Measurement Report: Seasonal variation and anthropogenic influence on cloud condensation nuclei (CCN) activity in the South China Sea: Insights from shipborne observations during summer and winter of 2021

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24 **Abstract**

25   Understanding seasonal variation in cloud condensation nuclei (CCN) activity and the impact of

26 anthropogenic emissions in marine environments is crucial for assessing climate change. In this study,

27 two shipborne observations in the South China Sea (SCS) during the summer and winter of 2021 were

28 conducted. During summer, higher particle number concentrations but lower mass concentrations of non-

29 refractory submicron particles (NR-PM$_1$) were observed. These differences were attributed to the

30 dominance of particles in the Aitken mode during summer and in the accumulation mode during winter.

31 Moreover, particles during summer were more hygroscopic with higher activation ratios (ARs) at all

32 supersaturation (SS). Based on backward trajectory analysis, the whole campaign was classified into

33 terrestrial and mixed air mass influence periods. Particles measured during the terrestrial period

34 consistently exhibited lower hygroscopicity values. Additionally, minor variations were shown for all

35 NR-PM$_1$ components under different air mass influences during summer, while the mass fraction of

36 nitrate increased significantly under terrestrial influence during winter. Particle number size distribution

37 (PNSD) exhibited unimodal distribution during terrestrial period and bimodal distribution during mixed

38 air mass influence period, with winter displaying a more pronounced bimodal pattern than summer. The

39 impact of PNSD on AR was greater than on aerosol hygroscopicity in summer, and vice versa in winter.

40 During terrestrial period, significant variations in PNSD were observed with the offshore distance, and

41 the largest variation was seen in Aitken mode during both summer and winter. Meanwhile, aerosol

42 hygroscopicity shows an increasing trend with the offshore distance, which is primarily attributed to the

43 increase of sulfate fraction during summer and the decrease of the black carbon fraction during winter.

44 Using a single parameterized PNSD in the N$_{CCN}$ prediction can lead to errors exceeding 100% during

45 both summer and winter, with dominant terrestrial air masses in the SCS atmosphere, while using a

46 constant hygroscopicity parameter would lower the errors in the N$_{CCN}$ prediction (~15% during winter

47 and ~10% during summer). Our study shows significant differences in aerosol properties between winter

48 and summer seasons and highlights the influence of anthropogenic emissions on the CCN activity in the

49 SCS.

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1.Introduction

Aerosols can act as cloud condensation nuclei (CCN), influencing cloud formation, lifespan, and albedo, thus indirectly impacting global radiative balance (Fletcher et al., 2011; Albrecht, 1989). The aerosol-cloud interaction currently represents the largest uncertainty in radiative forcing within climate models, ranging from -1.7 to -0.3 W m^{-2} (IPCC, 2021). This uncertainty can be attributed to the significant spatiotemporal variability in the aerosol size distribution and the ability of atmospheric aerosol particles acting as CCN (CCN activity) (Fitzgerald, 1973). Thus, field measurements of aerosol size distribution and physicochemical properties are needed to better understand the radiative forcing exerted by atmospheric aerosol particles.

Previous studies suggest that particle number size distribution (PNSD) is a primary factor influencing CCN concentrations (Dusek et al., 2006; Rose et al., 2010; Pöhlker et al., 2016; Burkart et al., 2011). The PNSD can account for 84–96% of the variability (Dusek et al. (2006) in CCN concentrations (N_{CCN}), while CCN activities may also play a significant role in CCN concentrations (Quinn et al., 2008; Cai et al., 2018; Ovadnevaite et al., 2017; Liu et al., 2018; Crosbie et al., 2015), which are primarily governed by the particle size, chemical composition, surface tension, and hygroscopicity (Köhler, 1936; Seinfeld and Pandis, 2016). Among these factors, the impact of hygroscopicity on CCN activities has received great attention in recent years (Petters and Kreidenweis, 2007; Ajith et al., 2022; Rose et al., 2010). Petters and Kreidenweis (2007) proposed the κ- Köhler theory based on the Köhler theory to quantify the ability of aerosol particles to absorb moisture and become CCN based on the aerosol hygroscopicity parameters (κ). Ajith et al. (2022) showed that 64% of particles can be activated as CCN when κ is equal to 0.37, whereas when κ decreases to 0.23, only 48% of particles can be activated.

Previous field observations have indicated significant seasonal variations in PNSD and hygroscopicity under both terrestrial and marine environments, leading to the seasonal variations in N_{CCN} (Crosbie et al., 2015; Schmale et al., 2018; Burkart et al., 2011; Bougiatioti et al., 2009; Sihto et al., 2011; Leena et al., 2016; Ross et al., 2003; Gras and Keywood, 2017; Quinn et al., 2019). Crosbie et al. (2015) revealed that in the urban area of Arizona particles had larger sizes, higher hygroscopicity, and N_{CCN} was also higher during winter, while a higher abundance of smaller particles was observed during summer owing to stronger photochemical reactions. In pristine environments like mountain, coastal, and forested

80   regions, seasonal variations in N_{CCN} and PNSD were more pronounced than urban and rural areas

81   (Schmale et al., 2018). Pöhlker et al. (2016) observed significant differences in N_{CCN} between the wet

82   and dry seasons in the Amazon rainforest, while the κ values remained relatively stable. They also noted

83   increased particle concentrations and aerosol hygroscopicity, both subject to the impact of long-range

84   transport originating from anthropogenic emissions. Observations in marine areas during different

85   seasons are relatively scarce compared with those in inland areas. Gras (1995) found that both particle

86   concentration and N_{CCN} in the Southern Ocean reached their peaks during summer and gradually decrease

87   to their valleys in winter. Quinn et al. (2019) showed that sea spray aerosols make a relatively significant

88   contribution to N_{CCN} only during winter in the Western North Atlantic, while in other seasons, the primary

89   contribution comes from biogenic aerosols oxidized from dimethyl sulfide (DMS). Zheng et al. (2020)

90   revealed that sulfate dominates the particle condensational growth to CCN sizes during summer in the

91   North Atlantic, while secondary organic aerosols played a significant role in particle growth throughout

92   all seasons. These results indicate that CCN activity and concentration could vary in a large range during

93   different seasons. Thus, further observations across different seasons in marine environments are needed

94   to enhance our understanding of marine CCN activities and their seasonal variations.

95   The South China Sea (SCS) is located between the Asian continent to the north and the Indonesian

96   archipelago to the south. The aerosols in the SCS can be influenced by ship emissions, marine

97   background emissions, and transport of terrestrial air masses from China, Indochinese Peninsula, and

98   Philippines, making their sources relatively complex (Geng et al., 2019; Liang et al., 2021; Sun et al.,

99   2023). The complex aerosol sources in the SCS lead to the complexity of PNSD and CCN activity in this

100   area. Atwood et al. (2017) found that under the influence of marine air masses, particles exhibited a

101   significant bimodal distribution with a κ of 0.65, whereas under the influence of continental air masses,

102   a unimodal distribution and a κ of 0.4 were observed. Another summer study in the northern SCS revealed

103   a predominantly unimodal aerosol PNSD with a peak centering at 60–80 nm and an average κ value of

104   0.4 (Cai et al., 2020). Moreover, particle and N_{CCN} decreased with increasing offshore distance,

105   highlighting the significant influence of mainland China on CCN activity in the northern SCS (Cai et al.,

106   2020). However, our understanding on CCN activity, particularly the variations in CCN activity in the

107   SCS across different seasons, remains largely unknown. Conducting observational studies on CCN

108   activity across different seasons would promote our understanding of CCN activity and, ultimately,

109   reduce uncertainties related to aerosol-cloud interactions and radiative forcing in the SCS.

110   In this study, we conducted two shipborne observations in the SCS during summer (May 5–June 9,

111   2021) and winter (December 19–29, 2021). Our observations with online instruments focused on

112   measuring aerosol chemical composition, PNSD, and CCN activation in the region. Our results provide

113   valuable insights into the differences in CCN activity between winter and summer, as well as the

114   influence of terrestrial transport on CCN activity in the SCS across different seasons.

