CLOUD CONDENSATION NUCLEI SIZES

James G. Hudson
and
Stephen Noble
Desert Research Institute
Reno, Nevada

1. INTRODUCTION

The physical size of cloud condensation nuclei (CCN) reveals important clues about CCN composition and origins. Among other considerations this can help distinguish natural from anthropogenic particles. The increasing importance of CCN to global climate considerations, the difficulty of making accurate CCN measurements and the sparsity of CCN measurements led Dusek et al. (2006) to investigate the possibility of deducing CCN concentrations and spectra from particle size measurements. These are much easier to obtain and are more readily available than CCN measurements.

Dusek et al. (2006) found a rather limited range of CCN sizes that suggested that indeed it may be possible to use particle size distribution measurements to estimate CCN spectra. However, Hudson (2007) pointed out that all of the measurements considered by Dusek et al. (2006) had apparently been made in rather polluted air masses. Hudson (2007) presented measurements in a variety of different air masses that showed so much variability in the sizes of CCN that it would be extremely difficult to deduce accurate CCN concentrations from particle size distribution measurements alone.

On the other hand Dusek et al. (2006) also considered the possibility that the relationship between particle size and CCN critical supersaturation (S_c) may depend on air mass. They suggested that the range of CCN sizes may still be sufficiently limited within each air mass so that CCN spectra can be accurately determined from particle size measurements. This assumes that an accurate relationship between particle size and CCN S_c can be determined in each air mass and that the air mass of the measurements can be correctly ascertained. Here we present further measurements of CCN sizes that address this controversy.

2. MEASUREMENTS

Size-S_c measurements are more easily done with the DRI instruments because they simultaneously provide the entire CCN spectrum. The mean S_c obtained from these instruments of measurements of narrow size slices from a differential mobility analyzer (DMA) produces the size-S_c data. The latest data are surface measurements in Reno, Nevada and Seoul, Korea and aircraft measurements in the 2007 PASE and ICE-L projects.

The Pacific Aerosol Sulfate Experiment (PASE) was conducted over the central Pacific near the equator directly south of Hawaii. This is as far as possible from continental or anthropogenic sources. The ICE-L project was conducted over Colorado and Wyoming. Hudson (2007) and Hudson and Da (1996) showed that maritime CCN tend to be smaller than continental or especially polluted CCN (i.e., Figs. 1 and 2). Figure 1 shows that the CCN sizes in maritime air tended to be like soluble salts such as NaCl or ammonium sulfate. On the other hand Figure 2 in continental air masses showed larger CCN sizes for the same S_c values.

3. RESULTS

Figures 3 and 4 show measurements from PASE that are largely consistent with previous marine measurements—relatively small CCN similar to Fig. 1. However at the largest sizes (above 150 μm) the S_c values tend to be higher than those of ammonium sulfate; i.e., more like the continental/polluted measurements. To compare different sizes or S_c Fitzgerald et al. (1982) introduced the hygroscopicity or solubility parameter, B, which is non-dimensional. For NaCl this is 1.23 and for ammonium sulfate it is 0.70. B is lower for less hygroscopic substances, which have to be larger to produce the same S_c values as more hygroscopic substances. B tends to be higher in maritime air masses and lower in more polluted air masses.

Figure 5 shows B for the data in Fig. 3. For most of the PASE measurements B, displayed this same inverse function of particle size. Volatility measurements during PASE indicated that the vast majority of CCN behaved like ammonium sulfate. The volatility measurements showed that very few CCN could be NaCl. The B values of the larger CCN suggested that they may have been internal mixtures of ammonium sulfate and insoluble or less soluble materials such as organics. Reanalysis of Hudson (2007) often showed somewhat the same tendency in other maritime air (Fig. 6). Figure 7 shows that B often tends oppositely in continental air where B often increased with CCN size. Figures 8-15 show further examples from PASE of the tendency for B to decrease with particle size. However, the relationship of B with particle size showed considerable variability; i.e., different slopes.

Figures 16 and 17 display one of the data points displayed in Figs. 4 and 10 respectively. Figures 16 and 17 display both the spectra that produced one of the data points in Figs. 4 and 10 and one of the calibration points used to produce the calibration curve that relates the raw data channels to S_c. The ambient spectra is plotted in green and the calibration spectra is plotted in red. Since these are for the same DMA sizes they clearly show the differences in behavior within the cloud chamber between the ammonium sulfate particles used for the calibrations and ambient particles. The ambient particles grew smaller droplets that were detected in lower voltage (droplet size) channels. This shows specifically how these ambient particles were not the same as ammonium sulfate particles; they have lower B values.

