troposphere, based on our sampling and analysis of remote sensing lidar data in previous studies, include exactly what the reviewer suggested: smoke, dust, polluted continental emissions. There also can be detrained aerosol from the marine boundary layer that is lofted up by cloud. The presence of these air mass types is now discussed in the manuscript:

"The sources of pollution impacting the study region vary in terms of the vertical layer being examined. More specifically, the predominant sources in the STBL are marine sea spray and biogenic emissions, and ship exhaust (e.g., Coggon et al., 2014; Modini et al., 2015), while the major sources impacting the FT originate from the continent, including biogenic emissions, wildfires, anthropogenic emissions, and crustal emissions (e.g., Crosbie et al., 2016; Wang et al., 2016). As it is challenging with the current dataset to separate the relative importance of the pollution type affecting the EIL, instead the focus of the subsequent discussion is on aerosol size distributions."

It is too difficult to separate the relative importance of each emissions source in affecting the EIL. As a result, we do not add text to address that part of the reviewer comment.

The reviewer provided the following comments/suggestions in their Supplement file in PDF format:

3- Make sure to use proper citation format. Comma and period are reversed with a few of these author citations.

Response: Done.

4- EIL Thickness column is a bit confusing. Maybe should stick with the EIL thickness and comment on the range of base and top altitudes for the entire campaign.

Response: The table has been edited accordingly and the range of base and top altitudes is provided now.

5- Could just same "Same as Figure 5 but CPC" or something like that.

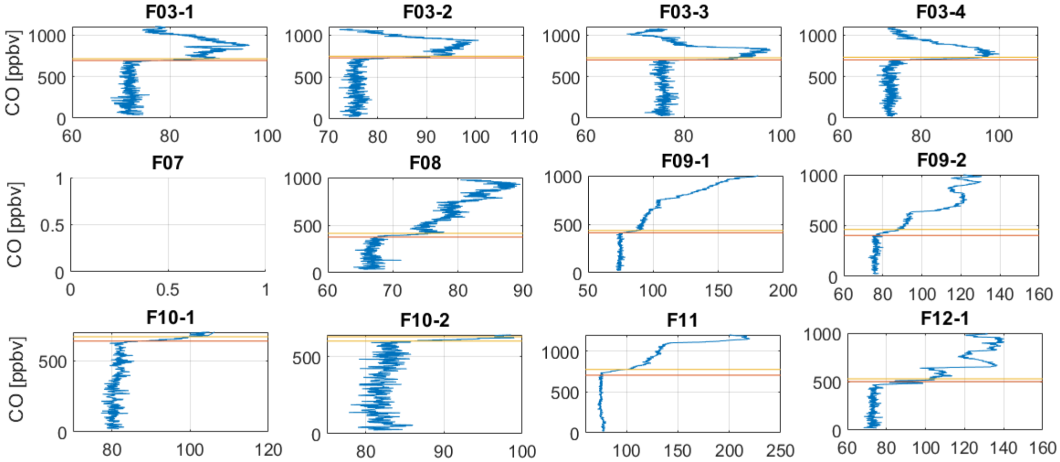
Response: Done.

Reviewer #2:

1- Table 1: PCASP showed higher concentration in EIL than those in STBL and FT in five cases. These are also among the cases when some of the highest concentrations of CN(3-10 nm) were observed. Does the highest PCASP concentration in EIL indicate a different airmass than those in STBL and FT? If so, for these cases, could the enhanced CN(3-10 nm) be a result of the different airmass instead of enhanced new particle formation? Were there any trace gas measurements that could provide information on the air masses and potential mechanism of the new particle formation in EIL?

Response: This is an excellent suggestion by the reviewer. We did conduct CO measurements during FASE, but not in NiCE. We checked CO profiles for these cases (shown below), and they reveal that EIL CO concentrations are between STBL and FT values. Based on these profiles, it is less likely that the EIL has a different airmass than the STBL and the lower FT, specifically FT1 and FT2. We now add the following text to the manuscript:

"Also, as a way to rule out the presence of a different air mass in the EIL that is distinctly different than those in the STBL and FT, vertical profiles of CO (not shown here) were examined for the cases in Table 1. CO exhibited a smooth transition in concentration in the EIL progressing from lower values in the STBL to higher values in the FT. Based on that result and the shallow depth of EIL, it is concluded that the EIL in the cases examined did not have a distinct air mass affecting it that was different from either that in the STBL or the lower FT."




2- Figures 5-6: Given enhanced new particle formation in the EIL is one of the major conclusions, I would suggest plot CN(3-10 nm) as function of altitude in EIL (similar to Figures 5 and 6). This may provide more insight into the mechanism of new particle formation.