115   **2. Methodology**

116   **2.1 Cruise information and onboard measurements**

117   **2.1.1 Cruise information**

118   This study consists of two research cruises conducted during the summer and winter of 2021,

119   respectively. The summer and winter cruises were carried out respectively by the vessels "Tan Kah Kee"

120   and "Sun Yat-sen University". The "Tan Kah Kee" is an oceanographic research vessel with a length of

121   77.7 meters, a beam of 16.24 meters, and a displacement of 3611 tons. The "Sun Yat-sen University" is

122   a comprehensive oceanographic training vessel with a total length of 114.3 meters, a beam of 19.4 meters,

123   and a displacement of 6880 tons.

124   The first cruise was from May 5th to June 9th, 2021. The cruise started from Xiamen Port and

125   traversed from the northern to the central-southern South China Sea, and then circled back near Hainan

126   Island, and finally returned to Xiamen Port. The second cruise was from December 19th to December

127   29th, 2021. It began from Gaolan Port in Zhuhai and reached the vicinity of Yongxing Island, and

128   ultimately returned to Gaolan Port (Fig. 1). On both cruises, most of the instruments were housed in a

129   single compartment and the sampling lines were extended from the window of the compartment to the

130   height of the ship's bridge (Fig. S1).

131   **2.1.2 Size-resolved cloud condensation nuclei activity measurement**

132   The size-resolved CCN activity was measured with a combination of a scanning mobility particle

133   sizer (SMPS) system and a cloud condensation nuclei counter (model CCNc-200, DMT Inc., USA), the

134   SMCA method initially proposed in Moore et al. (2010). The SMPS system consisted of a differential

135   mobility analyzer (DMA; model 3082, TSI., Inc.) and a condensation particle counter (CPC; model 3756,

136   TSI Inc.). The SMPS and the CCNc system were set to measure PNSD and size-resolved CCN number

137   concentration at a mobility size range of 10–500 nm and 10–593 nm in summer and winter campaign,

138   respectively.

139   The supersaturation (SS) of the CCNc was set at 0.2 %, 0.4 %, and 0.7 % in summer campaign and

140   0.1%, 0.2 %, 0.4 %, and 0.7 % in winter campaign, respectively. Before the measurements, the CCNc

141   was calibrated with ammonium sulfate ($(NH_4)_2SO_4$) particles at each set SS. Detailed description of the

142   instrument configuration and calibration can be found in Cai et al. (2018).

143   **2.1.3 Aerosol chemical composition measurement**

144   The chemical composition of atmospheric non-refractory submicron particulate matter (NR-PM_1),

145   including sulfate, nitrate, organics, ammonium, and chloride, was measured using an online time-of-

146   flight ACSM (ToF-ACSM; Aerodyne Inc., USA). The sampling time of the ToF-ACSM was

147   approximately 10 min. The relative ionization efficiency (RIE) values of the instrument were calibrated

148   using ammonium nitrate (NH_4NO_3) and ammonium sulfate ($(NH_4)_2SO_4$) both before the start and after

149   the completion of the campaigns. The RIE values for ammonium were 3.31 and 3.33 during the summer

150   and winter, respectively, while the ones for sulfate were 1.02 and 0.81 during the summer and winter,

151   respectively. The collection efficiency (CE) was determined as shown in Sun et al. (2023) and time-

152   independent CE values were used in this study. Detailed CE calculation can be found in the

153   supplementary (Text S2). The organic carbon (OC)/elemental carbon (EC) concentrations in $PM_{2.5}$ were

154   measured using a semi-continuous OC/EC analyzer (Model-4, Sunset Laboratory Inc., USA) based on

155   the thermal optical transmittance technique and detailed measurement process can be found in Sun et al.

156   (2023). The black carbon concentrations were measured with an aethalometer (AE33, Magee Scientific).

157   **2.1.4 Trace Gas and meteorological parameter measurements**

158   The concentrations of trace gases (CO, O_3, SO_2, and NOx) were measured using gas monitors

159   (T400U, T100U, and T200U; Teledyne API Inc., USA). The meteorological elements, including

160   temperature, relative humidity, wind speed, and wind direction, were measured by the combined

161   automatic weather station onboard the vessels.

162 **2.2 Data analysis**

163 **2.2.1 CCN activation**

164   The size-resolved number concentration of total praticle and cloud condensation nuclei were

165 obtained from the SMPS and CCNc thourgh the SMCA method. The activation diameter was determined

166 by fitting the activation ratio (AR, N_{CCN}/N_{CN}) and dry diameter at each supersaturation through the

167 following equation:

168 $$\frac{N_{CCN}}{N_{CN}} = \frac{B}{1+\left(\frac{D_P}{D_{50}}\right)^C},$$ (1)

169 where D_P represents dry particle diameter (nm); B, C, and D_{50} are the three fitting parameters,

170 representing the asymptote, the slope, and the inflection point of the sigmoid, respectively (Moore et

171 al., 2010). The D_{50} parameter, also known as the critical diameter, corresponds to the particle size at

172 which 50% of the particles are activated at a specific SS.

173   The hygroscopicity parameter (κ) which represents CCN activity according to κ-Köhler equation is

174 calculated as follows (Petters and Kreidenweis, 2007):

175 $$\kappa = \frac{4A^3}{27D_{50}^3(lnS_C)^2}, \quad A = \frac{4\sigma_{s/a}M_W}{RT\rho_W}$$ (2)

176 where ρ_w is the density of pure water (about 997.04 kg m^{-3} at 298.15 K), M_W is the molecular weight of

177 water (0.018 kg mol^{-1}), $\sigma_{s/a}$ corresponds to the surface tension of the solution-air interface and is assumed

178 to be equal to the surface tension of pure water ($\sigma_{s/a}$=0.0728 N m^{-1} at 298.15 K), R is the universal gas

179 constant (8.314 J mol^{-1} K^{-1}), T denotes thermodynamic temperature in kelvin (298.15 K), and D_{50} is the

180 critical diameter (in m).

181 **2.2.2 CCN concentration and activation ratio calculation**

182   The CCN concentration (N_{CCN}) can be predicted based on particle number size distribution (PNSD)

183 and D_{50} at a specific SS. It can be calculated by the following equation (Cai et al., 2018):

184 $$N_{CCN}(SS) = \int_{D_{50}(SS)}^{\infty} N_{CN}(D_P)dD_p$$ (3)

185 where N_{CCN}(SS) is CCN concentration at a specific SS, D_{50}(SS) is the activation diameter at a specific

186 SS from the SMCA method and $N_{CN}(D_P)$ is the particle number concentration under specific diameter

187 from SMPS measurement.

188   The AR can be calculated by:

189 $$AR = \frac{\int_{D_{50}(SS)}^{\infty} N_{CN}(D_P)dD_p}{\int_0^{\infty} N_{CN}(D_P)dD_p} \qquad (4)$$

190   To investigate the effect of PNSD and D_{50} on N_{CCN} and AR, we defined delta N_{CCN} (ΔN_{CCN}) and

191 delta AR (ΔAR). They are calculated by following equations:

192 $$\Delta N_{CCN} = \frac{N_{CCN,sim} - N_{CCN,actual}}{N_{CCN,actual}} \qquad (5)$$

193 $$\Delta AR = \frac{N_{CCN,actual} - N_{CCN,sim}}{N_{CN,actual}} \qquad (6)$$

194 The subscript "actual" represents the actual measured value, while the subscript "sim" represents the

195 simulated value.

2.2.3 Primary and secondary organic carbon concentration calculation

197   The concentrations of primary organic carbon (POC) and secondary organic carbon (SOC) were

198 calculated according to following equation:

199 $$POC = (OC/EC)_{pri} \times EC \qquad (7)$$

200 $$SOC = OC_{total} - (OC/EC)_{pri} \times EC \qquad (8)$$

201 where $(OC/EC)_{pri}$ is the OC/EC ratio in freshly emitted combustion aerosols, and OC_{total} and EC are

202 available from ambient measurements. The $(OC/EC)_{pri}$ was obtained from the minimum R squared (MRS)

203 method (Wu and Yu, 2016). The MRS approach provides more accurate estimation of SOC than

204 $(OC/EC)_{min}$ or $(OC/EC)_{10\%}$ approach and the results obtained from the MRS method are sensitive to the

205 magnitude of measurement uncertainty, but the bias does not exceed 23% if the uncertainty is within 20%

206 (Wu and Yu, 2016). In this study, the $(OC/EC)_{pri}$ values were 3.65 and 0.25 in summer when affected by

207 terrestrial air masses and terrestrial-marine mixed air masses, while they were 2.82 and 0.82 in winter

208 when affected by terrestrial air masses and terrestrial-marine mixed air masses, respectively. The

209 categorization method based on the influence of different air masses will be introduced in the next section.