Figure 18 shows a further complication for CCN size-S_c measurements that can only be revealed with spectral instruments such as the DRI CCN spectrometers. This is a bimodal spectrum that was produced here by the exhaust from a diesel generator superimposed upon the ambient aerosol. The right hand mode is the ambient maritime distribution similar to Figs. 16 and 17 whereas the left hand mode was caused by particles from the diesel generator. Although this was a somewhat contrived situation it probably represents a naturally mixed air mass that could occur when polluted air advects over the ocean. This shows the inadequacy of using only the mean S_c of a measurement. Figures 19 and 20 display the differences between the single mode and the bimodal distribution.

Figure 21 exhibits the complication of the width of the spectra. Panel A displays broad spectra that are typically observed in polluted air compared with the narrower spectra often observed in maritime air masses. This also shows the inadequacy of mean S_c alone to describe the relationship of size with S_c.

Figure 22 shows that B is not always related to the total particle concentration (CN), which is often used to characterize whether an air mass is clean (maritime) or polluted. This makes it more difficult to know which size-S_c relationship to use in order to deduce CCN from particle size measurements.

4. CONCLUSIONS

The results presented exhibit some of the difficulties associated with efforts to deduce CCN concentrations from particle size measurements.

Acknowledgements. This work was supported by NSF grants ATM-0342618, ATM-0313899, and NSF subaward 235435 from Drexel CCN in PASE.

References:

Dusek, U., G.P. Frank, L. Hildebrandt, J. Curtius, J. Schneider, S. Walter, D. Chand, F. Drewnick, S. Hings, D. Jung, S. Borrmann, M.O. Andreae (2006), Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science, 312*, 1375-1378.

Fitzgerald, J.W., W.A. Hoppel, and M.A. Vietti (1982), The size and scattering coefficient of urban aerosol particles at Washington, DC as a function of relative humidity, *J. Atmos. Sci.*, *39,* 1838-1852.

Hudson, J.G., 2007: Variability of the relationship between particle size and cloud-nucleating ability. *Geophys. Res. Let*., **34**, L08801, doi:10.1029/2006GL028850.

Hudson, J.G. and X. Da, 1996: Volatility and size of cloud condensation nuclei. *J. Geophys. Res., **101***, 4435-4442.

