

1      Reviewer-2

2      We appreciate your review and critique of the manuscript. Thank you.

3      Please note: Line numbers stated here are from the original manuscript.

4      **General Comments:**

5      The paper describes aerosol data obtained in a 3-month observational study at a coastal site in  
6      Chile. Aerosol observations in this part of the world are rare so the data should be of interest to  
7      the community. Hence, I support publication of this work.

8      I offer some comments below that the authors can consider in revision. In general, I think some of  
9      the discussion of standard instruments and approaches could be stream-lined or moved to the  
10     Appendix.

11     The analyses and findings are fairly straightforward. Implications could be strengthened by  
12     additional comparison to observations that are clearly “clean marine”.

13     This was addressed by revising the final sentences of Section 4.1:

14     “These averages are also statistically different ( $p < 0.01$ ), and again, the Arauco average is larger  
15     than that at THD. Based on averages presented in this section, and information provided in Table  
16     2, two summary statements are warranted: 1) During wintertime, the THD classifies as a  
17     moderately-polluted marine site, and the Arauco Site classifies between moderately-polluted  
18     marine and heavily-polluted marine. 2) These sites are not representative of conditions well  
19     removed from anthropogenic influence.”

20 **Specific Comments:**

21 Line 52: it's not clear how these aerosol indirect effects differ, as described here; please clarify. The  
22 Albrecht reference may refer to hypothesized increasing cloud lifetime and cloud cover due to  
23 increased aerosol?

24 We revised this:

25 "Consequently, upward reflection of solar radiation by liquid-only clouds (Twomey 1974), and upward  
26 reflection attributable to cloud fractional coverage (Albrecht 1989), increase with increased aerosol  
27 abundance."

28 Line 61: perhaps the VOCALS study should be cited as a contribution to Southern Hemisphere field  
29 work exploring aerosol-cloud interactions.

30 The references we picked contrast Southern and Northern Hemisphere aerosol and cloud  
31 properties. We are not aware of a VOCALS-related publication that does that. There is reference  
32 to VOCALS in Sections 4.4 (Snider et al. 2017; manuscript bibliography).

33 Line 70: I think you mean that the presence of SSA is associated with the presence of giant CCN that  
34 promote drizzle production.

35 We do not use the modifier “giant” when referring to a subclass of the aerosol. We did change  
36 the text to stress that most of the CCN are smaller than the class of SSA particles ( $D > 0.5 \mu\text{m}$ ) that we  
37 focus on. Here is how the paragraph is rewritten:

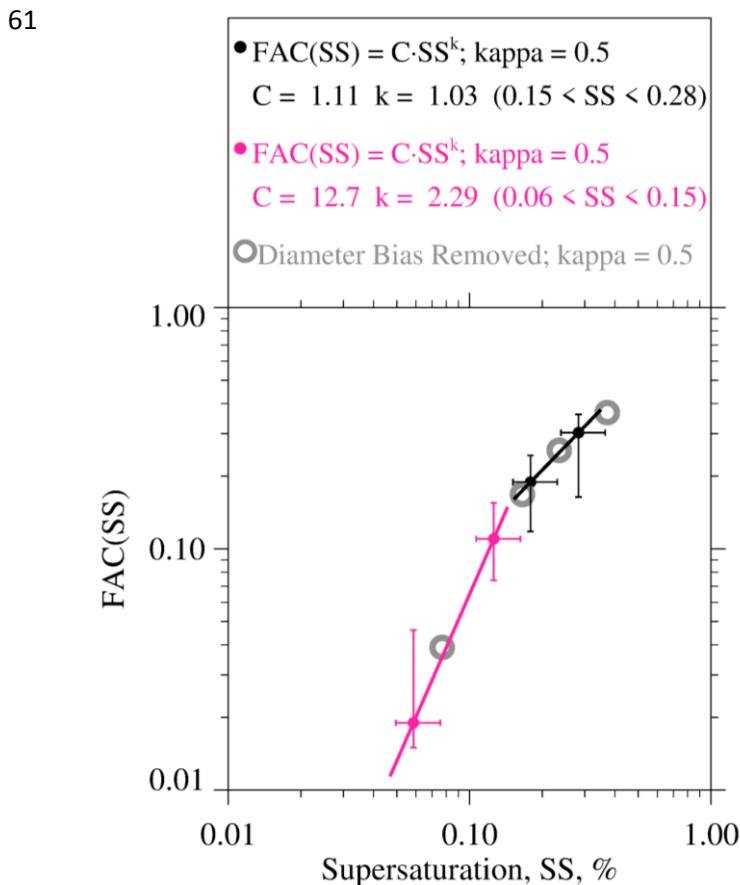
38 “We emphasize the following topics: 1) The parameterized relationship between sea salt aerosol (SSA)  
39 particles (diameter  $> 0.5 \mu\text{m}$ ) and wind speed; 2) The role as cloud condensation nuclei (CCN) of  
40 particles that are both smaller and more numerous than the above-mentioned SSA; 3) The  
41 parameterized relationship describing CCN activation spectra (Rogers and Yau, 1989; chapter 6), and 4)  
42 the potential application of the SSA and CCN parameterizations in numerical modelling of wintertime  
43 Southern Hemispheric clouds and precipitation. Motivating our investigation are modeling studies  
44 (Feingold et al. 1999), and analyses of field measurements (Gerber and Frick 2012), indicating that the  
45 reduction of rainfall due to increased CCN can be negated by SSA particles.”