2.2.4 Backward trajectory simulation

211   Backward trajectory calculations were performed using MeteoInfo, an open-source software (Wang,

212 2014) to determine potential source origins. Weekly GDAS1 (Global Data Assimilation System at a

213 resolution of 1°) files were downloaded from the NOAA Air Resource Laboratory (ARL) website

214 (https://www.ready.noaa.gov/gdas1.php). The calculation of backward trajectories is performed every 12

215   hours based on the location of the ship, generating 48-hour backward trajectories at 50m, 150m, 500m,

216   and 1000m heights.

217   **3. Results and discussion**

218   **3.1 Overview**

219   Figure 2 shows the timeseries of PNSD (a1 and a2), PM$_1$ mass concentrations and fractions (b1 and

220   b2, c1 and c2), number concentrations of CCN (d1 and d2), and hygroscopicity κ-values (e1 and e2)

221   during two campaigns in summer and winter. Based on the backward trajectories and source origins, both

222   campaigns were classified into two periods: the period influenced by terrestrial-marine mixed air masses

223   (31%) and the period influenced by terrestrial air masses only (69%) (Fig. S3). During the summer,

224   terrestrial air masses primarily traverse through the Philippines region, whereas during the winter, they

225   predominantly originate from mainland China. As shown in Fig. 2, a higher total particle number

226   concentration during summer (11195 cm^{-3}) than during winter (6358 cm^{-3}) was obtained when the marine

227   atmosphere was mainly influenced by terrestrial air masses. This is in line with previous studies that a

228   higher particle number concentration of exceeding 15000 cm^{-3} was observed in Manila (the largest port

229   city in the Philippines) than that of approximately 10000 cm^{-3} reported in Guangzhou and Hong Kong

230   (Liu et al., 2008; Cai et al., 2017). Similar particle concentrations (2114 cm^{-3} and 1840 cm^{-3}) in winter

231   and summer were shown when the marine atmosphere was predominantly influenced by mixed air

232   masses. Interestingly, they are much lower than those (~3400 cm^{-3}) observed in the northern South China

233   Sea (Cai et al., 2020), but higher than those observed in the remote SCS (975 cm^{-3}) when similar mixed

234   influences of air masses (Atwood et al., 2017).

235   The average mass concentration of NR-PM$_1$ was 3.60 μg m^{-3} in summer and significantly increased

236   to 18.11 μg m^{-3} in winter. However, this summer concentration was much lower than the concentrations

237   (9.11 μg m^{-3} and 10.65 μg m^{-3}) measured in similar summer periods of 2018 and 2019 in the northern

238   SCS (Liang et al., 2021; Sun et al., 2023). As aforementioned, the particle number concentrations were

239   higher in summer than in winter when the atmosphere was primarily influenced by terrestrial air masses.

240   However, the particles in summer were predominantly in the Aitken mode, resulting in a lower

241   contribution to the mass loading than those in winter which were mainly in the accumulation mode (Fig.

242   3). Additionally, higher NR-PM$_1$ concentrations were found in both summer and winter under the

9

243   influence of terrestrial air masses than mixed ones, with decreases of approximately 0.6 μg m^{-3} (from

244   3.84 to 3.20 μg m^{-3}) and 17 μg m^{-3} (from 23.52 to 6.32 μg m^{-3}) in summer and winter, respectively. The

245   dramatic decrease for the latter case indicated a more pronounced impact of inland anthropogenic

246   emissions on the aerosol mass concentration in the SCS region during winter.

247   As shown in Fig. 2 (d1 and d2), the N_{CCN} values at three supersaturation levels (0.2%, 0.4%, and

248   0.7%) in winter (1660, 2356, and 3053 cm^{-3}) were lower than those in summer (2899, 5450, and 5770

249   cm^{-3}), respectively. Specifically, the N_{CCN} (5450 cm^{-3}) at 0.4% SS in this study during summer is much

250   higher than that (1544 cm^{-3}) measured at a similar SS (0.34%) in summer 2018 in the similar northern

251   SCS region (Cai et al., 2020). This is primarily due to most of the particles being in the Aitken mode

252   particles during summer (Figs. 3a and c). For comparison, the average total particle number concentration

253   in summer is twice that in winter, however, no such a significant difference in CCN concentration was

254   seen.

255   The median values of κ were approximately 0.5 in summer and 0.3 in winter (Fig. 2e), respectively.

256   During summer, the median κ value was slightly higher than that (~ 0.4) measured in 2018 in the similar

257   northern SCS (Cai et al., 2020), but lower than that (0.65) in the southern SCS (Atwood et al., 2017).

258   Comparatively, under the influence of terrestrial air masses, the κ values ranged from 0.41 to 0.50 (Fig.

259   3a), close to the above value of 0.4 in 2018 (Cai et al., 2020). Furthermore, under the influence of mixed

260   air masses, the κ values ranged from 0.49 to 0.61 (Fig. 3c), similar to those observed in the southern SCS

261   (0.54) and the Western North Pacific (0.49–0.64), but lower than those in the southern SCS (0.65) and

262   the Western North Pacific (0.48–0.84) under the influence of marine air masses (Atwood et al., 2017;

263   Kawana et al., 2022). During winter, under the influence of terrestrial air masses, the κ values ranged

264   from 0.17 to 0.38 (Fig. 3b), very close to those (0.14–0.37) measured in Guangzhou (Fig. 3d) (Cai et al.,

265   2018). This specific winter scenario indicates that aerosols in the northern SCS are significantly

266   influenced by the air masses transported from the mainland China during winter season. Compared to

267   terrestrial air masses, the κ values were respectively higher at the set SS under the influence of mixed air

268   masses (Fig. 3d). The above differences on hygroscopicity are likely attributed to variations in aerosol

269   chemical composition during different seasons and under the influence of different air masses. In both

270   winter and summer, the PNSD exhibits a unimodal distribution under the influence of terrestrial air

271   masses, while it shows a bimodal distribution under the influence of mixed air masses (Fig. 3). The

272   bimodal distribution may result from a mixture of particles from marine primary emissions and from

273 transported anthropogenic emissions across the region, while the unimodal distribution was primarily

274 composed of particles from inland-transported anthropogenic emissions (Frossard et al., 2014; Atwood

275 et al., 2017).

3.2 Impact of chemical composition on hygroscopicity

277   Chemical composition is a crucial factor influencing aerosol hygroscopicity due to distinct

278 hygroscopic properties exhibited by each chemical component (Petters and Kreidenweis, 2007). We

279 observed two significant differences in aerosol hygroscopicity between summer and winter (Fig. 3).

280 Firstly, aerosol hygroscopicity was considerably higher in summer than that in winter. Secondly, aerosol

281 hygroscopicity under terrestrial-influencing air masses was lower than that under mixed-influencing air

282 masses. Additionally, hygroscopicity decreased with increasing particle size in summer, whereas

283 opposite effect was observed in winter. In this section, we primarily discuss the differences in aerosol

284 hygroscopicity between summer and winter, as well as the variations observed when influenced by

285 different air masses, from the chemical composition perspective.

3.2.1 Impact of inorganic components

287   In summer, the highest proportion of NR-PM$_1$ components measured by ACSM is sulfate (40.3%),

288 followed by organic (37.3%), ammonium (14.0%), nitrate (6.2%), and chloride (2.3%) (Fig. 4). However,

289 in winter, organic (39.2%) has the highest proportion, followed by nitrate (22.1%), sulfate (19.5%),

290 ammonium (17.7%), and chloride (1.5%) (Fig. 4). Compared to summer, the significant increase of

291 nitrate proportion and decrease of sulfate in winter were mainly attributed to the difference in air masses.

292 Winter terrestrial air masses originate from mainland China and primarily pass through the PRD region

293 to reach the South China Sea, while the summer ones primarily originate from the Philippines region.

294 Stronger transportation and industrial emissions in the PRD than in the Philippines lead to higher NOx

295 concentrations and hence higher nitrate concentrations (Wang et al., 2019). A high proportion of nitrate

296 sulfate in the PRD during winter was previously reported in NR-PM$_1$ (Yang et al., 2022), whereas sulfate

297 was found the major component in the Philippines (Tseng et al., 2019).