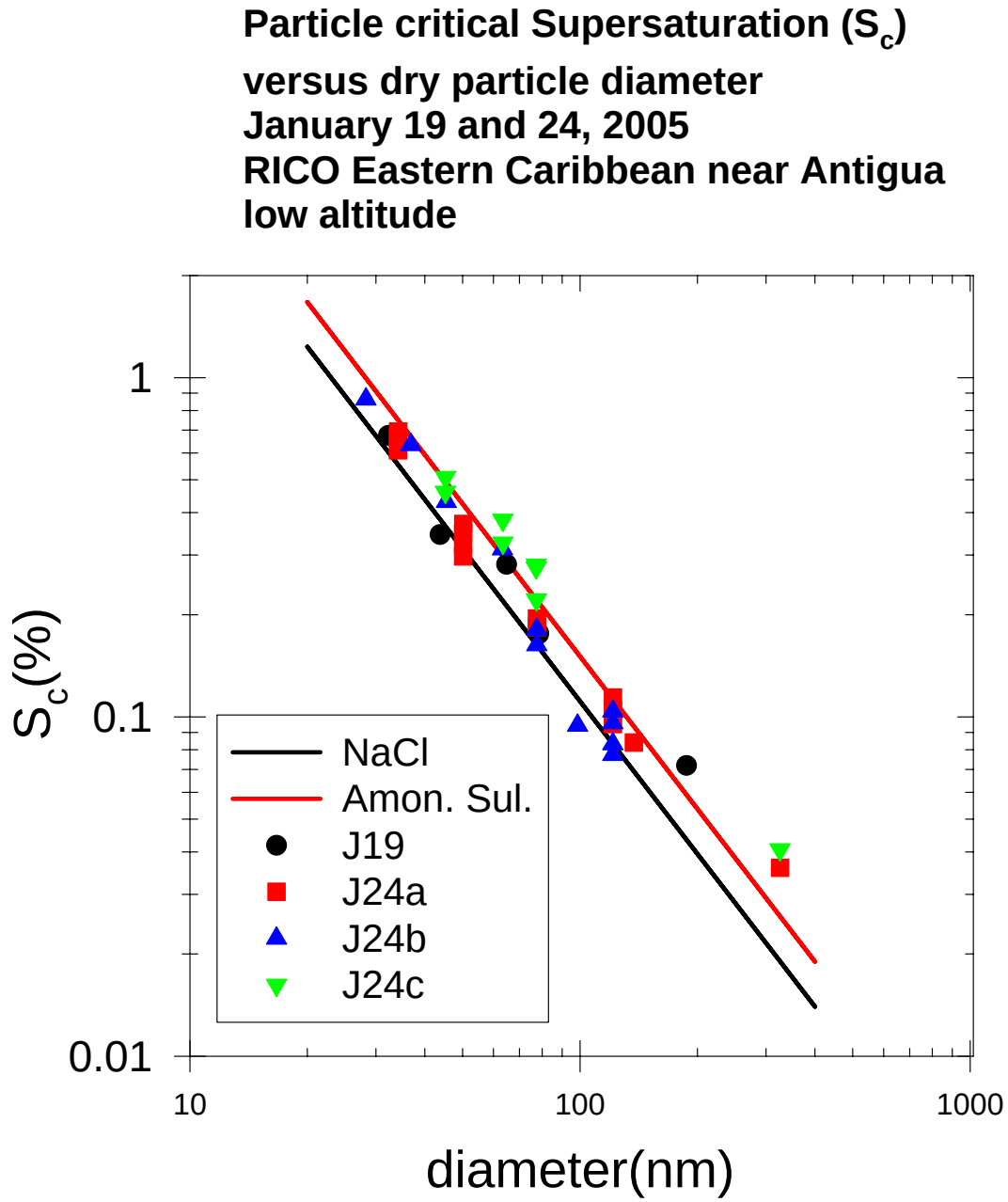


Figure 1. Critical supersaturaiton (S_c) versus dry particle size for Several measurements in clean maritime air masses (from Hudson [2007]).

