Reviewer-1

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

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

The paper presents results from a measurement campaign (CCOPE) on the Chilean Pacific Coast. The data consist of particle number concentrations measured with a condensation particle counter (CPC) and size distributions measured with a high-resolution optical particle counter (UHSAS) at a measurement station near the town of Arauco. The data are used for parameterizations of aerosol properties relevant to cloud and precipitation processes: number-to-volume ratios, concentrations of cloud condensation nuclei and sea-salt aerosol. The goal is to use these parameterizations for interpreting other data collected during the campaign on the Nahuelbuta Mountains about 30 – 100 km south of Arauco. The paper is basically well written and I can recommend its publication in ACP after some corrections and more detailed explanations.

It is in a way pleasant to see that it is still possible to make relevant observations even with such very simple traditional aerosol instrumentation when the setup in most similar campaigns today consists of several instruments measuring both physical properties and chemical composition. On the other hand, the lack of knowledge of size distributions at sizes smaller than those measured with the UHSAS, chemical composition and hygroscopicity increase the uncertainty of the interpretations. Discuss this.

We feel that the last four paragraphs of Section 4.4 address this. Please see Section 4.4 of the revised manuscript. Finally, since estimates of the effective supersaturation (Hudson 1984) are generally smaller than 0.2%, at least in liquid-only stratocumulus (Snider et al. 2017), we do not think that lack of knowledge at $D < 0.06$ μm is a limiting factor.

Hudson, J. G., 1984: Cloud condensation nuclei measurements within clouds. J. Climate Appl. Meteor., 23, 42–51, doi:10.1175/1520-0450(1984)023,0042:CCNMWC.2.0.CO;2.

Snider, J.R., D.Leon and Z.Wang, Droplet Concentration and Spectral Broadening in Southeast Pacific Stratocumulus, J. Atmos. Sci., 74, 719-749, 2017

The trajectories were calculated with HYSPLIT by using the GDAS wind data with a 0.5° spatial resolution. This is so coarse that the effects of local topography are not properly taken into account. The measurement site is very close to the town of Arauco and the sea, Gulf of Arauco is to the north of it and to the west of Arauco there are some hills higher than 300 m. As a result, even when the HYSPLIT trajectories show that wind blows from the west local wind in Arauco may have blown from other directions bringing anthropogenic aerosol from the town. The main goal of the paper is to use the parameterizations in the CCOPE data interpretations and modeling. During westerly winds the Nahuelbuta Mountains are definitely not affected by the anthropogenic sources around the Gulf of Arauco whereas your measurement station obviously is – the average total particle number concentration in air that you classified as "clean" was 2759 ± 1827 cm^{-3}. This is high compared with marine aerosol essentially everywhere, possibly also on the coast directly to the west of the Nahuelbuta Mountains. In light of this, discuss the validity of the results for CCOPE.

Yes, spatial resolution of the GDAS is a factor limiting our ability to stratify measurements made at the Arauco Site. In spite of the limitation, our conditional sampling does demonstrate that aerosol surface area at the Arauco site is, on average, smaller than that reported by Hegg and Kaufman (1998) over the western Atlantic in air that had advected from the United States. The comparison of aerosol surface area is discussed in Sect. 5 of the manuscript. Related to your point about representativeness, the Arauco CPC data can be used to generate lower and upper quartile values of N_{CPC} ensemble. The quartiles are 789 and 2151 cm^{-3}, respectively. We did not present these N_{CPC} quartiles in the manuscript, but they are easily derived using the N_{CPC} ensemble described in the Supplementary Material (manuscript) or using the data reader we provided (see section titled “Data Availability”). The lower quartile N_{CPC} (789 cm^{-3}) indicates that 25% of the time conditions were comparable to the wintertime average at THD (Section 4.1).