Response: We thank the reviewer for this great suggestion. We revised our Figures 5 and 6 by plotting profiles for different size ranges, including 110-3400 nm and 10-110 nm. The overall behavior remained similar. We added a new figure (Figure 7; see below), which is a profile of particle number concentration in the diameter range of 3-10 nm within the EIL. Figure 7 demonstrates that concentrations of particles in this size range exhibit a different, and non-linear, relationship with altitude in the EIL as compared to the other two size ranges. We have added the following text to the paper:

"Figure 7 demonstrates that concentrations of particles in the $D_p$ range between 3 and10 nm exhibit a different, and non-linear, relationship with altitude in EIL as compared with the other two size ranges. This non-linear relationship of particle concentration with altitude is likely due to nucleation of particles within the EIL. "

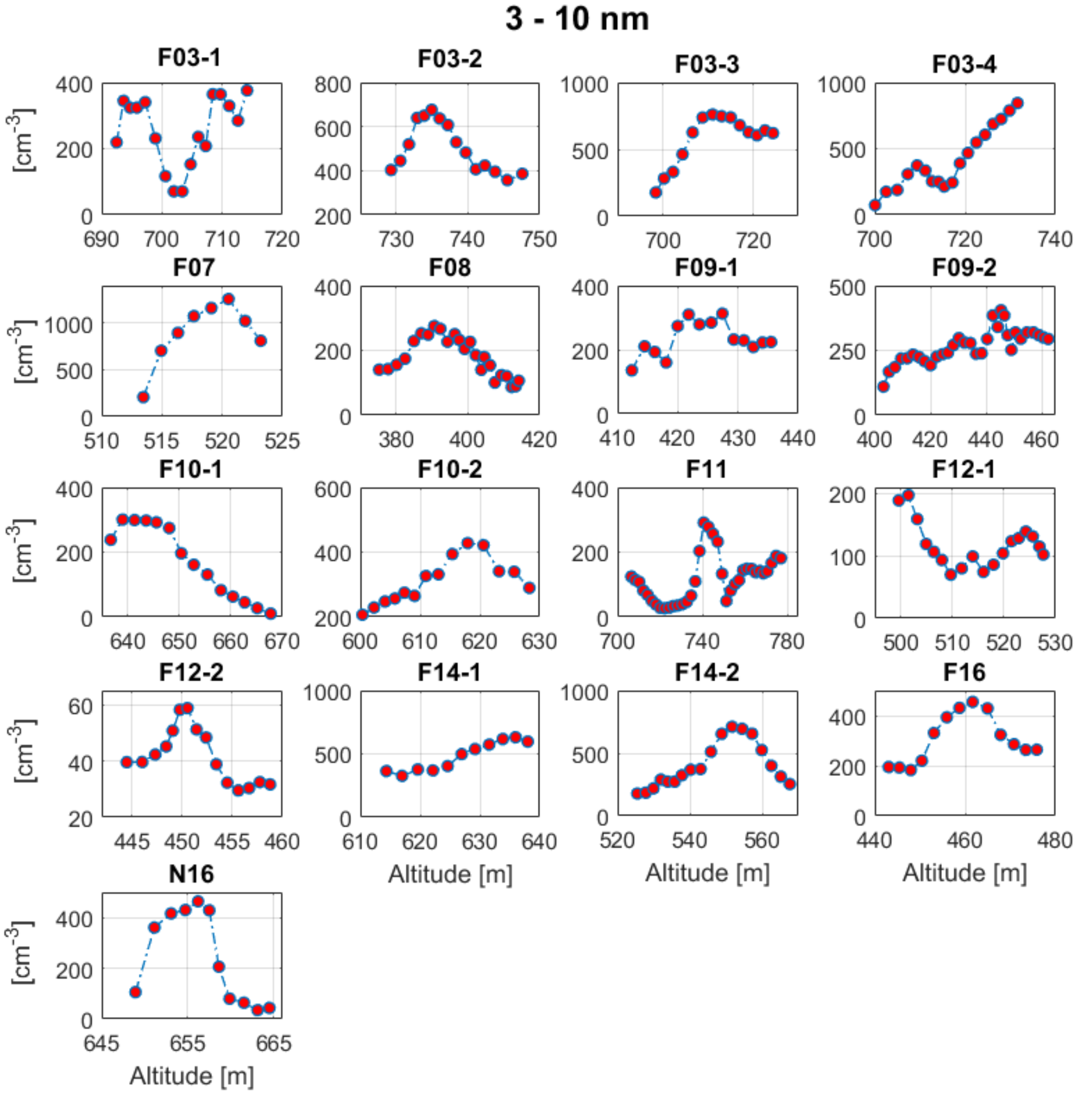



**Figure 7: Particle concentration in diameter range 3-10 nm as a function of altitude in the EIL.**

3- Line 168-169: Based on Figure 3, the highest number concentration for particles with Dp between 10 and 110 nm was in FT1 and FT2 layers, instead of FT2 and FT3.