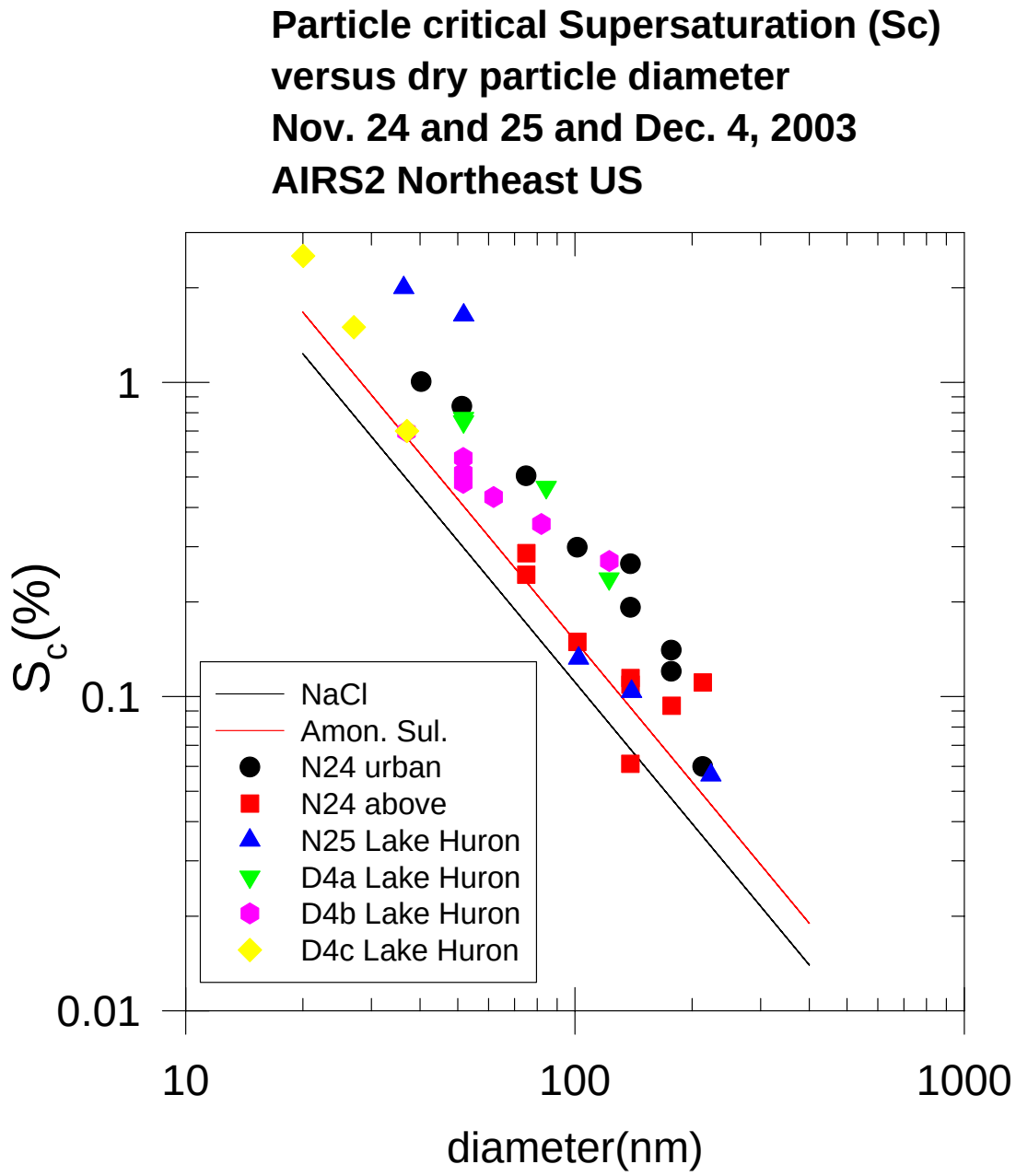


Figure 2. As Fig. 1 but for continental air masses (from Hudson [2007]).

