

## Supporting Information for

# **Marine boundary layer aerosol in Eastern North Atlantic: seasonal variations and key controlling processes**

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## S1. Data filtering methods

The Graciosa Airport is located nearby the ENA site, with the east-west extending aircraft runway 116 m north of the site (Wood et al., 2015). Pollution episodes associated with aircraft and road vehicles at Graciosa Airport are identified and screened out based on temporal variation of CN, following a similar method as described in Zheng et al. (2016). Briefly, the change rate of total aerosol number concentration (CN) is calculated first. Subsequently, all time points with change rates larger than  $60 \text{ cm}^{-3} \text{ s}^{-1}$ , which is roughly the 95<sup>th</sup> percentile of all CN change rates, are identified as episode candidates. Starting from these candidates, the program will look forward / backward until all pollution periods is found (Zheng et al., 2016). This method worked satisfactorily as a conservative estimate that can remove the most obvious local pollution periods, which typically constitutes 0 %~20 % of the data within the averaging interval of 1-h.

## 10 S2. Optical data corrections

$B_{\text{abs}}$  can be derived by Bond's correction of:  $B_{\text{abs}} = B_{\text{PSAP}} - 0.0164 B_{\text{NEPH}}$  (Bond et al., 1999). For this purpose, conversion of  $B_{\text{NEPH}}$  from Nephelometer measurement wavelengths (450, 550, and 700 nm) to  $3\lambda$ -PSAP measurement wavelengths (464, 529, and 648 nm) is needed, through interpolation based on the Scattering Ångström Exponent (SAE) (Costabile et al., 2013). For example,  $B_{\text{NEPH}}$  at 529 nm are derived by:

$$15 \quad SAE_{\text{NEPH}}(550) = -\frac{\log(B_{\text{NEPH}}(700)/B_{\text{NEPH}}(450))}{\log(700/450)}, \text{ and } B_{\text{NEPH}}(529) = B_{\text{NEPH}}(550) \left[ \left( \frac{529}{550} \right)^{-SAE_{\text{NEPH}}(550)} \right]$$

where the numbers in parenthesis denoted the wavelengths in nm. Similarly,  $B_{\text{NEPH}}$  at 464 and 648 nm were derived from those measured at from 450 and 700 nm, using SAE derived by 450 nm /550 nm and 550 nm /700 nm wavelength pairs. Note that there's another correction method proposed by Virkkula et al. (Bond et al., 1999; Virkkula et al., 2005; Virkkula, 2010). Based on our data, that correction will result in a 6 % lower value in  $B_{\text{abs}}$  at 529 nm, but showing the same trend with Bond' correction ( $R^2=0.99$ ). As here we only discussed about relative trends in  $B_{\text{abs}}$  here, the detailed correction method would not influence our conclusions.

For  $B_{\text{sca}}$ , truncation and angular illumination corrections using SAE are applied, following the method proposed by Anderson and Ogren (1998). Briefly, that is  $B_{\text{sca}} = (a+b \cdot \text{SAE}) \cdot B_{\text{NEPH}}$ , where the wavelength-dependent correction coefficient  $a(\lambda)$  and  $b(\lambda)$  are taken from Müller et al. (2011), with the “sub- $\mu\text{m}$ ” and “no cut” coefficients applied for  $\text{PM}_{10}$  and  $\text{PM}_{10}$  signals, respectively. After this correction,  $B_{\text{sca}}$  is further converted to PSAP-corresponding wavelengths through the SAE interpolation method as described above. Again, take data at 529 nm for example, it was derived by:

$$25 \quad SAE(550) = -\frac{\log(B_{\text{sca}}(700)/B_{\text{sca}}(450))}{\log(700/450)}, \text{ and } B_{\text{sca}}(529) = B_{\text{sca}}(550) \left[ \left( \frac{529}{550} \right)^{-SAE(550)} \right]$$

## S3. Estimation of $k_{\text{CCN}}$ and $k_{\text{CN}}$

30 For  $\text{CCN}(0.1 \%)_{\text{SSA}}$ , the influence of secondary processing is expected to be negligible. SSA over 100 nm would all have been activated under typical cloud  $ss$ , thus the ratio of  $\text{CCN}(0.1 \%)_{\text{SSA}}$  to  $N_{400}$  would be conserved during coalescence scavenging. Non-cloud processing would reduce  $N_{\text{Ac}}$  through coagulation loss, while this influence is expected to be small (section 6.2, Table 3). Thus we have  $k_{\text{CCN}} = 1$ .