Response: This change has been made.

4- Line 214-216, The concentration of CN(3-10 nm) is substantially lower than CN(10-110nm) in both EIL and FT. I think the new particle formation is likely slow, and the growth of newly formed particles to CCN and optical active sizes is also very slow. I am quite convinced that these nucleated particles have significant impact on marine CCN budget. Could the author comment on the rates of new particle formation and growth?

Response: Based on the reviewer's great suggestion, we added the following text:

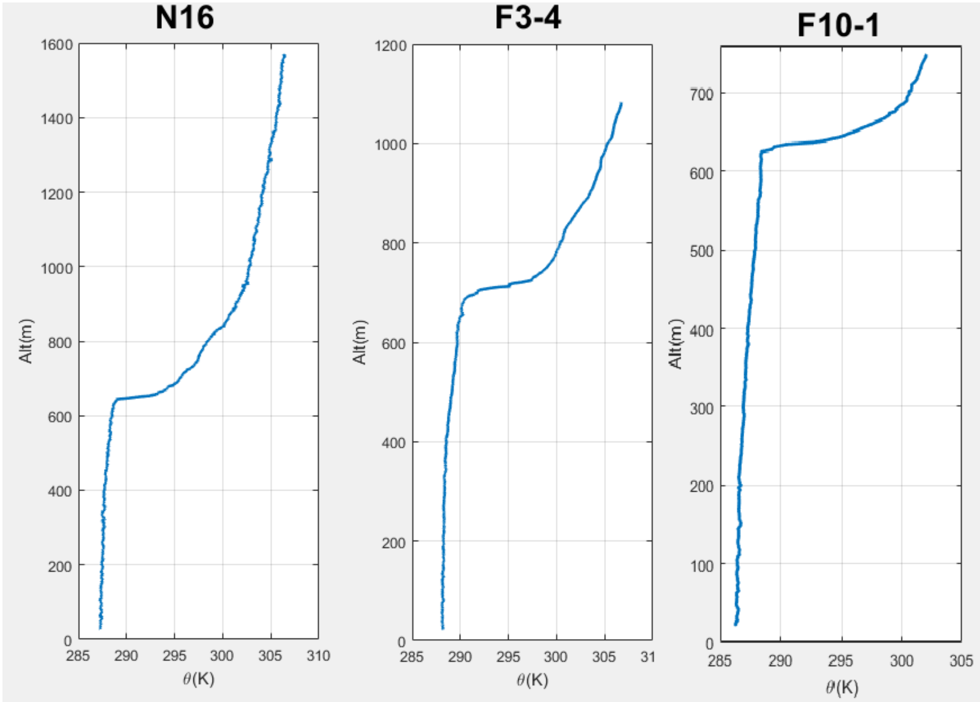
"The potential significance of nucleation in the EIL is that these particles impact the transfer of solar radiation owing to both directly scattering light and contributing to the marine atmosphere's cloud condensation nuclei (CCN) budget after growth to sufficiently large sizes. It is not possible with the current dataset to accurately calculate either nucleation rates in the EIL or the growth rate of nucleated particles to CCN-relevant sizes. However, a comparison of particle concentrations for $D_p$ between 3 and 10 nm in the EIL versus the SUB layer suggests that the nucleation rate in the former layer is greater by a factor of five. Others have reported particle growth rates in the Pacific Ocean MBL to be in the range of 3-10 nm $h^{-1}$ (Hoppel et al., 1994; Weber et al., 1998; Jennings and O'Dowd, 2000). Using a global aerosol microphysics model, Merikanto et al. (2009) estimated that in the marine boundary layer, 55% of CCN (0.2%) are from nucleation, with 45% entrained from the FT and 10% nucleated directly in the boundary layer. Therefore, nucleation in the EIL is significant for the CCN budget in the marine atmosphere."

5- Line 229: Should "..EIL presumably insight: : :" be ": : :FIL provide insight: : :" instead?

Response: Thank you for finding this error. We have made the change.

6- Figure 9 and related discussion. Could the influence of STBL on EIL aerosol properties be related to the strength of the inversion? I would suggest include the vertical profile of potential temperature to these plots.

Response: We thank the reviewer for this great suggestion. The figure below shows potential temperature profiles for the three cases examined. The potential temperature gradients for N16, F3-4, and F10-1 are 0.2, 0.18, and 0.19 K/m, respectively. The approximate equal potential temperature gradients for these three cases is indicative of a similar inversion strength. We added text in the manuscript to summarize this result: "An interesting feature of these three cases is that the strength of the temperature inversion at cloud top was similar ($d\theta/dz$ within the EIL was ~ 0.2 K $m^{-1}$)." We think that it is sufficient to add this text rather than to add another figure.



N16
F3-4
F10-1
Alt(m)
θ(K)