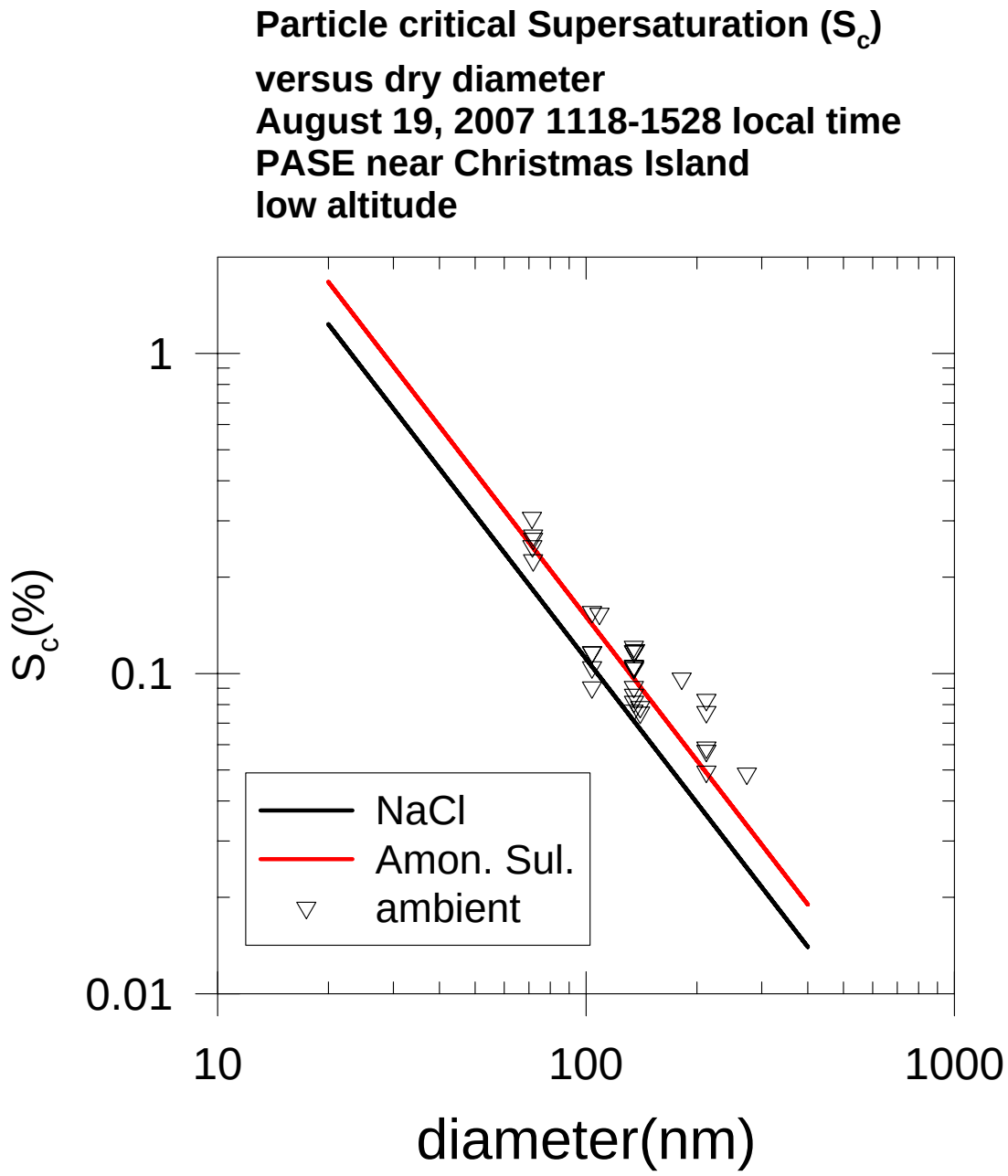


Figure 3. As Fig. 1 but for the recent PASE project.