In terms of  $CN$ , the situation could be more complicated since it includes both  $CCN$  and interstitial aerosols. Coalescence scavenging would reduce  $CCN(0.1\%)_{SSA}$  without impacting the interstitial aerosols, thus elevating actual  $CN/CCN(0.1\%)_{SSA}$  ratio and underestimating the  $CN_{SSA}$ . To the contrary, coagulation loss and interstitial scavenging could be a major non- $CCN$  loss term (section 6.2, Table 3), which will result in a lower actual  $CN/CCN(0.1\%)_{SSA}$  ratio than theoretically predicted, and thus an overestimation in estimated  $CN_{SSA}$ . The overall influence depends on the relative strength of these two mechanisms. Assuming that both the  $CCN(0.1\%)_{SSA}$  and the interstitial aerosols from SSA,  $N_{INT,SSA}$ , are in steady state (namely  $\partial_t CCN(0.1\%)_{SSA} = 0$  and  $\partial_t N_{INT,SSA} = 0$ ), then we have:

$$\int_{\ln 100}^{\ln 1000} f_{SSA}(\ln D_p) d \ln D_p = (E_{CCN|PRCP} + E_{CCN|COND} + E_{CCN|FT,dilute}) CCN(0.1\%)_{SSA}$$

$$\int_{\ln 10}^{\ln 100} f_{SSA}(\ln D_p) d \ln D_p = (E_{CN-CCN|INT} + E_{CN-CCN|COAG} + E_{CN-CCN|COND} + E_{FT,dilute}) N_{INT,SSA}$$

$$= (E_{CN-CCN|INT} + E_{CN-CCN|COAG} + E_{CN-CCN|COND} + E_{FT,dilute})(CN_{SSA} - CCN(0.1\%)_{SSA})$$

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where  $E_{FT,dilute}$  is the dilution efficiency of FT air being  $-\omega_e/H_{MBL}$  (section 5). Combing these two equations, we have:

$$\frac{CN_{SSA} - CCN(0.1\%)_{SSA}}{CCN(0.1\%)_{SSA}} = \frac{E_{CCN|COALES} + E_{CCN|COND} + E_{FT,dilute}}{E_{CN-CCN|INT} + E_{CN-CCN|COAG} + E_{CN-CCN|COND} + E_{FT,dilute}} \frac{\int_{\ln 10}^{\ln 100} f_{SSA}(\ln D_p) d \ln D_p}{\int_{\ln 100}^{\ln 1000} f_{SSA}(\ln D_p) d \ln D_p}$$

Thus  $k_{INT}$  could be estimated by:

$$k_{INT} = \frac{E_{CCN|COALES} + E_{CCN|COND} + E_{FT,dilute}}{E_{CN-CCN|INT} + E_{CN-CCN|COAG} + E_{CN-CCN|COND} + E_{FT,dilute}} < \frac{E_{CCN|COALES}}{E_{CN-CCN|INT} + E_{CN-CCN|COAG}}$$

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The estimated coalescence scavenging efficiency (17% ~ 40%  $CCN \text{ day}^{-1}$ ) is 1.7-4.4 times stronger than the overall efficiency of coagulation loss and interstitial scavenging (Table 3). Correspondingly, the upper limit of  $k_{INT}$  is expected to be between 1.7 and 4.4 (Table 4).