46

47

48 Line 132: the particle size overestimate due to not being fully dried is discussed and a ballpark %  
49 given. However, it seems the data were not corrected for this. The CCN estimate will therefore be  
50 affected since critical supersaturation is very sensitive to size. Why wasn't this factored in? (Since a  
51 kappa is assumed, the data could be corrected for water content if RH is known.) Could this  
52 overestimate be used to add uncertainties into the parameterization?

53 Our analysis of the 20% particle-size overestimate is in the figure below. The pink and black  
54 data points, and their uncertainties and fit lines, are replicated from Fig. 8 (manuscript). In  
55 addition, gray circles are plotted at critical SS values corresponding to diameters 20% smaller  
56 (kappa = 0.5 is assumed). This demonstrates that a decreased lower-limit diameter, and the  
57 resultant increased fractional aerosol concentration (FAC), propagate to an insignificant departure  
58 of the perturbed data points (gray circles) from the FAC relationship in Fig. 8. Certainly, the  
59 perturbed points remain within the uncertainties described in Section 4.4. This explains why we  
60 did not factor in a 20% particle-size overestimate into our analysis of uncertainty in Fig. 8.



62 Line 136: what height was the inlet? (this is specified only later on line 175, as 2 m) It seems to me  
63 that the aerosol inlet was much lower than is typically done for aerosol sampling campaigns (e.g.,  
64 THD has an aerosol inlet at 10m). What is the impact on the data?

65 Our main concern was keeping rain out of the Arauco inlet. We accomplished this by  
66 sampling below an eave on the west side of the residence at the Arauco Site (L136). In the  
67 revision, we modified the sentence starting on L174:

68 "An important distinction between the sampling at THD and Arauco is the above ground level  
69 (a.g.l.) height of the aerosol inlets. This is 10 and 2 m a.g.l. at THD and Arauco, respectively. We  
70 cannot state with any certainty if the lower-height sampling at Arauco made those measurements  
71 unrepresentative."

72

73 Line 141: there is a lot of detail about the CPC principle of operation, yet this is a very commonly  
74 applied and simple instrument. In general I think the descriptions of instrumentation could be  
75 much briefer.

76 The two paragraphs were shortened and merged. However, relevant connections to the  
77 CPC at THD, maximum detectable concentration, and data recording were retained.

78 Here is the revised text:

79 "The CPC counts particles larger than  $D = 0.010 \mu\text{m}$  (Table 1)<sup>1</sup> up to a maximum concentration of  
80  $10,000 \text{ cm}^{-3}$ . The UHSAS measures scattering produced when aerosol particles are drawn through light  
81 emitted by a solid state laser ( $\lambda = 1.05 \mu\text{m}$ ). By reference to a calibration table (Cai et al. 2008; Cai et al.  
82 2013), the UHSAS microprocessor converts scattered light intensity to particle size and accumulates  
83 the derived sizes in a 99 channel histogram. Channel widths are logarithmically uniform ( $\Delta \log_{10} D =$   
84 0.013) over the instrument's full range ( $0.055 < D < 1.0 \mu\text{m}$ ). UHSAS concentrations were recorded  
85 every 10 seconds and CPC concentrations were recorded once per second (Table 1)."