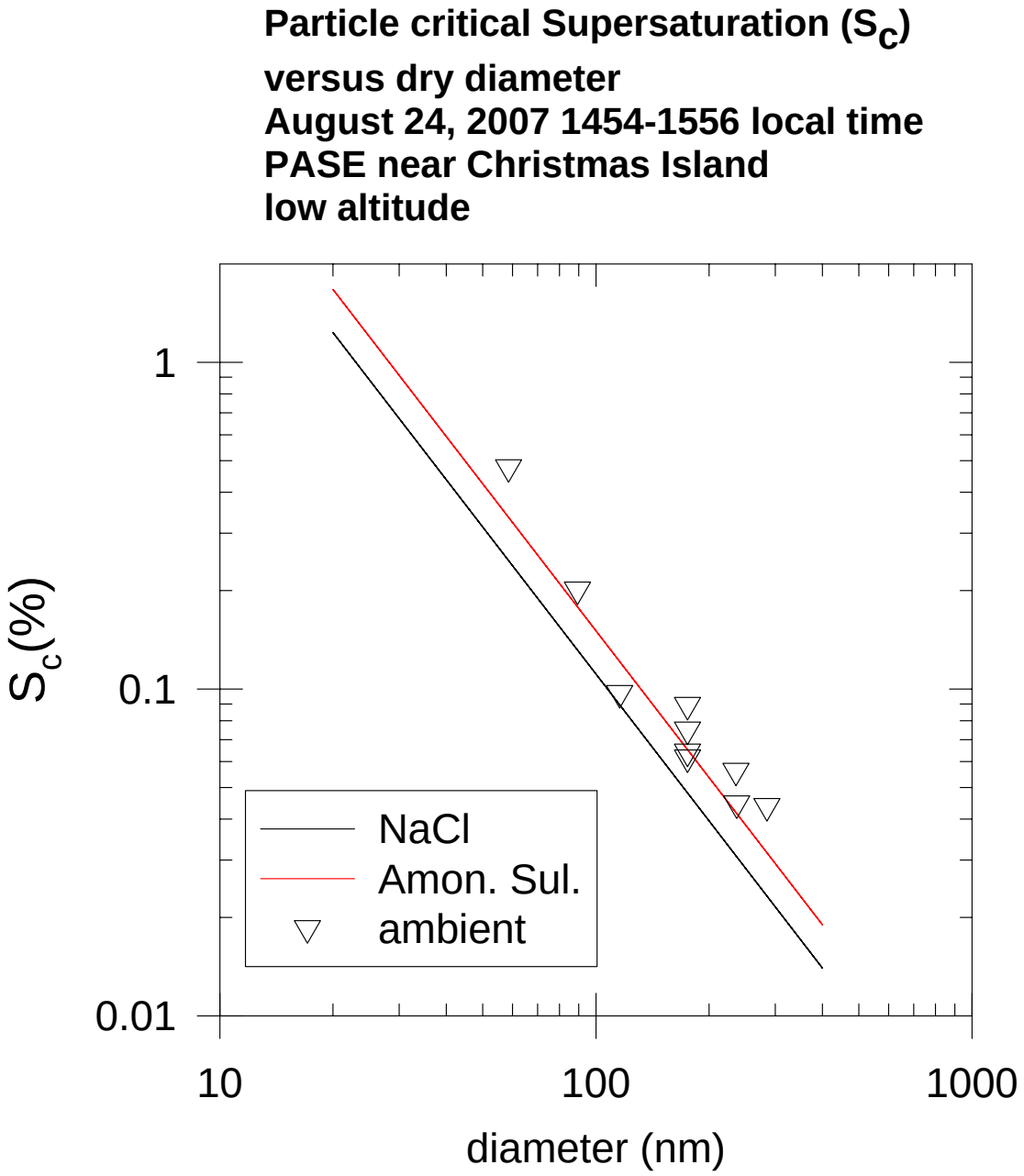


Figure 4. As Fig. 3 for a different flight.