You also assert that “..directly west of Nahuelbuta Mountains..” a more pristine aerosol state may exist. We are not convinced this is true. In Fig. 1 (revised manuscript), Lebu (population 24,000) and Cañete (population 32,000) are included. Another small city (Curanilahue) was in Figure 1 of the original manuscript. These small cities increase the possibility that cloud and precipitation over the Nahuelbuta are impacted by anthropogenic aerosols, even in a westerly flowing air. Furthermore, source/receptor relationships for aerosols on the Central Chilean Coast depend on source strength and

a host of meteorological factors (e.g., extratropical cyclone track, thermal stability, and etc.). Onshore/offshore flow that occurs during meteorologically quiescent periods (sea/land breeze circulations), could also be significant. For example, if the sea/land circulation creates a “strip” of aerosol contamination within the near-shore zone, and this air is brought onshore during episodes of persistent westerly airflow. A “coastal strip” of larger cloud droplet concentration is evident in analyses of satellite retrievals in Wood et al. (2012; their figure 4). The latter compliments the retrievals of Bennartz (2007), who we cite in the manuscript (Sect. 5). However, neither Wood et al. (2012) nor Bennartz (2007) segregate the satellite data into wintertime and summertime ensembles. As we state in the manuscript (Sect. 5), further analysis of the satellite retrievals are needed to investigate if the coastal strip exists both in winter and in summer.

The previous paragraph focused on aerosol-cloud interactions occurring within the planetary boundary layer; an additional dimension of the problem is aerosol resident above the planetary boundary layer. We acknowledge this in Sect. 5 (original and revised manuscript).

In summary, we feel that the caveats provided in the manuscript (Sections 5 and 6) are sufficient for numerical modelling of wintertime Chilean Coastal clouds and precipitation. We are confident that such modelling will extend understanding beyond the analyses provided here and in Massmann et al. (2017).

Hegg, D. A., and Y. J. Kaufman, Measurements of the relationship between submicron aerosol number and volume concentration, J. Geophys. Res., 103, 5671-5678, 1998

Massmann, A.K., J.R. Minder, R.D. Garreaud, D.E. Kingsmill, R.A. Valenzuela, A. Montecinos, S.L. Fults, and J.R. Snider, 2017, The Chilean Coastal Orographic Precipitation Experiment: Observing the Influence of Microphysical Rain Regimes on Coastal Orographic Precipitation. J. Hydrometeor., 18, 2723–2743, https://doi.org/10.1175/JHM-D-17-0005.1, 2017

Bennartz, R., Global assessment of marine boundary layer cloud droplet number concentration from satellite, J. Geophys. Res., 112, D02201, 2007

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

Detailed comments

Section 2.1. Add information on the distance of the Arauco measurement site from the sea, from the town of Arauco, the paper mill, the Curanilahue measurement station and the rest of the CCOPE campaign area.

A distance scale is provided in Fig. 1 (revised manuscript). Also, a city Coronel (population 110,000), and two small cites Lebu (population 24,000) and Cañete (population 32,000) are included in the revised Fig. 1.

L145-146 "*... CPC concentrations were recorded once per second and once every 10 seconds (Table 1)*."

The expression "CPC concentrations" would mean there are many Condensation Particle Counters flying in the air. That is not quite correct. Use "... CPC data were recorded..."

Corrected

Another thing I don't understand, is the logic of saving data once per s and once per 10 s. The 1-s data has it all, from it 10-s data can be picked up if needed. What is the logic?

The text was revised:

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

105 The expressions "***CPC concentration***" and "***UHSAS concentration***" have been used in some sentences
106 also later. As I wrote above, these should be rewritten. For example title of section 4.1 should rather
107 be "Comparison of particle number concentrations..."

108 Corrected.

109 L256-258 " *… 194 classify as clean sector. For both sites we required a clean sector wind speed > 1.5 m*
110 *s-1 in* addition to the clean sector directional criteria (Fig. 2)."

111 You started wind measurements at Arauco on 19 June. Did you use only the aerosol data after that in
112 this comparison?