298   Previous studies showed that nitrate has slightly higher hygroscopicity than sulfate (Gysel et al.,

299 2007; Topping et al., 2005). For example, the κ values for ammonium sulfate ((NH$_4$)$_2$SO$_4$), ammonium

300 bisulfate (NH$_4$HSO$_4$), and ammonium nitrate (NH$_4$NO$_3$) are 0.48, 0.56, and 0.58, respectively (Huang et

301 al., 2022). In some marine environments, aerosols exhibit a high acidity with low ammonium

302 concentrations, where sulfate and nitrate primarily exist in the form of sulfuric acid (H_2SO_4, κ: 0.7) and

303 nitric acid (HNO_3, κ: 0.85), respectively, thereby enhancing aerosol hygroscopicity (Chang et al., 2011;

304 Siegel et al., 2022). Here, we use the ratio of NH_4^+ (measured) to NH_4^+ (predicted) to roughly determine

305 an aerosol being "acidic" or "alkaline" (Guo et al., 2015): more acidic if this ratio is smaller one because

306 the imbalance indicates the possible presence of acidic compounds such as H_2SO_4, HNO_3, or HCl in the

307 aerosol (Zhang et al., 2021). The median values of this ratio in summer and winter are 0.82 and 1.11,

308 respectively, exceeding the threshold of 0.75 (Fig. S5), a rough estimate for equal molecular numbers of

309 $(NH_4)_2SO_4$ and NH_4HSO_4. This indicates that sulfate and nitrate are primarily present in the form of

310 $(NH_4)_2SO_4$ and NH_4NO_3 both in summer and winter in the SCS. Since the hygroscopicity values of

311 $(NH_4)_2SO_4$ and NH_4NO_3 are similar, changes in their proportions may not be the primary driver of the

312 observed aerosol hygroscopicity variations between winter and summer. Interestingly, we found that the

313 mass concentrations of BC and EC were higher in winter (1.79 μg m^{-3} and 1.16 μg m^{-3}) than those in

314 summer (0.61 μg m^{-3} and 0.64 μg m^{-3}), which might be one of the reasons for the lower aerosol

315 hygroscopicity in winter because of the hydrophobicity nature of BC (κ=0) (Weingartner et al., 1997;

316 Siegel et al., 2022). Another reason may be attributed to the hygroscopicity variations of organic

317 components (κ_{org}), which will be discussed in the following section.

318   As introduced at the beginning of the section, the hygroscopicity decreased with increasing particle

319 size during summer, in contrast to the increasing trend during winter, which may be attributed to the

320 oxidation of dimethyl sulfide (DMS). DMS oxidation produces methanesulfonic acid (MSA) which is

321 further oxidized to form non-sea salt (NSS) sulfate in smaller particles (Raes et al., 2000). The ratio of

322 sulfate to MSA (SA/MSA) can be used to assess the impact of DMS oxidation and anthropogenic

323 emissions on sulfate (Savoie et al., 2002; Zhang et al., 2007; Cai et al., 2020). A lower ratio indicates a

324 more significant influence of DMS oxidation on sulfate, while a higher ratio suggests a greater influence

325 of anthropogenic emissions. Previous studies have found that SA/MSA is approximately 15–625 in the

326 northern South China Sea region, significantly higher than that (18–20) in the open ocean (Zhang et al.,

327 2007; Savoie et al., 2002). Cai et al. (2020) reported this ratio in a range of 100–10000 using Modern-

328 Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) reanalysis data in a

329 similar northern SCS region. In this study, since we could not accurately distinguish MSA from other

330   organic components using Tof-ACSM due to its low mass resolution, we employed the same method to

331   analyze the SA/MSA distribution in the South China Sea region during the observation period (Fig. S6).

332   Our results showed that this ratio ranged from approximately 100 to 3000 in summer, whereas it exceeded

333   10000 in winter, indicating a greater impact of anthropogenic emissions than DMS oxidation on the NSS

334   sulfate in the SCS, with a more pronounced influence in winter than in summer. Higher DMS

335   concentrations in summer than in winter are likely due to higher sea surface temperatures which promote

336   the production rate of DMS by phytoplankton (Bates et al.,1987). Therefore, it is likely that there is a

337   higher amount of NSS sulfate produced through DMS oxidation during summer, contributing to the

338   sulfate production and hence increasing aerosol hygroscopicity in smaller particles during summer.

339   **3.2.2 Impact of organic components**

340   The mass concentrations of organics exhibit significant differences between winter (7.15 $\mu g\ m^{-3}$)

341   and summer (1.34 $\mu g\ m^{-3}$), while the total fractions in NR-PM$_1$ remain relatively consistent (40% and

342   37% in winter and summer, respectively). Despite their similar proportions in winter and summer, the

343   differences in the organic components may likely play an important role in the variations of

344   hygroscopicity. Previous studies showed that the measured hygroscopic parameter was linearly

345   correlated with the organic mass fraction (f_{org}) (Bhattu et al., 2016; Huang et al., 2022; Dusek et al., 2010).

346   Here, we employed linear regression to relate κ obtained at 0.2% SS ($\kappa_{ss:0.2\%}$) to f_{org} measured by ToF-

347   ACSM (Fig. 5). Two reasons for choosing $\kappa_{ss:0.2\%}$: (1) It represents hygroscopicity of larger particles

348   (~100 nm) than that at higher SSs. (2) The κ values at 0.1% SS ($\kappa_{ss:0.1\%}$) is only available in winter. In

349   addition, the measured mass concentrations focused on larger particles which were represented by those

350   of $\kappa_{ss:0.2\%}$. We consider κ as hygroscopicity of organics (κ_{org}) when f_{org} approaches 1 and as that of

351   inorganics (κ_{inorg}) when f_{org} is nearly 0.

352   During summer and winter under the influence of terrestrial air masses, the κ_{org} value was

353   approximately 0.09, while under the influence of mixed air masses, it was approximately 0.21 and 0.20,

354   respectively. This result suggests that no significant differences of κ_{org} were found between summer and

355   winter under the same influence of air masses. However, pronounced differences of κ_{org} were indeed

356   observed under the influence of different air masses which may be associated with different types of

357   organic compounds responsible for hygroscopicity. This was further supported by variations of the POC

358   and SOC fractions. In general, POC is associated with primary organic aerosol (POA), primarily

359   originating from combustion sources, such as fossil fuel, biomass burning, and biogenic emissions, while

360   SOC is associated with secondary organic aerosol (SOA), primarily originating from the oxidation of

361   POA (Wu and Yu, 2016). In addition, SOC exhibits higher hygroscopicity than POC (Jayachandran et

362   al., 2022; Safai et al., 2014). Previous studies have indicated that highly aged organic compounds exhibit

363   higher hygroscopicity than the less oxidized counterparts (Lambe et al., 2011; Jimenez et al., 2009).

364   Previous studies showed that the κ_{org} values are in a range of 0.02–0.17 when the O/C ratio varies from

365   0.2 to 0.6 (Jimenez et al., 2009) and κ_{POA} (κ corresponds to POA, similar to κ_{org}) and κ_{SOA} are respectively

366   in the ranges of 0–0.1 and 0.1–0.3 due to different types of organic aerosol (Liu and Wang, 2010; Kuang

367   et al., 2020). A higher proportion of POC was observed during the terrestrial periods, whilst a significant

368   increase in the fraction of SOC was shown under the influence of mixed air masses (Figs. 4 h-m). As a

369   result, the increasing oxidation degree of SOA and the decreasing proportion of POA lead to higher κ_{org}

370   under mixed air masses than those under continental air masses. This is also consistent with a recent

371   study in the SCS (Sun et al., 2023), revealing that a combined effect of photo-oxidation and liquid-phase

372   reactions play an essential role in the BBOA aging processes.