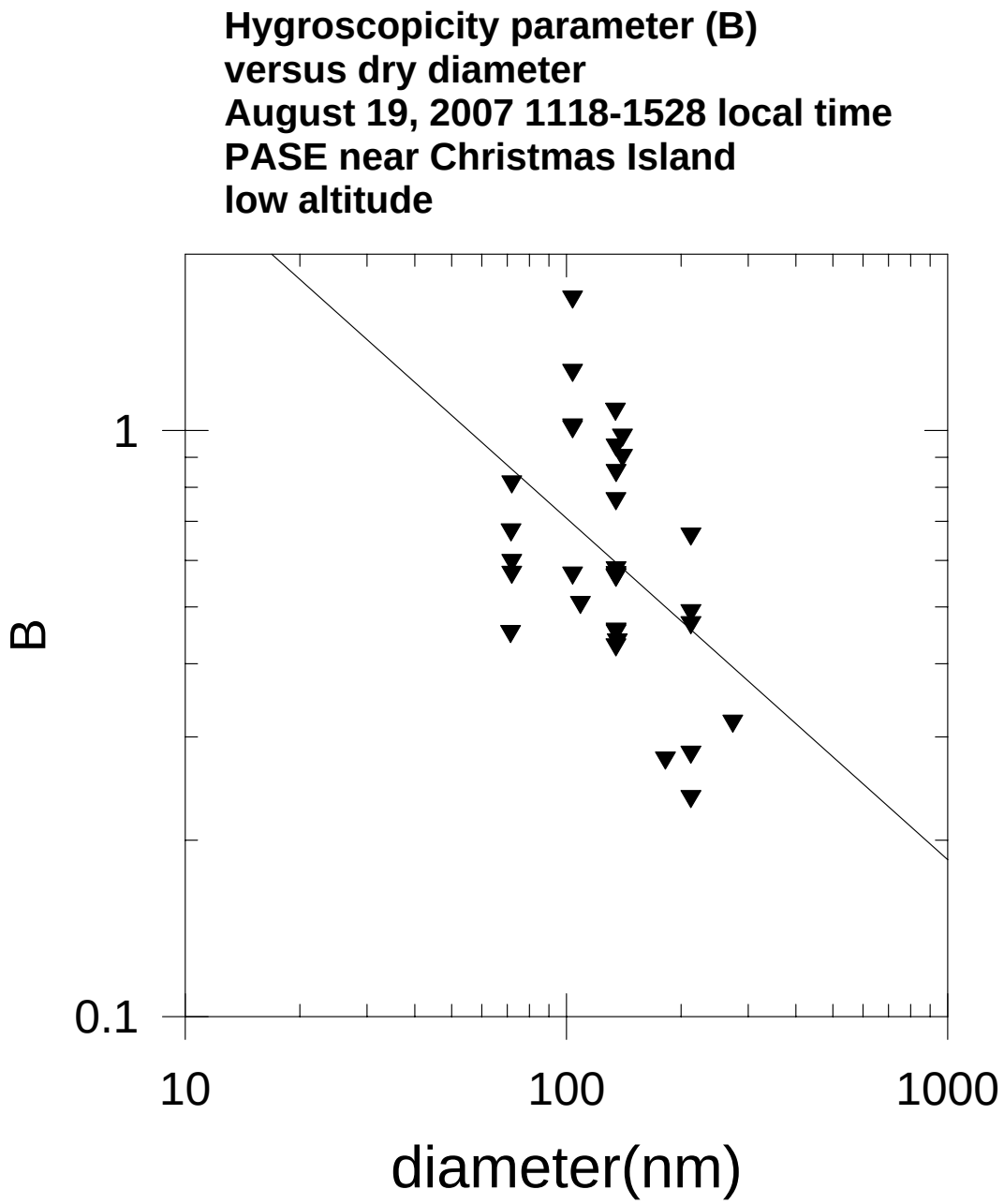
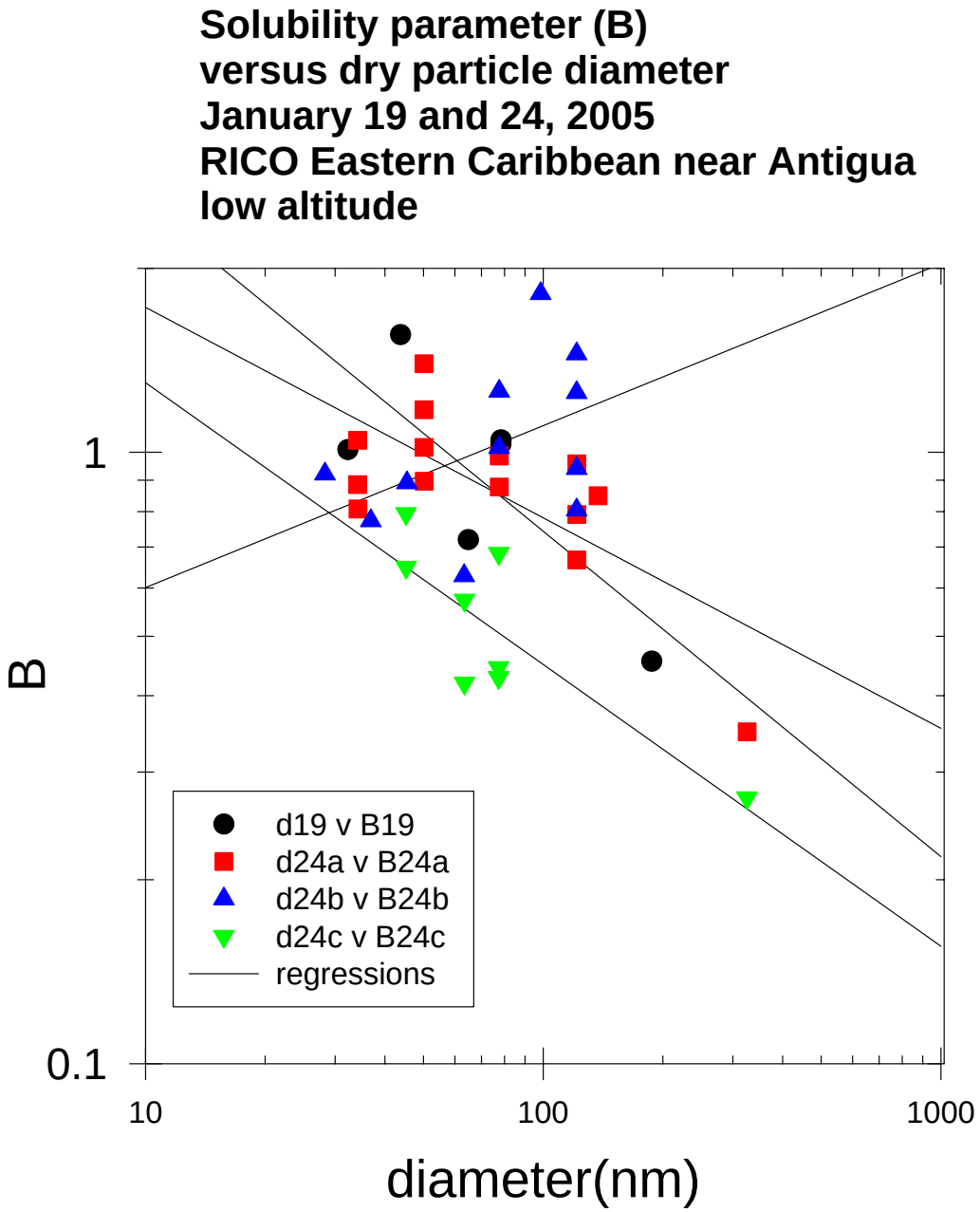