113 Yes.

114 L286-289 " ***During this two-hour data segment, centered on 00 UTC June 9 (9 pm local time), winds were light at***
115 ***Arauco and Curanilahue (< 2 m s_{-1}) and the wind direction was variable at Curanilahue (Arauco Site wind direction***
116 ***measurements are only available after 19 June 2015; Sect. 2.1).***"

117 You wrote that wind measurements at Arauco started on 19 June. How can you then write that the
118 wind at Arauco was < 2m/s on 9 June?

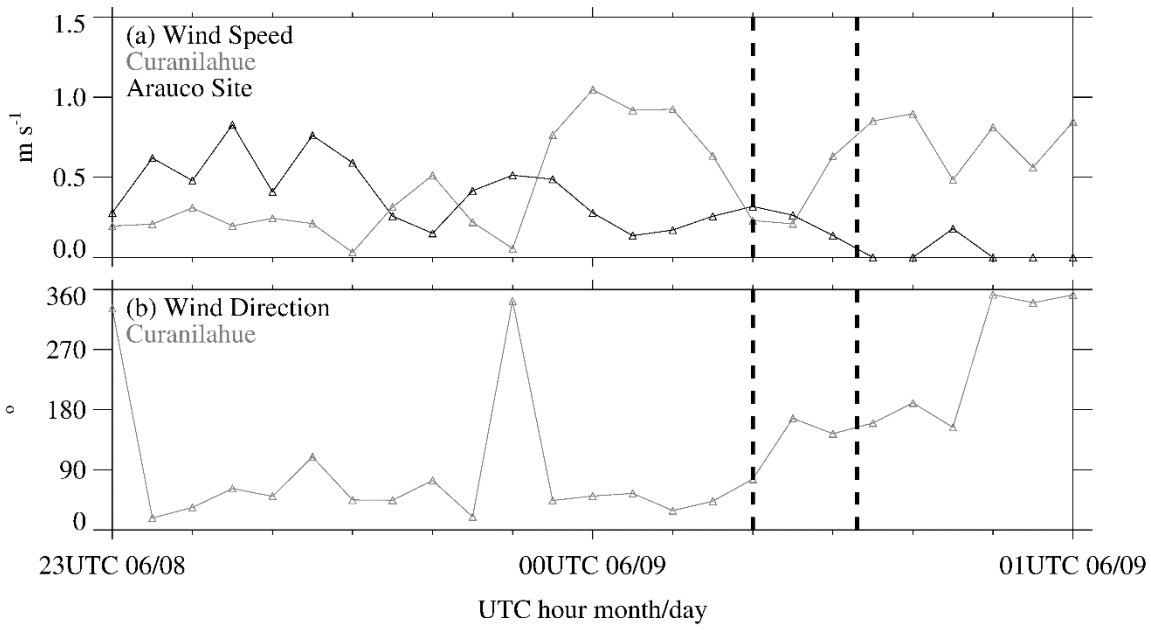
119 Meteorological measurements (minus wind direction) were acquired from 29 May to 14 August
120 and meteorological measurements (including wind direction) were acquired 19 June to 14 August. This
121 is stated in Section 2.1 (original and revised manuscript).

122

123 The distance between Arauco and Curanilahue is approximately 25 km, the measurement site of
124 Curanilahue is at > 100 m ASL and there are quite a few valleys and hills higher than 100 m ASL between
125 the two sites. So the local winds at these sites may have been completely different. How justifiable is it
126 to use Curanilahue in interpreting Arauco data?

127 Reviewer #1 also commented on this, and we responded. Wind speeds were light at both
128 locations and direction was variable at Curanilahue. A graph of the data is provided below. In general,
129 the effect of wind on aerosol is very difficult to interpret.

130
131
132



Section 4.3

In calculating the N/V ratio, justify using N_{UHSAS} and not N_{CPC} for N?