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<sup>1</sup> The CPC minimum detectable diameters we report are in fact diameters that a CPC detects particles with efficiency = 50 %. The CPC detection efficiency is a steep function of particle diameter (Weidensholer et al. 1997).

86 Line 161: the presence of the paper mill immediately render this as a non-pristine site. Later,  
87 on lines 476, the prevalence of wood burning is mentioned. Even with onshore winds, complex  
88 coastal flows will likely result in influences from these aerosol sources. Probably it needs to be  
89 stated upfront that this site is not representative of a “clean marine” location even when data are  
90 segregated by sector.

91 This is stated, after relevant analysis, in two places in the original manuscript: 1) L279 to  
92 L282, and 2) L307 to L311. We feel this is sufficient. Also, please see our reply to your General  
93 Comment.

94 Line 182: there is no mention of topography in the description of the site and surrounding  
95 area. This seems critical to understanding how the site is affected by transport.

96 The topography is provided in Fig. 1. Also, we assert that further analysis of satellite  
97 retrievals are needed to address this outstanding issue. Please see Sect. 5 where we discuss  
98 satellite-based cloud droplet concentration retrievals in Bennartz (2007).

99 Line 191: Just a comment: in the end there are only a few days (five days?) of data with  
100 onshore flow + UHSAS data that can be used to characterize the “marine” sector.

101 As we state on L191 to L192, there are 20 onshore trajectories that overlap with the availability  
102 of UHSAS measurements. Table 3, which is discussed later in the manuscript, has the dates and  
103 times of the onshore trajectories. These occurred on seven different days in June, 2015.  
104 Please note that the arrival times are static: 00, 06, 12, and 18 UTC.

105

106 Line 231-233: I don't think these equations are needed in the text – perhaps in the  
107 supplement if you think they are necessary, but they are pretty standard.

108 Yes they are standard, however, our analysis and presentation relies on these  
109 moments (zeroth, second, and third), and our CCN parameterization relies on an integral  
110 similar to Eq. 2. We prefer to leave these definitions.

111 Line 265: the T-test is a fairly standard statistical test and doesn't need a lot of description.

112 Apparently, there are a few tests in the category of "t-test". We prefer this one, and document  
113 by citing Havlicek and Crain (1988).

114

115 Line 434: internal mixing is probably not a good assumption as claimed, since many  
116 observations have shown that organics content of marine aerosol increases with decreasing  
117 size. However, it is hard to justify another assumption here, and perhaps the best way to  
118 address is to discuss some prior observations and add estimates of uncertainty?

119        Given that our parameterizations are aimed at multi-dimensional models of aerosol  
120 and cloud and multi-dimensional models of aerosol, cloud, and precipitation, where the  
121 mixing state in the activation scheme is nearly always “internal”, we do not see merit in  
122 exploring this issue. Further, we note that aerosol dynamics calculations confirm this  
123 assumption provided coagulation (of aerosol particles) and condensation (of trace gas onto  
124 aerosol particles) has gone on for 24 hours (Fierce et al. 2017; their Figure 2). The action of  
125 coalescence scavenging (Wood et al. 2006), occurring within clouds, is ignored in the  
126 calculations of Fierce et al. (2017), and would further shorten the time needed for the  
127 internal mixing assumption to be valid. Please note, we cite Fierce et al. (2017) in this  
128 paragraph of the manuscript.

129        Fierce, L., N. Riemer, and T.C. Bond, Toward Reduced Representation of Mixing State  
130 for Simulating Aerosol Effects on Climate. Bull. Amer. Meteor. Soc., 98, 971–980,  
131 <https://doi.org/10.1175/BAMS-D-16-0028.1>, 2017

132        Wood, R. ( 2006), Rate of loss of cloud droplets by coalescence in warm clouds, J.  
133 Geophys. Res., 111, D21205, doi:10.1029/2006JD007553.

134

135 CCN parameterization: why aren't the size distributions used more directly, and why fit with  
136 the exponential relationship? The latter is clearly not physical despite its long history of use  
137 on the community, although for marine stratus that do not reach high supersaturations, it is  
138 reasonable within the expected supersaturation bounds.

139 Size distributions are used in a manner that is direct. This is explained in the revised  
140 Section 4.4. Our explanation is enhanced by addition of Eq. 5 (revision).