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**Fig. S1 to S5**

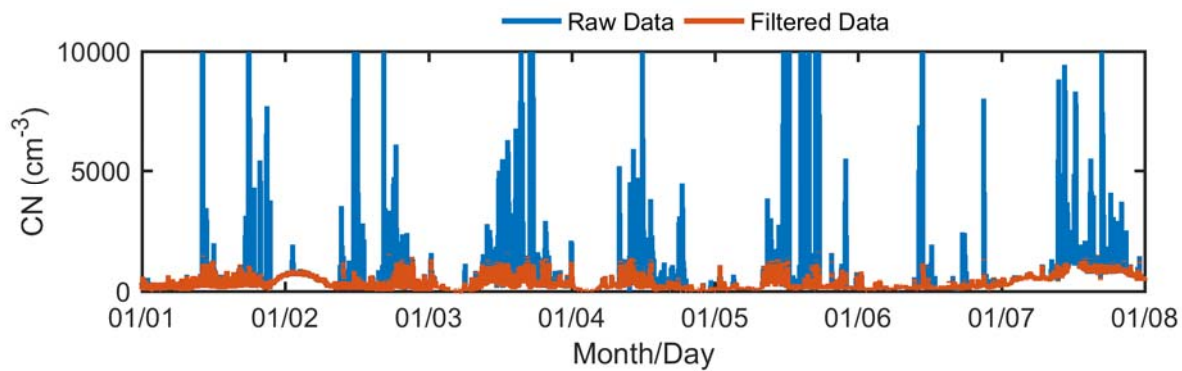
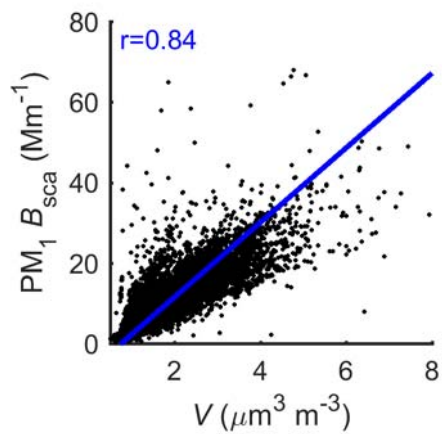
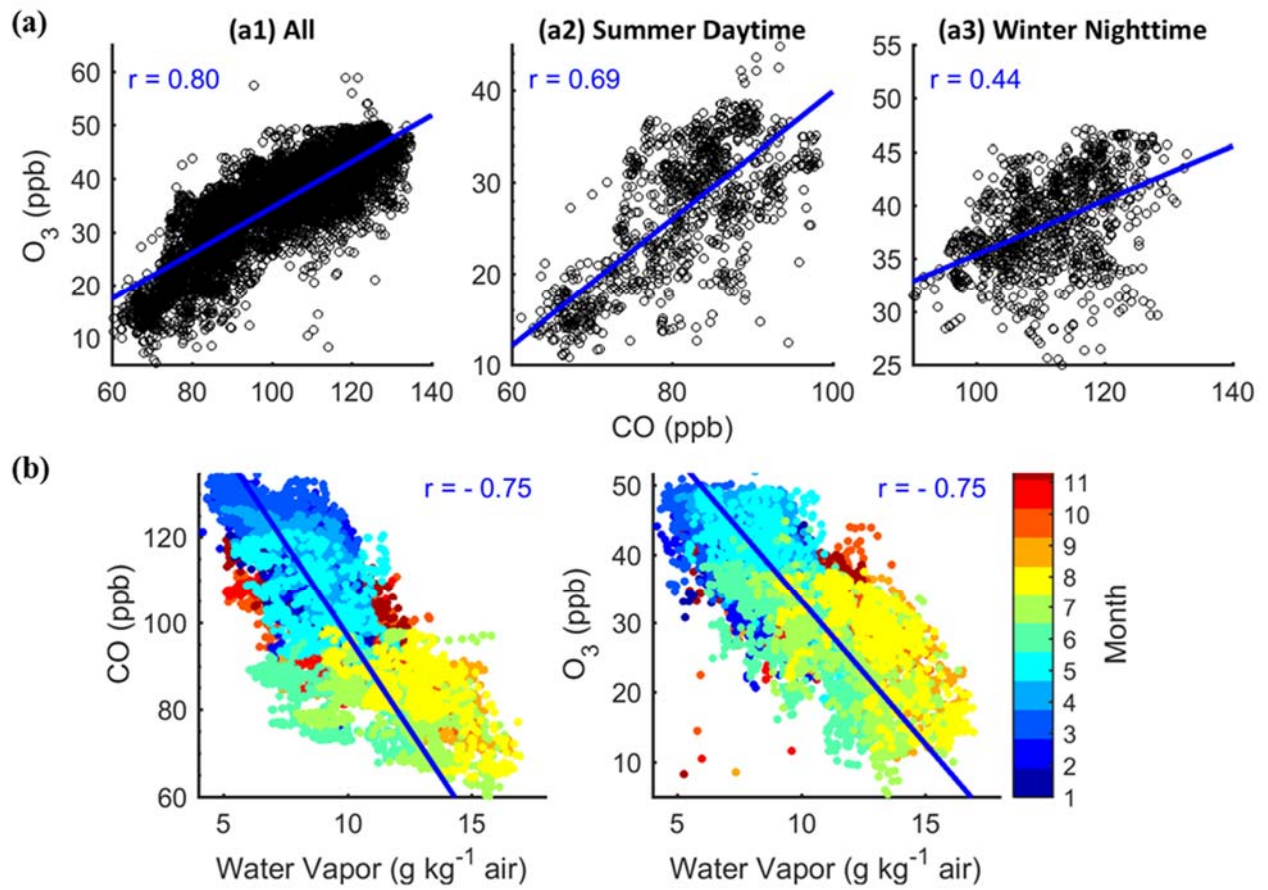