We rewrote this section of the manuscript. We feel the revision justifies what you commented on:

"In this section we analyze two ASD moments (Section 3.3). These are symbolized N_{UHSAS} and V_{UHSAS}, respectively. The ratio of N_{UHSAS} (aerosol concentration) and V_{UHSAS} (aerosol volume) – generically the *N/V* ratio - is of interest for several reasons. First, for both operational and theoretical reasons the *N*/*V* ratio is evaluated for particle diameters larger than ~ 0.1 μm (VD00; Hegg and Kaufman 1998, hereafter HK98), and importantly, the model developed to evaluate aerosol exchange between an overlying free troposphere (FT) and the marine boundary layer (MBL) successfully predicts the *N*/*V* ratio in the MBL (VD00). Second, a value of the ratio can be derived by fitting measurements of *N* and *V* (HK98). Third, aerosol mass loading, and thus an aerosol volume corresponding to an assumed particle density [1], are relatively easy to evaluate. A method routinely used to evaluate aerosol mass loading involves pulling aerosol-laden air through a filter and evaluating the accumulated mass gravimetrically. Fourth, the product of an *N*/*V* ratio and an ambient aerosol volume (aerosol mass) has been proposed as a scheme for estimating cloud droplet concentration in marine stratocumulus clouds (HK98 and VD00).

HK98 used a passive cavity aerosol spectrometer probe (PCASP) to evaluate *N*, *V* and the *N/V* ratio. Since the UHSAS counts down to a smaller diameter (0.055 μm) than the PCASP (0.12 μm), it is expected that the *N/V* ratios we derive using the UHSAS will be larger than those in HK98. The main reason for this is that decreasing the lower-limit diameter increases *N* more than *V* (VD00). "

Hegg, D. A., and Y. J. Kaufman, Measurements of the relationship between submicron aerosol number and volume concentration, J. Geophys. Res., 103, 5671-5678, 1998

van Dingenen, R., A. O. Virkkula, F. Raes, T. S. Bates, A. Wiedensohler, A simple non linear analytical relationship between aerosol accumulation number and sub-micron volume, explaining their observed ratio in the clean and polluted marine boundary layer, Tellus, 52B, 439-451, 2000

[1] In the case of ambient particles containing hygroscopic materials, density values range between 1.5 and 1.8 g cm^{-3} (McMurry et al. 2002)

What did HK98 and VD00 use?

This information is provided in Sect. 4.3. First we present *N*/*V* ratios derived with the lower-limit diameter set at the minimum particle diameter detected by the UHSAS. Next, we repeat the analysis with the lower-limit diameter equal to the value applied by HK98. Results are in Tables 3 and 4. The "headline" of these Tables provides the distinction. Additionally, VD00 integrate from minimum diameter = 0.08 μm, but we do not consider that case.

Section 4.4

L377-385 This is an important part of the paper and it should be understood properly in order to understand the parameterization FAC(SS) presented later. Now it is not quite clear to me. You have earlier presented some of the simplest possible aerosol equations, Eqs. (1) – (4), which is fine, they are good to be shown. But now when it comes to a clearly more complicated issue, equations are missing which is not logical. And on line 379 it is written "***... kappa–Köhler formula of Petters and Kreidenweis (2007, their Eq. (11))***" but their Eq. (11) shows the relationship of growth factor, dry particle diameter, kappa, and relative humidity. How is this used to "***...interpret a FAC's lower-limit diameter as an upper-limit SS***" as was stated on line 377? Is the referred equation right? Write the proper equation and explain the steps of the calculation in more detail so that readers can repeat the calculation for their own data.

The relevant equation from Petters and Kreidenweis (2007) was cited incorrectly. This is changed in the revised manuscript. For calculating critical SS, corresponding to prescribed values of dry diameter and kappa, we used Eq. 6 (Petters and Kreidenweis 2007). This is corrected in the revised manuscript. Additionally, our explanation is enhanced by inclusion of Eq. 5 (revision).

Here is the revised text:

"Our first step is to select a particle diameter, apply this as a lower-limit diameter in an integration of the UHSAS size distribution, and divide the integral by the coincident CPC-measured concentration. The resultant is referred to as the *fractional aerosol concentration* (*FAC*).

$$FAC(D) = \frac{1}{N_{CPC}} \cdot \int_{D}^{1\mu m} \left(dN / d\log_{10} D\right) \cdot d\log_{10} D \qquad (5)$$

Figs. 7a - b have graphical representations of *FAC*(*D*=0.055 μm) and *FAC*(*D*=0.120 μm).

