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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Large increases in N_{cn} and N_{ccn} together with a nucleation-modeparticle pool over the northwestern Pacific Ocean in the spring of 2014" *by* Juntao Wang et al.

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Anonymous Referee #2

General comments:

This paper presents potentially interesting information on particle number size distributions and concentrations as well as CCN concentrations in a marine boundary layer. However, before recommending the acceptance of this paper for publications, several issues need to be discussed in more detail and care. My main points in this regard are given below.

Response: Thanks for reviewer's comments. We try our best to revise the manuscript accordingly.

Section 1. The authors need to define the scientific aims of this study more clearly in the paper. It is not enough to say what is being studied in the paper. The sentence "Several new findings have. . ." on page 3 sounds strange here.

Response: Agree and revise accordingly. In the revision, page 2, lines 14-19, we add "Due to practical difficulties, direct observations of Nccn in the remote marine atmosphere are still limited and this restrains to gain reliable estimates of the Nccn over the oceans, leading to the results of aerosol-cloud interaction estimates suffering from a larger error (Rosenfeld et al., 2019; Sato and Suzuki, 2019).", Page 2, Lines 29-31, we add "Moreover, modeling studies show that the NWPO likely suffer from the largest increase in surface sea temperature and experience the largest increase in CO2 sink under warming climate (John et al., 2015; Lauvset et al., 2017). This further demonstrates the importance to study Ncn and NCCN and related potential climate effects therein." Page 3, Lines 3-5, we add "Through a comprehensive comparison with those observations in literature, we illustrated the characteristic of Ncn and Nccn over the NWPO in 2014 and revealed the changes in Ncn and Nccn against the results measured two decades ago. In addition, the influences of dust and BB aerosols on Ncn and Nccn were also analyzed on the monthly time scale."

Section 2.1. Description of the methods is incomplete. Nothing is said about the detection limits of gas-phase instruments, or the performance of any of the instruments during the campaign. Was it tested whether the instruments (e.g. CCNC) performed during the measurements? The authors mention a correction factor of 1.25 for the FMPS but do not explain where this factor comes from. What are the potential effects of uncertainties in FMPS measurements on the results discussed in this paper? (something is mentioned on page 6, lines 23-26, but probably more is needed).

Response: We revise the method parts accordingly. The detection limits of gas mon-

itors have been added in the revision. Based on our recent measurements made by an on-line ion chromatography in remote marine atmospheres, we reluctantly used the data measured by the gas monitors to characterize the background concentrations of gaseous pollutants therein. We only used the data to help screen out ship self-emission. This has been clarified in the revision. It is almost impossible to test the performance of instruments during the cruise, but the instruments were tested after the campaign.

According to side-by-side measurements between the FMPS and a CPC during several campaigns before and after, the empirical coefficient of 1.25 was obtained. A comparison result has been added in supporting information (Fig. S2). The particle size reported by FMPS suffers from the errors against the results measured by the scanning mobility particle sizer (SMPS) (Lee et al., 2013), but the errors can be reasonably corrected using the empirical correction procedure proposed by Zimmenrman et al. (2015) to obtain highly consistent results with SMPS. We thereby conducted the correction in this study. This has been added in the revision.

Section 3.1, last paragraph. There is a larger number of papers reporting CCN concentrations in the scientific literature. What was the basis for selecting these few studies when comparing results from this study? And why a single study conducted in Arctic was chosen here?

Response: Agree. We add a long discussion on the comparison among those measurements in various marine atmospheres in the revision. Please see our revised Section 3.1.

Section 3.3. After the more than 20-year-old papers citied here, a large number of studies (even reviews) on marine number size distributions have been published. The authors should make better use of these, more recent studies.

Response: Agree. In the revision, we add "As reviewed by Vu et al. (2015), the particle number size distributions in the marine atmospheric boundary layer usually

showed two modes, such as Aitken mode and accumulation mode, with nucleation mode to be observed occasionally (Koponen et al., 2002; Ueda et al., 2016; Zhu et al., 2019). For example, the particle size number concentrations exhibited a bimodal distribution with an Aitken mode ($\sim$ 50 nm) and an accumulation mode (150-180 nm) during the fall campaign over the western North Pacific in 2008 (Mochida et al., 2011). The bimodal distributions were also reported during a winter campaign over the tropical and subtropical Pacific Oceans from 2011 to 2012 (Ueda et al., 2016) and during a campaign over the western North Atlantic in June-July 2013 (Kristensen et al., 2016). However, the Aitken mode and the accumulation mode were sometimes overlapped in the particle number size spectra measured over marginal seas influenced by polluted air masses (Lin et al., 2007; Nair et al., 2013; Zhu et al., 2019)."