141 What we develop is a power-function relationship between a CCN activation spectrum  
142 and supersaturation:  $N(SS) = N_{CPC} \cdot FAC(SS) = N_{CPC} \cdot C \cdot SS^k$ . As is the case for all power functions  
143 relating cumulative CCN concentration ( $N(SS)$ ) and supersaturation ( $SS$ ), cloud droplet  
144 concentration can be calculated with the activation spectrum parameters ( $C$  and  $k$ ) and with  
145 measured (or assumed) updraft velocity (e.g., Johnson 1981). Thus, an analytical link between  
146 CCN, cloud updraft, and cloud microphysics is established. Caveats associated with this  
147 approach, and why such a calculation of droplet concentration can differ somewhat from a  
148 calculation based on a numerical parcel model, are discussed in Johnson (1981).

149 Johnson, D.B., 1981: Analytical Solutions for Cloud-Drop Concentration. J. Atmos. Sci.,  
150 38, 215–218, [https://doi.org/10.1175/1520-0469\(1981\)038<0215:ASFCDC>2.0.CO;2](https://doi.org/10.1175/1520-0469(1981)038<0215:ASFCDC>2.0.CO;2)

151

152 What about comparing with other published spectra for coastal aerosol?

153 As far as we can tell, no published CCN activation spectra are available for the Central Chilean  
154 Pacific coast (e.g., Schmale et al. 2018). Our group has published *summertime* measurements of CCN  
155 spectra (Snider et al. 2017; their Table 2). These were acquired over the subtropical Southeast Pacific,  
156 within the summertime marine boundary layer (Snider et al. 2017; Figure 1). A comparison is shown  
157 below. Since this is an open response, we have elected to show the comparison here, but not as an  
158 addition to the manuscript. First we compare our parameterized fractional aerosol concentration (*FAC*)  
159 function to the analysis in Andreae (2009), and then we compare CCN activation spectra.

160 Fig. a (see below) reproduces the parameterized *FAC* curve presented in the manuscript (Fig. 8).  
161 As we discussed in the manuscript, this was derived using size distribution and CPC measurements  
162 (please see Eq. 5 in the revised manuscript), and using the kappa–Köhler formula of Petters and  
163 Kreidenweis (2007, their Eq. (6)). The value  $\kappa = 0.5$  is assumed for the curve we show in Fig. a. A data  
164 point derived using values in Table 2 of Andreae (2009) is also presented. Different from our approach,  
165 the measurements Andreae (2009) analyzed are from a set of CCN( $SS=0.4\%$ ) and CPC measurements.  
166 Those measurements were acquired at a variety of locations. The locations are classified as Clean  
167 Marine, Clean Continental, Polluted Marine, and Polluted Continental (Andreae 2009). The averaged  
168  $N(SS=0.4\%) / N_{CPC}$  ratio for these conditions is 0.36 (Andreae 2009; their table 2). At the large  $SS$  end of  
169 our parameterization (Fig. a), we see reasonable agreement between with Andreae (2009).

170 Two activation spectra – derived as  $N_{CPC} \cdot FAC(SS) = N_{CPC} \cdot C \cdot SS^k$  (Section 4.4) - are shown in Fig.  
171 b (see below). These go with upper and lower quartile values of the  $N_{CPC}$  ensemble described in the  
172 Supplementary Material (manuscript). Also presented is the averaged CCN activation spectrum based  
173 on the 36 spectra from Table 2 of Snider et al. (2017).

174 At  $SS = 0.3\%$  there is consistency between the Southern Hemisphere (SH) averaged  
175 summertime spectrum (Snider et al. 2017) and SH wintertime spectrum, provided the latter is  
176 compared using the lower-quartile- $N_{CPC}$  value (see previous paragraph). However, these averaged  
177 spectra have different slopes and they therefore diverge at  $SS < 0.3\%$ . A smaller slope in the  
178 summertime setting could be due to a less prominent Aitken mode (summertime), compared to a  
179 more prominent Aiken mode (wintertime).