In a second step we interpret a *FAC*'s lower-limit diameter as an upper-limit *SS*. We do this by applying a value for the kappa hygroscopicity parameter, which we set at κ = 0.5, and by applying the kappa–Köhler formula of Petters and Kreidenweis (2007, their Eq. (6)). This transformation from lower-limit *D* to upper-limit *SS* converts the *FAC* in Fig. 7a to *FAC*(*SS* = 0.41 %) and the FAC in Fig. 7b to *FAC*(*SS* = 0.13 %). We also evaluated how a range of the kappa parameter ($0.3 < \kappa < 0.7$) translates to a range of *SS*. Our upper-limit κ comes from airborne measurements made over the Southeast Pacific Ocean during

summer (Snider et al., 2017), and our lower-limit κ is the value recommended by Andreae and Rosenfeld (2008) for simulating aerosol indirect effects over continents."

Additionally, we rewrote the paragraph explaining how FACs are derived for onshore trajectories. The revised paragraph is this:

"The *FAC*s in Figs. 7a – b are two of the many available from CCOPE. One way to aggregate these is to calculate a *FAC* for each of the 20 onshore trajectories. For example, if we select the lower-limit diameter at D = 0.055 μm, plot numerator values (Eq. (5)) vs denominator values (Eq. (5)), and fit with the equation $Y = a \cdot X$, the "a" we derive is the $FAC(D = 0.055\ \mu m)$ for a particular trajectory. *FAC*s calculated in this way, and with lower-limit D selected = 0.120 μm, are presented in the seventh columns of Tables 3 and 4. Correlation coefficients presented in the eighth columns of these tables mostly exceed 0.5. By averaging over the 20 onshore trajectories, we calculated the overall averages presented at the bottom of the two tables. These overall averages are $FAC(D = 0.055\ \mu m) = 0.35 \pm 0.13$ (Table 3) and $FAC(D = 0.120\ \mu m) = 0.13 \pm 0.07$ (Table 4). This decrease of the *FAC* results because a larger lower-limit D (Eq. (5)), implies a smaller numerator (Eq. (5)), and thus a smaller $FAC(D)$."

Section 4.5

Refer also to O'Dowd, C. D. and de Leeuw, G. (2007) and consider comparing your results also with the parameterization they presented

O'Dowd, C. D. and de Leeuw, G.: Marine Aerosol Production: a review of the current knowledge, Phil. Trans. R. Soc. A., 365,1753–1774, doi:10.1098/rsta.2007.2043, 2007

O'Dowd and de Leeuw (2007) summarize the sea spray research of Geever et al. (2005) and Clarke et al. (2006). The latter two references are not compiled in Lewis and Schwartz (2004) (hereafter LS04). We reference LS04 and Clarke et al. (2006) in the manuscript (original and revised).

Clarke et al. (2006) report a particle size-dependent flux function. As discussed in de Leeuw et al. (2011) (their section 6.5), a *size-dependent flux* can be transformed to a *concentration*, corresponding to a specified range of particle size, but this requires a steady-state, an assumed value for atmospheric residence time, and an assumed value for the depth of the MBL. Geever et al. (2005) investigated sea spray from particles smaller than 1 μm, but did not report a size-dependent flux function.

Using the Clarke et al. (2006) parameterization with a range of wind speeds (3, 6, and 12 m/s), we transformed to concentrations assuming residence time = 3 day and MBL depth = 500 m (de Leeuw et al. (2011); their section 6.5). The SSA concentrations we calculated are within a factor = 3 of the CCOPE curve in Fig. 9. Specifically, the calculated values are smaller at 3 m/s (Fig.9-to-calculated ratio = 1.3) and larger at 12 m/s (Fig.9-to-calculated ratio = 0.33). Given that there is significant variability in residence time and MBL depth, and in the wind speed scaling applied in Clarke et al. (2006), the result in Fig. 9 (manuscript) seems reasonable.