Section 3.4. Again, there are a number of more recent airborne studies on new particle formation in and above MFL in the scientific literature.

Response: In the revision, Page 9, bottom paragraph, we add "Recent measurements further supported that NPF events most likely occurred in the free troposphere over different oceanic zones (Dadashazar et al., 2018; Rose et al., 2015; Sanchez et al., 2018; Takegawa et al., 2014). Several factors such as the lower temperatures and lower relatively humility, lower condensation sinks, mixed precursors from the continent and marine sources lower in free troposphere have also been argued why new particle formation occurred therein."

In the revised last paragraph of Section 3.4, we add "Including the increase in Ncn by NPF events, a few studies proposed that the nucleation mode particles may grow to be larger, even reach CCN size in the free troposphere (Rose et al., 2015; Sanchez et al., 2018) and have size growth during subsidence process from free troposphere to MBL. Sanchez et al. (2018) estimated that the contributions of NPF in the free troposphere to the Nccn at SS of 0.1% in the clean marine atmosphere over the North Atlantic were 31% and 33% in late-autumn and late-spring, respectively. Merikanto et al. (2009) reported that 55% of CCN at SS of 0.2% in the marine boundary layer were

from nucleation, with 45% entrained from the free troposphere and reminding 10% nucleated directly in the boundary layer. However, the growth of newly formed particles to CCN size was not observed in this study."

Section 3.5. What is the purpose of the two sentences on lines 27-29 in this section? Also the discussion at the end of this section is a bit confusing.

Response: In revision, we add "Regarding of the vertical distribution of the Nccn over the marine atmosphere, three scenario are hypothesized: 1) the Nccn aloft was larger than those in the atmosphere near the sea level; 2) the Nccn in vertical direction was homogenous; 3) the Nccn aloft was lower than those in the atmosphere near the sea level. Varying wind speeds may change the convection, which in turn affected the Nccn in the atmosphere near the sea level. For example, Clarke et al. (2013) reported that CCN activated in MBL clouds were strongly influenced by entrainment from the free troposphere (FT). Zheng et al. (2018) also argued that entrainment of FT aerosols was a vital source of accumulation mode particles over the eastern North Atlantic, which could activate as CCN easily."

The last part has been revised as "Based on vertical backward air mass trajectories (Fig. S6), air masses were transported mostly from the Asian continent at high altitude (>2000 m a.m.s.l.) to the reception zones, indicating that air masses were affected by the entrainment of FT aerosols. Therefore, it is reasonable to argue that the Nccn mixed downward from FT may be an important source of the Nccn in the MBL over the NWPO. However, modeling studies are needed to quantify the contribution in the future."

Technical issues:

Why do the authors use such complicated format when presenting concentrations (M+-N x 10-3). Would it be much simpler just to give the numbers as they are?

Response: Considering analytic errors of FMPS and the effective number of number

concentrations to be consistent with analytic errors, we used this format to present our results.

Page 6, line 11: Following those in the literature,. . . ????

Response: The part has been revised as "As proposed in previous studies, e.g., Dusek et al. (2006) and Kalivitis et al. (2015), the total number concentration (N>Dp) of particles larger than a threshold diameter (Dp) can be used as a proxy for the Nccn. Specifically, aerosol particles with the size exceeding 60∼70 nm could be activated as CCN at SS of 0.4% (Dusek et al., 2018). In this study, N>Dp with Dp varying from 50 nm to 80 nm was calculated and a linear correlation was conducted with the values of Nccn measured at SS of 0.4%. A good correlation was obtained between Nccn and N>60 nm, with the slope of 0.98 closer to unity and R2=0.94 (Fig. 3)."

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Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-1089/acp-2018-1089-AC2-supplement.pdf

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