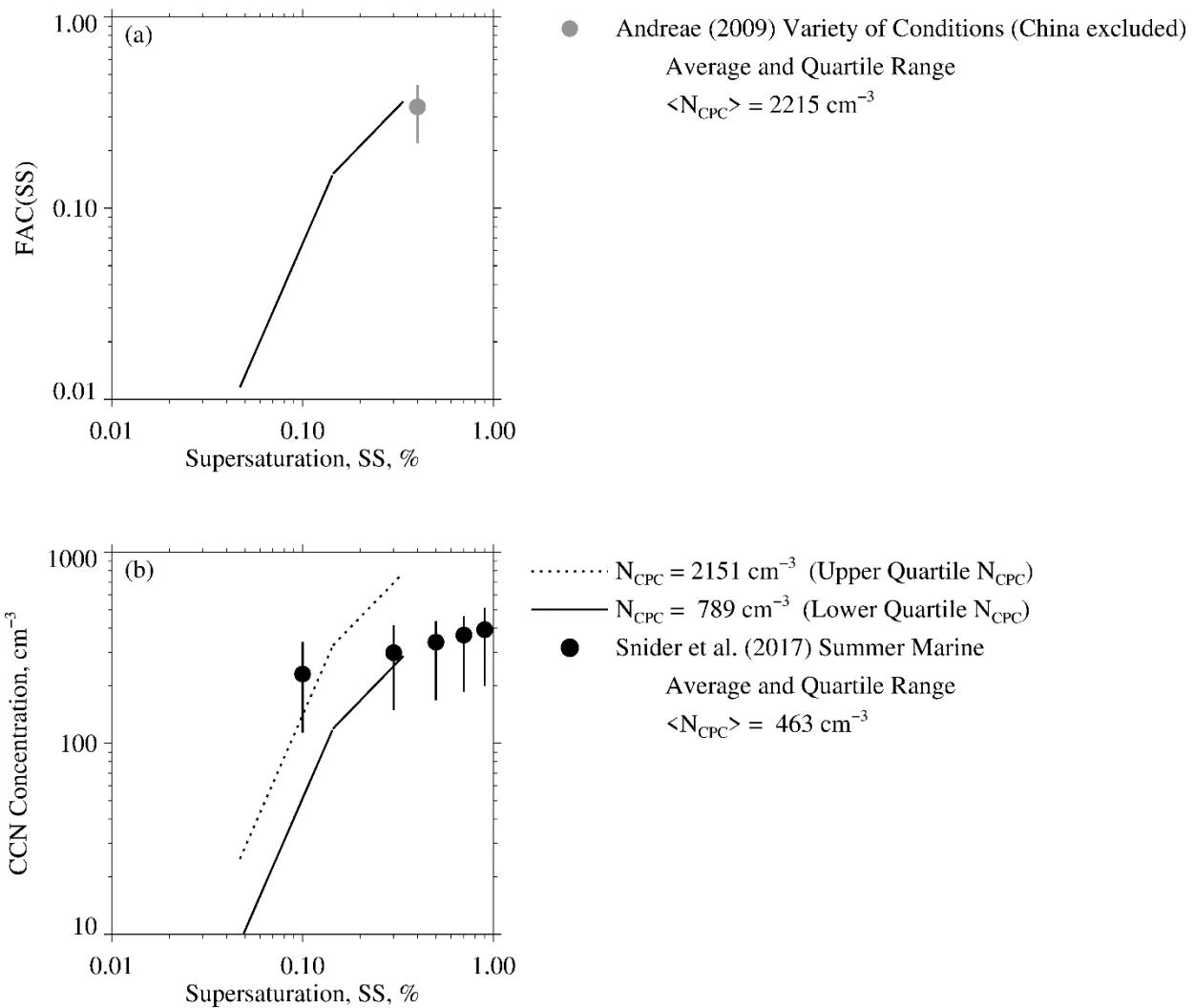
180            Although this comparison is limited, we do not see a significant discrepancy between the FAC  
181 parameterization we developed, and the approach of Andreae (2009) (Fig. a). Some discrepancy is  
182 apparent between the CCN activation spectra we derive, for relatively clean wintertime conditions,  
183 with  $N_{CPC} = 789 \text{ cm}^{-3}$ , and the averaged CCN spectrum in marine conditions over the Southeast Pacific,  
184 albeit during summer and at lower latitude. This discrepancy increases with decreasing SS. More  
185 comparison data is needed to fully validate the FAC parameterization we developed in our manuscript.