3.3 Differences in the CCN activation ratio during different seasons

374   The CCN activation ratio (AR), which quantifies the number fraction of aerosol particles capable

375   of acting as CCN at a specific supersaturation level, is an important parameter for characterizing the

376   CCN activity (Dusek et al., 2006). In addition, AR is also a useful parameter in the CCN prediction

377   (Pruppacher, 2010; Deng et al., 2013). Figure 6 illustrates the variations in AR and N_{CCN} under the

378   influence of different air masses during different seasons. The median AR values were higher in summer

379   (0.39, 0.67, and 0.85 at 0.2%, 0.4%, and 0.7% SS, respectively) than in winter (0.21, 0.36, 0.49, and 0.64

380   at 0.1%, 0.2%, 0.4%, and 0.7% SS, respectively). In summer under terrestrial air masses, the ARs (0.34–

381   0.81 at 0.2%–0.7% SS) in this study were lower than those (0.49–0.85 at 0.18%–0.59% SS) in a previous

382   study in the same region (Cai et al., 2020), but higher than those (0.18–0.48 at 0.11%–0.60% SS) in the

383   Western North Pacific (Kawana et al., 2022). However, under mixed air masses, the AR values were

384   higher than those (0.31–0.71 at 0.18%–0.59% SS) reported in Cai et al. (2020) and close to those (0.42–

385   0.85 at 0.11%–0.60% SS) in the Western North Pacific (Kawana et al., 2022). In winter under terrestrial

386   air masses, the AR values were in a range of 0.14 to 0.59 at 0.1%–0.7% SS, consistent with those

387   measured in the Guangzhou (0.26–0.64 at 0.1%–0.7% SS) and Hong Kong (0.16–0.65 at 0.15%–0.7%

388   SS) (Cai et al., 2018; Meng et al., 2014).

389   The differences of AR between different seasons under different air masses could be attributed to

390   different PNSD and hygroscopicity. When the PNSD concentrates particles in the size range beyond the

391   D_{50}, more particles can be activated as CCN, leading to higher ARs. Meanwhile, more hygroscopic

392   particles intend to have smaller D_{50} values with which more particles can be activated. Previous studies

393   showed that the influence of the above two factors may vary under different environments. The PNSD

394   played a more important role in most cases in continental area (Dusek et al., 2006; Tao et al.,2021), while

395   hygroscopicity can also have significant effects in some environments, such as boreal forest and coastal

396   area (Roldin et al., 2019, Bougiatioti et al., 2016). Here, we investigate the relative importance of aerosol

397   PNSD and chemical composition (hence hygroscopicity) in the SCS during different seasons under

398   different air masses. We define ΔAR as the difference between the actual and estimated AR using Eq.

399   (7), following a procedure illustrated in Fig. S8. AR was calculated using Eq. (4) based on the average

400   D_{50} and the PNSD obtained during different seasons and periods (Fig. S8). Figure 7 shows that the ΔAR

401   values at 0.2%–0.7% SS between summer and winter due to D_{50} were from -22% to 29%, while they

402   were from -10% to 12% due to PNSD, indicating more significant influence of D_{50} than PNSD on N_{CCN}

403   in the South China. In fact, the peak diameters of PNSD during both winter and summer are around 70

404   nm, and their variations are relatively small compared to the changes in D_{50} (Fig. S9).

405   We further investigate the influence of PNSD and hygroscopicity on AR under different air masses

406   in the same season. In summer, the influence of PNSD on the AR can reach 6.0% to 9.8% between

407   different periods (Figs. 7 b1 and c1), with the most significant impact at 0.2% SS (D_{50} > 90 nm). This

408   can be attributed to the fact that large variations in accumulation mode particles significantly impact AR

409   with peaking at a larger size for terrestrial air masses (151 nm) than mixed air masses (140 nm) (Figs. S9

410   a1-a3). In comparison, the impact of hygroscopicity (based on D_{50}) on AR is relatively small, for example,

411   the influence is estimated to be around 1.3%–3.4% at 0.2% and 0.7% SS, and ~6% at 0.4% SS. In winter,

412   the impact of hygroscopicity on AR at all SS levels (2.6%-5.1%) was more significant than that of PNSD

413   (1.2%–3.2%) (Fig. 7). We attribute this difference to two possible reasons: (1) Particles tend to be more

414   concentrated in larger sizes (Fig. S9) and the changes in PNSD under different air masses have a

415   relatively minor impact on AR. (2) the D_{50} values in winter are larger than those in summer and are closer

416   to the peak sizes of the PNSD (Figs. S9 b1-b3). The results suggest that PNSD was the most important

417   factor influencing N_{CCN} in summer, while hygroscopicity became more important in winter.

3.4 Influence of spatial distribution of particle properties on N_{CCN}

419   This section focuses on how the spatial distribution of aerosol concentration and physicochemical

420   properties influences CCN concentrations under different air masses, particularly the variations occurring

421   with offshore distance. Typically, N_{CCN} in a specific region is predicted in models with a single

422   parameterized PNSD and it can be used to simulate the hygroscopic growth process (Yu and Luo, 2009).

423   Here, we further investigated the impact of spatial variations in PNSD and hygroscopicity on the

424   prediction of N_{CCN}. In summer, terrestrial air masses primarily pass through the Philippines and the

425   impact varies with longitude, while during winter, terrestrial air masses are predominantly from mainland

426   China, especially the PRD region, and the impact varies with latitude (Fig. S3). Hence, we investigate

427   the influence in summer based on longitude and in winter based on latitude. Figure 8 illustrates the

428   variations of the N_{CCN} and number concentration of different mode particles, κ at 0.4% SS ($\kappa_{ss:0.4\%}$), the

429   ratio of SOC to OC, mass concentration of BC, and NR-PM$_1$ component proportions with increasing

430   offshore distance.

431   In summer, the N_{CCN} showed a decreasing trend with increasing offshore distance under the

432   terrestrial air masses, while no significant variation was observed under the mixed air masses.

433   Furthermore, the number concentration of Aitken mode particles significantly decreased with increasing

434   offshore distance (from 118°E to 113°E), from nearly 10^4 to approximately 10^3 (Fig. 8a2). Similarly, the

435   number concentration of accumulation mode particles exhibited a decreasing trend. In contrast, no

436   significant variation was observed for the number concentration of nucleation mode particles, indicating

437   that those particles were less affected by the terrestrial air masses. The $\kappa_{ss:0.4\%}$ values exhibit an increasing

438   trend with increasing offshore distance under the terrestrial air masses, while no obvious trend was shown

439   under the mixed air masses (Figs. 8 a3 and b3). The above observations can be attributed to three possible

440   reasons: (1) A higher degree of oxidation for the organic component enhances the hygroscopicity of the

441   particles, as evidenced by the increasing trend of the SOC/OC ratio with increasing offshore distance

442   (0.4 to 0.7 from 118°E to 113°E, Fig. 8a4). (2) A significant decrease in the BC concentration (0.5 to 0.3

443   μg m^{-3} from 118°E to 113°E, Fig. 8a5) may contribute to an increase of the aerosol hygroscopicity. (3)

444   An increase of sulfate fraction with increasing offshore distance may lead to a decrease of organic

445   compounds (Fig. 8a6). The increase of hygroscopicity with increasing offshore distance lowered the D_{50}

446   values and led to the increase of N_{CCN}, while the decrease of N_{CCN} was primarily attributed to the change

447   of the particle concentrations in Aitken mode and accumulation mode.

448   In winter, the increasing trend with increasing offshore distance is similar to that in summer under

449   the terrestrial air masses, while no significant N_{CCN} variation was observed under the mixed air masses

450   (Figs. 8c1-c2 and d1-d2). Meanwhile, the aerosol hygroscopicity exhibits a slight increasing trend with

451   offshore distance (a κ value of 0.19 to 0.21 from 22°N to 19°N). The increase of aerosol hygroscopicity

452   with increasing offshore distance can be explained by two possible reasons: (1) The nitrate mass fraction

453   decreased significantly and organics became the predominant component with increasing offshore

454   distance. Hence, the increased oxidation degree of the organic component was an important factor for

455   the enhancement of aerosol hygroscopicity which is supported by the increase of the SOC/OC ratio (Fig.

456   8c4). (2) The decrease of the BC concentrations with increasing offshore distance was more pronounced

457   during winter (2.1 to 0.6 μg m^{-3}) than during summer (0.5 to 0.3 μg m^{-3}), indicating that the decrease of

458   the BC mass fraction is also a significant contributor to the overall increase of aerosol hygroscopicity.

459   Interestingly, the slight decrease of the average κ value further offshore (19°N to 20°N) was caused by

460   the impact of nearby ship emissions which was further supported by the decrease of SOC/OC ratio.