Summary: Because of assumptions necessary to transform a size-dependent flux to a concentration, we have not compared our result to sea spray research other than the comparison to wind-speed-dependent concentrations presented in O'Dowd and Smith (1993).

Clarke, A., V. Kapustin, S. Howell, K. Moore, B. Lienert, S. Masonis, T. Anderson, and D. Covert, Sea-salt size distribution from breaking waves: Implications for marine aerosol production and optical extinction measurements during SEAS, J. Atmos. Ocean.Technol., 20, 1362–1374, 2003

Geever, M., C. D. O'Dowd, S. van Ekeren, R. Flanagan, E. D. Nilsson, G. de Leeuw, and Ü. Rannik, Submicron sea spray fluxes, Geophys. Res. Lett., 32, L15810, doi:10.1029/2005GL023081, 2005

de Leeuw, G., E. L Andreas, M. D. Anguelova, C. W. Fairall, E. R. Lewis, C. O'Dowd, M. Schulz, and S. E. Schwartz, Production flux of sea spray aerosol, Rev. Geophys., 49, RG2001, doi:10.1029/2010RG000349, 2011

O'Dowd, C. and G. de Leeuw, Marine aerosol production: a review of the current knowledge, Phil. Trans. R. Soc. A., 365,1753–1774, doi:10.1098/rsta.2007.2043, 2007

O'Dowd, C.D., and M.H. Smith, Physicochemical properties of aerosols over the Northeast Atlantic: evidence for wind-speed-related submicron sea-salt aerosol production, J.Geophys. Res.,98, 1137-1149, 1993

Fig 1. Add a distance scale.

Fig. 1 (revised manuscript) has a distance scale. The revised map is shown below. Small cites Cañete and Lebu, and the city Coronel, are included in the revised Figure 1.

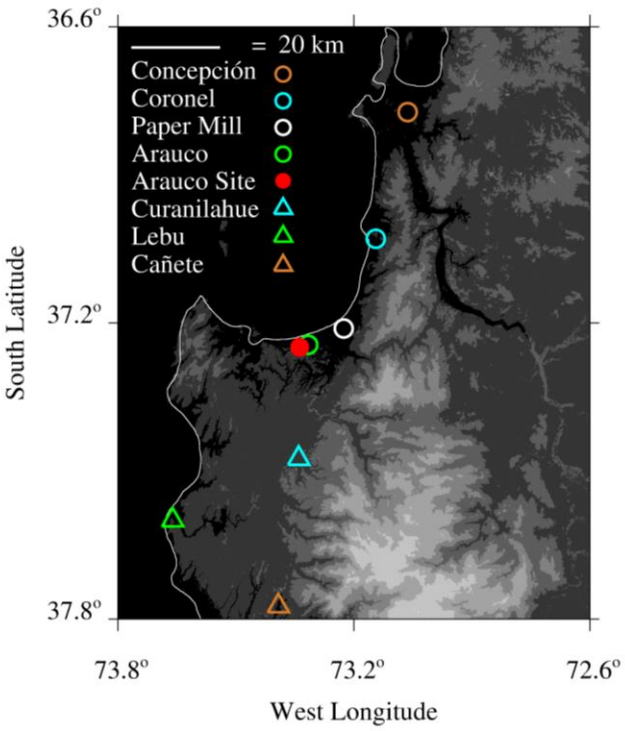


Fig. 3b. Why is the y axis reverse? Why is the lowest pressure 920 hPa? A sensible scale would be 990-1020 hPa.

An air parcel's barometric pressure is output by the HYSPLIT model. Fig. 3b (original manuscript) has this pressure on the Y axis. Pressure, decreasing upward on the Y axis, is a proxy for altitude. In the revised Fig. 3b (see below), the MSL altitude of the air parcel is plotted. MSL altitude was calculated using the pressure output by HYSPLIT (parcel barometric pressure) and the ICAO equation for the Standard Atmosphere (1993). MSL altitude increases if a larger sea-level is pressure applied in the ICAO equation. This sensitivity is ~ 8 m / hPa.