186            Andreae, M.O., Correlation between cloud condensation nuclei concentration and aerosol  
187 optical thickness in remote and polluted regions, *Atmos. Chem. Phys.*, 9, 543-556, 2009

188            Petters, M. D., and S. M. Kreidenweis, A single parameter representation of hygroscopic growth  
189 and cloud condensation nucleus activity. *Atmos. Chem. Phys.*, 7, 1961–1971, 2007

190            Schmale, J., Henning, S., Decesari, S., Henzing, B., Keskinen, H., Sellegri, K., Ovadnevaite, J.,  
191 Pöhlker, M. L., Brito, J., Bougiatioti, A., Kristensson, A., Kalivitis, N., Stavroulas, I., Carbone, S., Jefferson,  
192 A., Park, M., Schlag, P., Iwamoto, Y., Aalto, P., Äijälä, M., Bukowiecki, N., Ehn, M., Frank, G., Fröhlich, R.,  
193 Frumau, A., Herrmann, E., Herrmann, H., Holzinger, R., Kos, G., Kulmala, M., Mihalopoulos, N., Nenes,  
194 A., O'Dowd, C., Petäjä, T., Picard, D., Pöhlker, C., Pöschl, U., Poulain, L., Prévôt, A. S. H., Swietlicki, E.,  
195 Andreae, M. O., Artaxo, P., Wiedensohler, A., Ogren, J., Matsuki, A., Yum, S. S., Stratmann, F.,  
196 Baltensperger, U., and Gysel, M.: Long-term cloud condensation nuclei number concentration, particle  
197 number size distribution and chemical composition measurements at regionally representative  
198 observatories, *Atmos. Chem. Phys.*, 18, 2853-2881, <https://doi.org/10.5194/acp-18-2853-2018>, 2018.

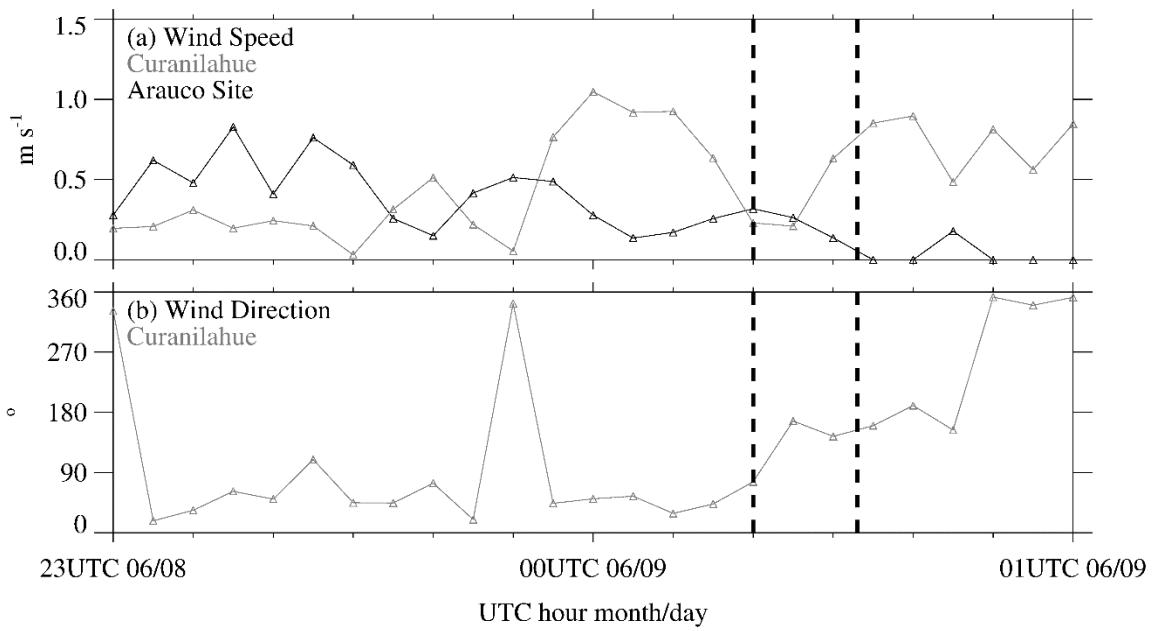
199



201

202 Figure 6: perhaps add local wind speed and direction to this figure?

203 We feel the verbal description – provided in the manuscript - is adequate. The graph is  
204 provided below, but this graph is not in the revised (or original) manuscript. In general, the  
205 effect of wind on aerosol is very difficult to interpret.



214

215 **Technical Corrections:**

216 Line 482: "was" should be "were"

217

218 **Corrected**