461   The impact of spatial distribution of D_{50} and PNSD on N_{CCN} was further investigated through

462   calculating N_{CCN} using D_{50} or PNSD (Eq. (3)) according to the scheme shown in Fig. S10. For example,

463   to investigate the impact of PNSD on N_{CCN}, the PNSD which are closest and furthest from the shore and

464   the observed D_{50} were used to calculate N_{CCN}. Here, we define ΔN_{CCN} as the ratio of the difference

465   between calculated N_{CCN} and observed N_{CCN}, divided by the observed N_{CCN} (Eq. (7)).   However, under

466   the mixed air masses, as mentioned above, since no obvious variations of PNSD and aerosol

467   hygroscopicity with offshore distance were observed (Figs. 8 b2-b3 and d2-d3), the impact of spatial

468   distribution of D_{50} and PNSD on N_{CCN} was less significant (Fig. S11). Hence, below we only discuss the

469   scenarios under the terrestrial air masses.

470   Figure 9 shows ΔN_{CCN} at different offshore distances under the terrestrial air masses. In summer, a

471   monotonic trend of ΔN_{CCN} was observed with a value of up to approximately 70% at 0.2 SS under the

472   effect of hygroscopicity. In winter, the impact of hygroscopicity was less pronounced than that in summer,

473   reaching only up to around 10% (Figs. 9 c1 and c2). This indicates that a single hygroscopicity parameter

474   can represent the overall hygroscopicity in SCS under the influence of terrestrial air masses in winter,

475   whereas during summer, this approach may not be sufficient. The ΔN_{CCN} nearshore caused by the impact

476   of PNSD can exceed 300% and can reach up to 80% furthest offshore during summer, indicating that the

477   spatial distribution of PNSD has a greater impact on N_{CCN} than hygroscopicity. Similarly, although the

478   impact was less significant during winter, the ΔN_{CCN} nearshore can still reach 80% (Fig. 9 d1 and d2).

479   Hence, in summary, using a single PNSD for the representation of PNSD in the region of SCS can lead

480   to significant uncertainties in the prediction of N_{CCN} under the terrestrial air masses during both summer

481   and winter.

482   **4. Conclusion**

483   In this study, we investigated the seasonal variations of CCN activity in the SCS and explored the

484   impact of anthropogenic emissions, based on shipborne observations conducted during the summer (May

485   5–June 9) and winter (December 19–29) of 2021. CCN activity, chemical composition, and particle

486   number size distribution (PNSD) over the SCS were measured using several onboard instruments

487   including a ToF-ACSM, a CCNc, an SMPS, an OC/EC analyzer, an AE33, and several monitors for trace

488   gases (i.e., SO_2, NOx, CO, and O_3). Our results show that the particle number concentration (N_{CN}) and

489   CCN number concentration (N_{CCN}) during summer were higher than those during winter, while the NR-

490   PM_1 mass concentration was lower in summer, which can be attributed to the predominance of the Aitken

491   mode particles in summer than a significant higher concentration of the accumulation mode particles

492   during winter. Additionally, the aerosol hygroscopicity was found to be higher in summer than in winter,

493   likely due to the enhanced terrestrial air masses and the increased proportions of black carbon and

494   decreased sulfate concentrations from DMS oxidation.

495   Based on backward trajectories, both campaigns could be divided into two periods: period affected

496   by terrestrial air masses and period affected by both terrestrial and marine (mixed) air masses. In summer,

497   the terrestrial air masses originate primarily from the Philippines, whereas in winter, they predominantly

498   originate from the PRD region of China. The hygroscopicity under terrestrial air masses was lower than

499   that under mixed air masses during both seasons, and it was similar to that observed in the PRD region

500   during winter. The PNSD distribution exhibited a unimodal pattern under the terrestrial air masses,

501   whereas a bimodal pattern was observed under the mixed air masses. During winter, the N_{CCN} values

502   were higher under terrestrial air masses in contrast to higher AR values under mixed air masses. During

503   summer, the AR ratio was primarily influenced by PNSD, whereas during winter, it was more strongly

504   influenced by aerosol hygroscopicity than by PNSD.

505   Our study demonstrated significant variations in the aerosol concentration and physicochemical

506   properties with increasing offshore distances under terrestrial air masses during both summer and winter.

507   The decreasing trends observed in various gas concentrations (NO_X and CO) with increasing offshore

508   distances suggest a diminishing influence of anthropogenic emissions. The aerosol hygroscopicity

509   increased with increasing offshore distances, primarily due to the decrease of the organic fraction, the

510   oxidation degree of the organic component, the decreased proportions of black carbon, and the increased

511   sulfate ratio. We found that the predicted CCN concentrations nearshore based on a single PNSD under

512   terrestrial air masses could lead to significant error (15%–360%). In contrast, using a representative

513   hygroscopicity parameter value nearshore can reduce the error in predicting N_{CCN} (5%–10%). Hence, the

514   PNSD had a greater impact on N_{CCN} prediction than hygroscopicity. Our study highlights the significant

515   differences of CCN activity during summer and winter in the SCS and significant influence of

516   anthropogenic emissions on the CCN activity. Future studies should include observations during spring

517   and autumn to explore the impact of mixing state on aerosol hygroscopicity for a more comprehensive

518   understanding of CCN activity in this region.

519

520   *Data availability.* Data from the measurements are available at

521   https://doi.org/10.6084/m9.figshare.25472545 (Ou et al., 2024).

522

523   *Supplement.* The supplement related to this article is available online at xxx.

524

525   *Author contributions.* **HO, MC, and JZ** designed the research. **YZ, XN, BL, and CS** performed the

526   measurements. **HO, MC, QS, and SM** analyzed the data. **SZ and HW** provided useful comment on the

527   paper. **HO, MC,** and **JZ** wrote the paper with contributions from all co-authors.

528

529   *Competing interests.* The authors declare that they have no conflict of interest.

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776

777 FIGURE CAPTION

778 Figure 1. The cruies of two shipborne observation.

779 Figure 2. Timeseries of (a) particle number size distribution, (b) mass concentration of NR-PM1, and (c)

780 its fraction, (d) mass concentration of organic carbon and elemental carbon, (e) number concentration of

781 total particle and cloud condensation nuclei under the supersaturation of 0.1%, 0.2%, 0.4%, and 0.7%,

782 and (f) aerosol hygroscopicity. The number 1 means timeseries in summer and number 2 means it in

783 winter.

784 Figure 3. Particle number size distribution under effect of terrestrial air masses in summer (a) and winter

785 (b); Particle number size distribution under effect of mixed air masses in summer (c) and winter (d); The

786 red markers represent the activation diameters and hygroscopicity parameters corresponding to 0.1%,

787 0.2%, 0.4%, and 0.7% supersaturations in this study (without 0.1% in summer). The green markers

788 represent the hygroscopicity parameters reported in Atwood et al. (2017) for the southern South China

789 Sea during summer. The gray markers represent the hygroscopicity parameters documented in Cai et al.

790 (2018) for the Pearl River Delta region during winter.

791 Figure 4. The mass concentration of NR-PM1 , primary organic carbon, secondary carbon, elemental

792 carbon, and black carbon and their fraction under effect of different air masses in summer and winter.

793 Figure 5. Scatter plot of κ under the supersaturation of 0.2% and organic mass fraction with linear

794 regression.

795 Figure 6. Activation ratio at supersaturation of 0.1%, 0.2%, 0.4%, and 0.7% under effect of terrestrial

796 and mixed air masses in summer and winter. The box extends from the first quartile (Q1) to the third

797 quartile (Q3) of the data, with a line at the median. The box extends from Q1 to Q3 of the data, with a

798 line at the median. The whiskers extend from the box by 1.5 times of the inter-quartile range (IQR). Flier

799 points are those passing the end of the whiskers.

800 Figure 7. Differences in activation fraction calculated from different particle size distributions and

801 activation diameters.

802 Figure 8. Variations of κSS:0.4%, trace gases (NOx and CO), OC/EC, BC, SOC/OC, mass fraction of

803 NR-PM1 chemical composition, as well as number concentrations of nucleation mode, Aitken mode, and

804 accumulation mode with offshore distance under effect of terrestrial and mixed air masses in summer

805 and winter.