Figure S1. Raw and filtered CN during the first week in Jan. 2017, as an example of the outcome of the pollution episode filtering method used here.



5 **Figure S2. Correlation of  $PM_1 B_{sca}$  and total volume concentration derived from UHSAS measurements from 2015 to 2017.** The value of  $r$  given referred to the Pearson correlation coefficient, while the regression line based on York et al. (2004) is also shown for reference.



**Figure S3. Evidences of FT domination on ENA CO and O<sub>3</sub>.** (a) O<sub>3</sub>-CO correlations during (a1) all periods, (a2) summer daytime and (a3) winter nighttime, where daytime indicated 8:00 to 20:00 LT. (b) Correlation of CO and O<sub>3</sub> with water vapor. Data during the identified dust and biomass burning episodes (section 3.3) is excluded here. The value of  $r$  given referred to the Pearson correlation coefficient, while the regression line based on York et al. (2004) is also shown for reference.

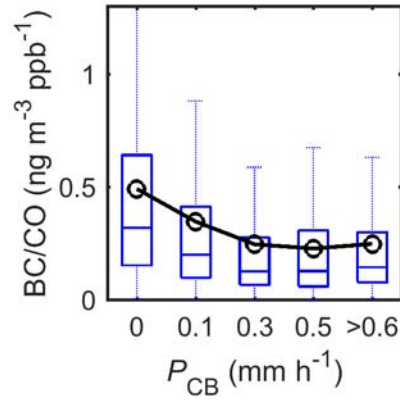


Figure S4. Dependence of BC to CO ratio with precipitation rate at cloud base ( $P_{CB}$ ).

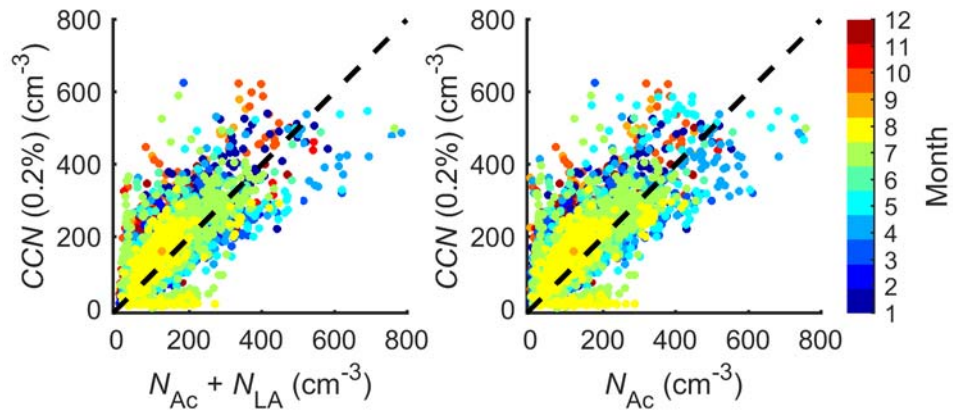


Figure S5. Comparison of observed  $CCN$  concentrations with relevant modal number concentrations. The 1:1 line is also shown for reference.

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