International Civil Aviation Organization (ICAO), Manual of the ICAO Standard Atmosphere: extended to 80 kilometres (262500 feet), 3rd ed., ISBN-92-9194-004-6, 1993

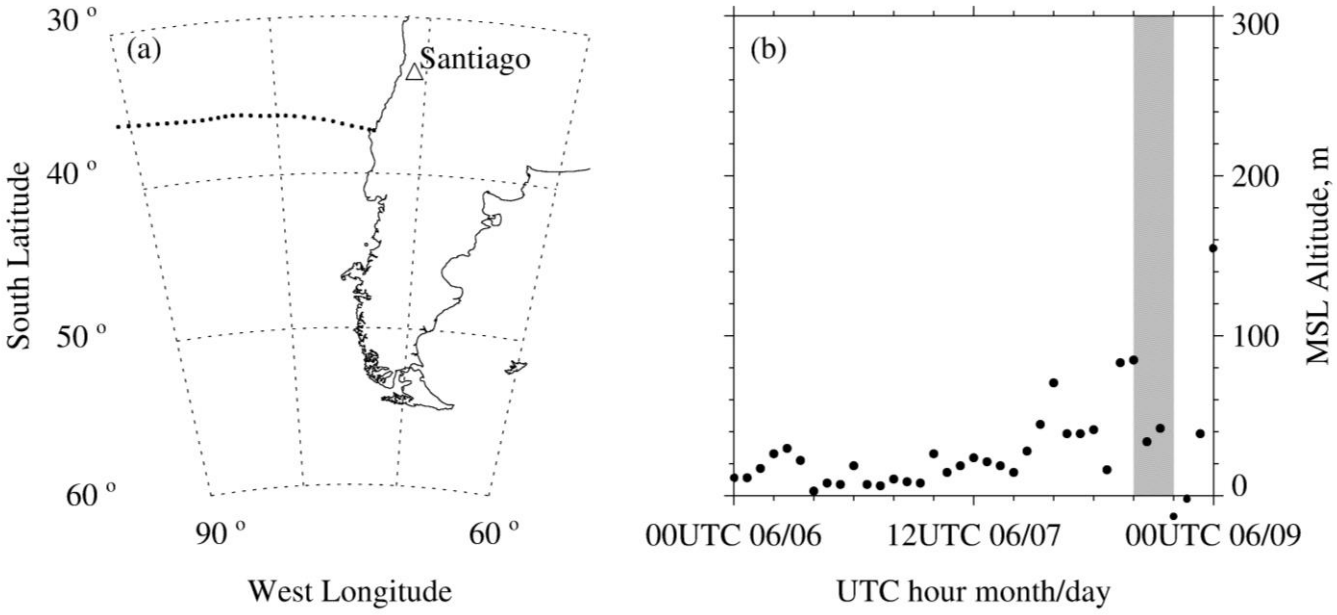


Fig. B1. What is the vertical dashed line at ~11:33 UTC?

This is explained in the original manuscript (Appendix B). Readers are referred to Appendix B at L194. The first paragraph of Appendix B (revised manuscript) was revised for clarity. Here is the revised text:

"For each of the onshore trajectories (Sect. 3.1), a two-hour segment, centered on the trajectory arrival time was analyzed. An example is in Figs. B1a – e. The first panel (Fig. B1a) shows the sequence of CPC values sampled every second (i.e., 1-s samples referred to as *fast* N_{CPC}), and Fig. B1b shows CPC values sampled every 10 seconds (i.e., 10-s samples referred to as *slow* N_{CPC}). The following procedure was used to attenuate the narrow perturbations that were likely the result of local aerosol emissions (e.g., within the time interval indicated by vertical dashed lines in Figs. B1a, B1b, and B1d)."