806   Figure 9. The difference between the calculated cloud condensation nuclei number concentration (NCCN)

807   using the D50 from the farthest and nearest offshore distances, along with the measured particle number

808   size distribution (PNSD), and the measured NCCN under the effect of terrestrial air masses in summer

809   (a1 and a2) and winter (c1 and c2); The difference between the calculated NCCN using the PNSD from

810   the farthest and nearest offshore distances, along with the measured D50, and the measured NCCN under

811   the effect of terrestrial air masses in summer (b1 and b2) and winter (d1 and d2). ΔNCCN refers to the

812   difference between calculated NCCN and observed NCCN, divided by the observed NCCN.

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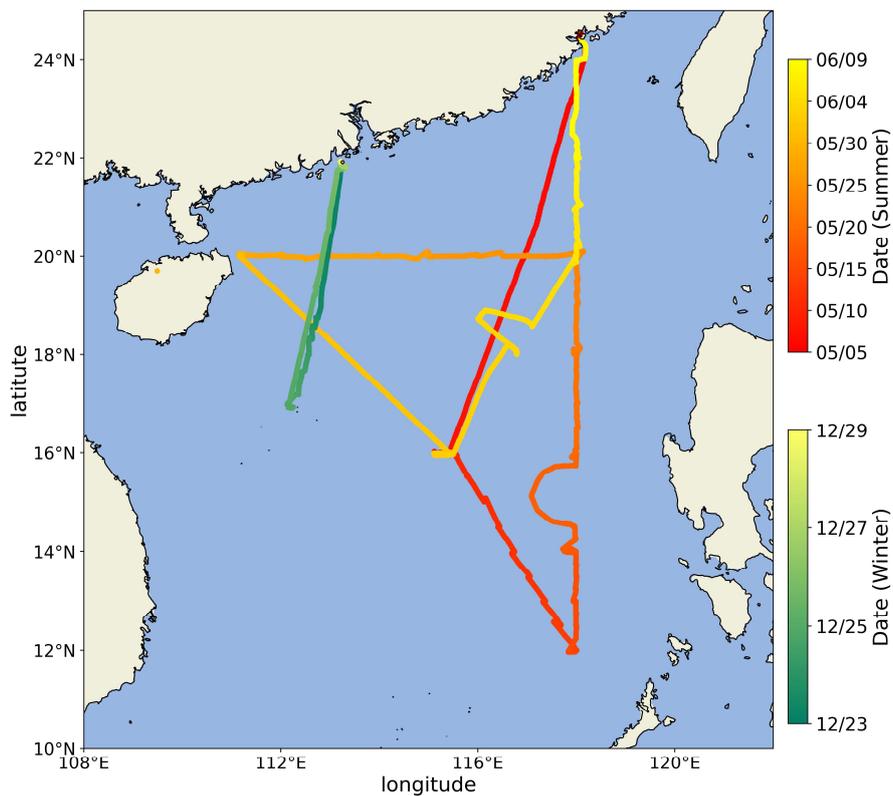
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816   Fig. 1
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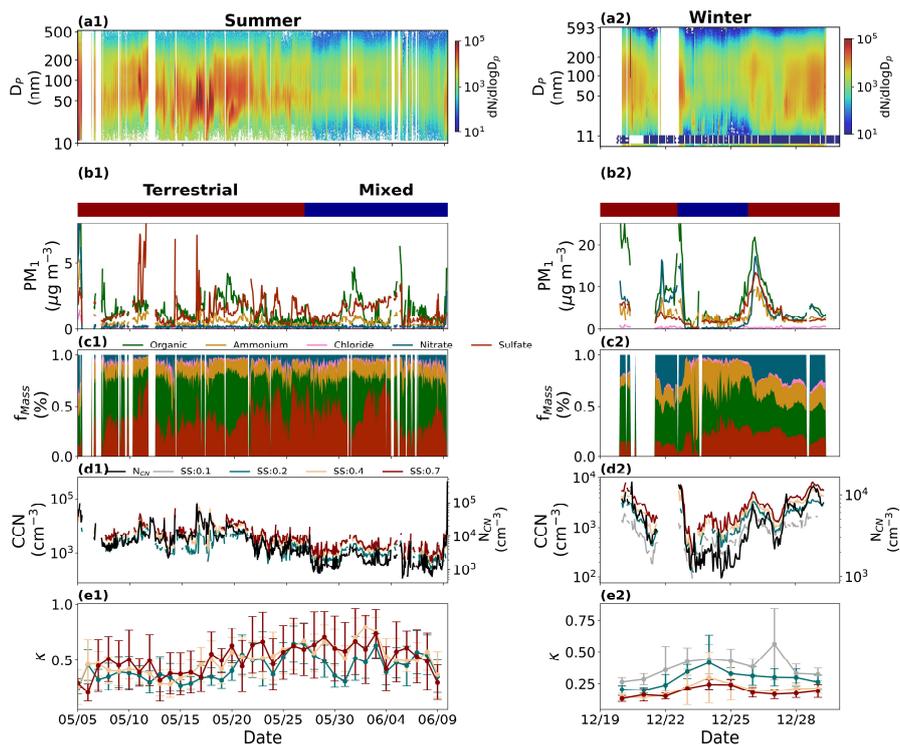
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819   Fig. 2

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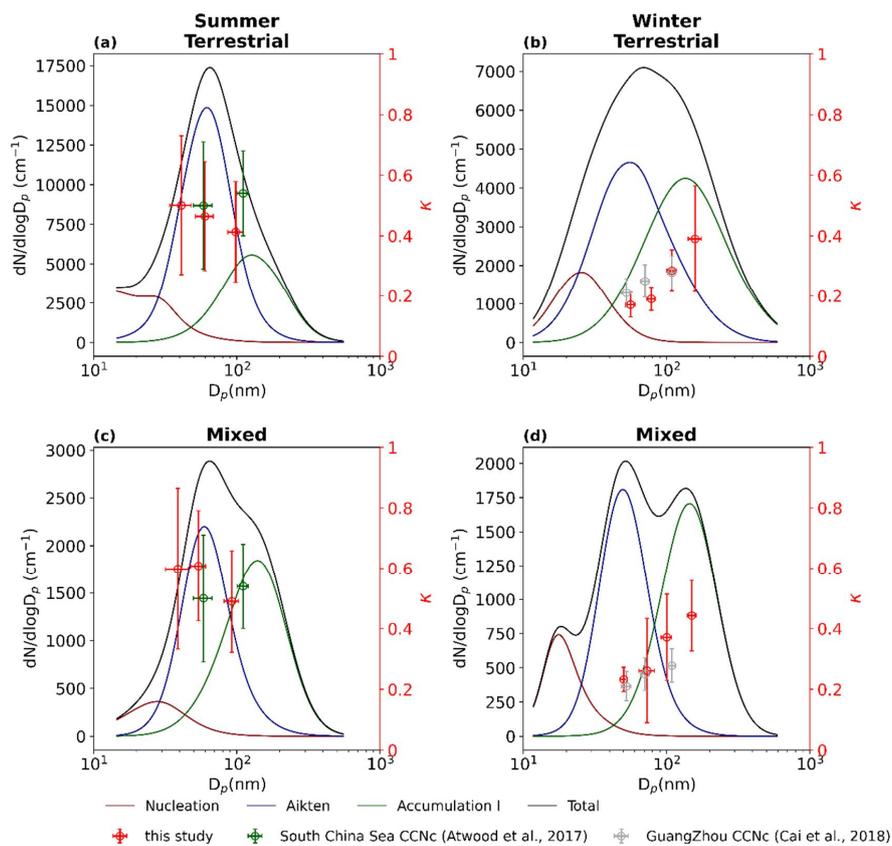
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822   Fig. 3

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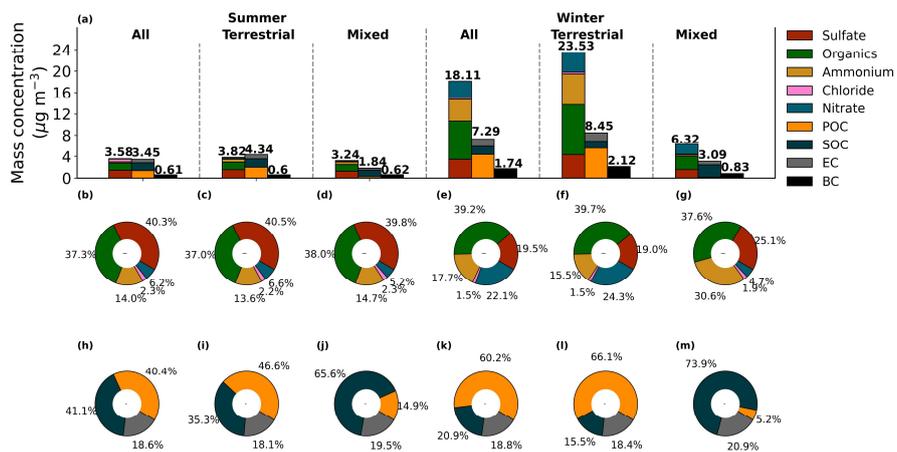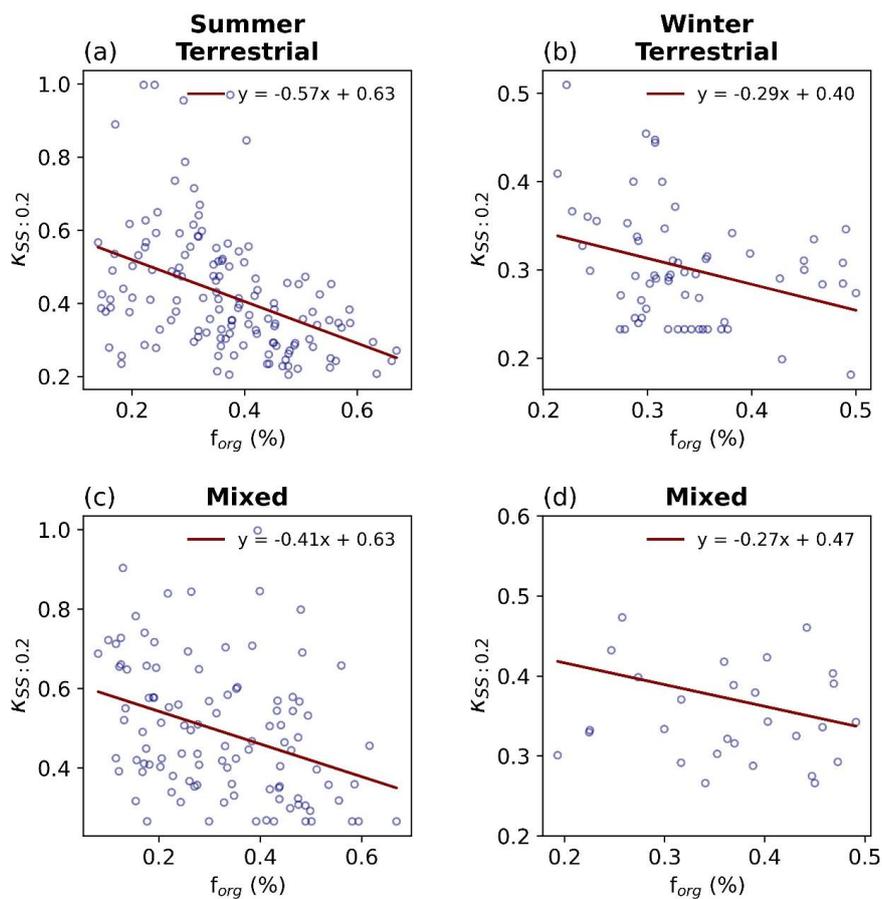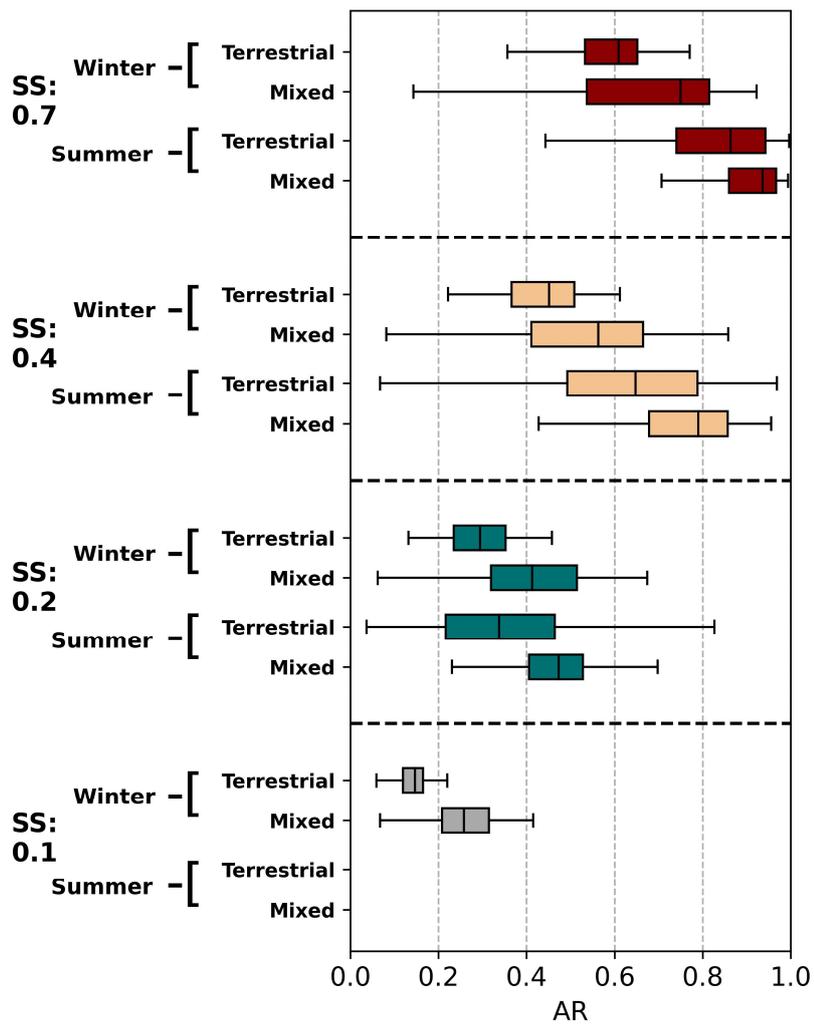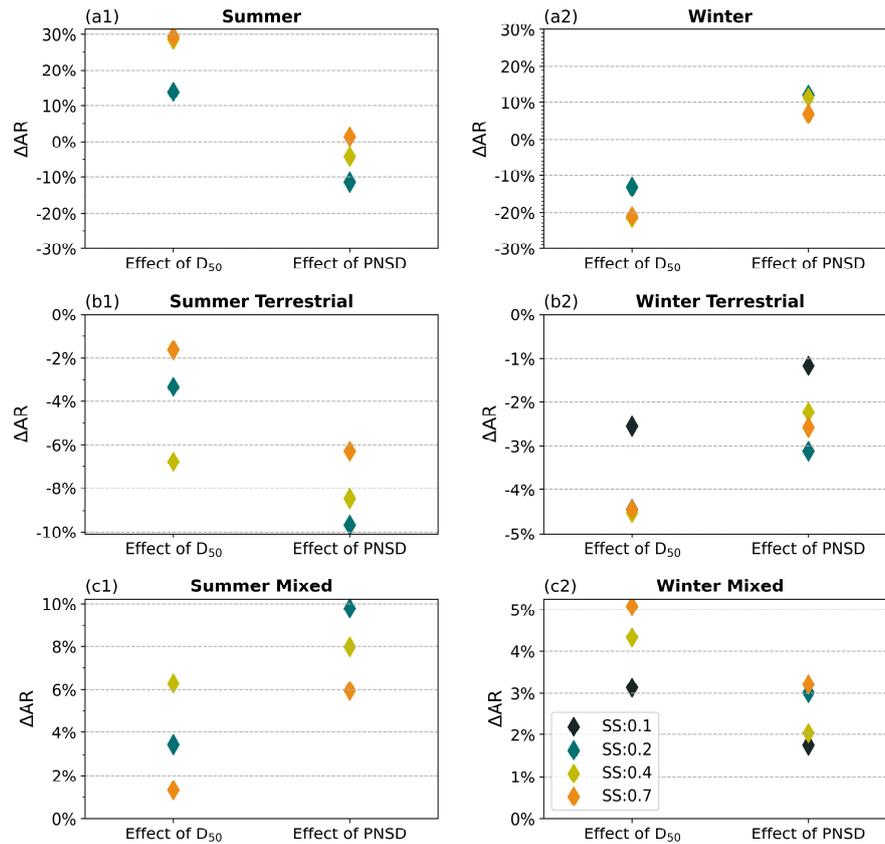
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835   Fig. 7

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