Figure 5. Hygroscopicity parameter (B) versus dry particle size for the data displayed in Fig. 3.



D19
y = 0.94 -0.53x
r ² = 0.59

D24a
y = 0.58 -0.34x
r ² = 0.51

D24b
y = -0.49 + 0.26x
r ² = 0.20

D24c
y = 0.57 -0.46x
r ² = 0.69

Figure 6. As Fig. 5 but for the data displayed in Fig. 1. Also shown are the linear regressions.

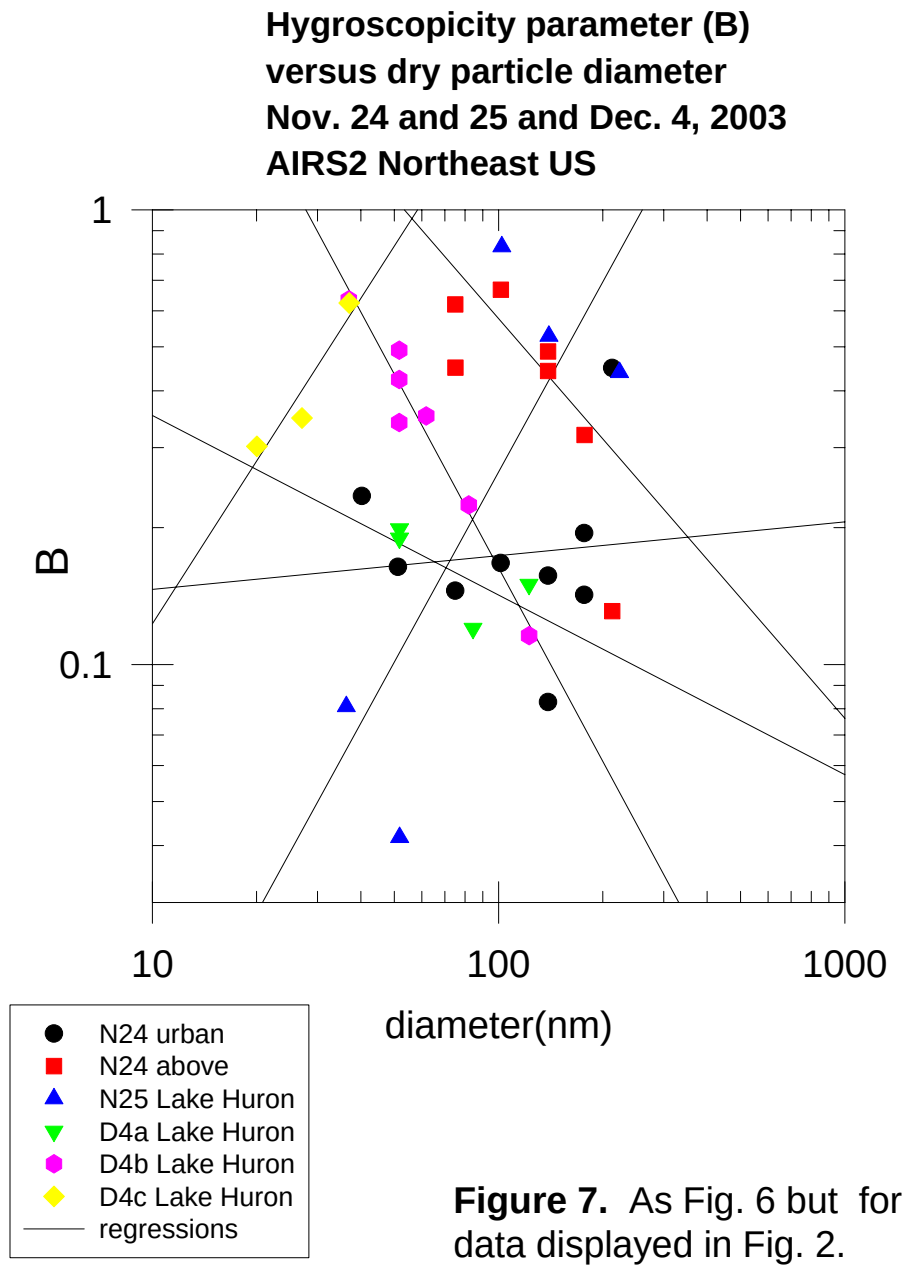


Figure 7. As Fig. 6 but for the data displayed in Fig. 2.

N24 urban
$y = -0.91 + 0.07x$
$r^2 = 0.009$

N24 above
$y = 1.52 - 0.88x$
$r^2 = 0.23$

N25 Lake Huron
$y = -3.35 + 1.39x$
$r^2 = 0.62$

D4a Lake Huron
$y = -0.057 - 0.40x$
$r^2 = 0.51$

D4b Lake Huron
$y = 2.04 - 1.41x$
$r^2 = 0.96$

D4c Lake Huron
$y = -2.10 + 1.19x$
$r^2 = 0.90$

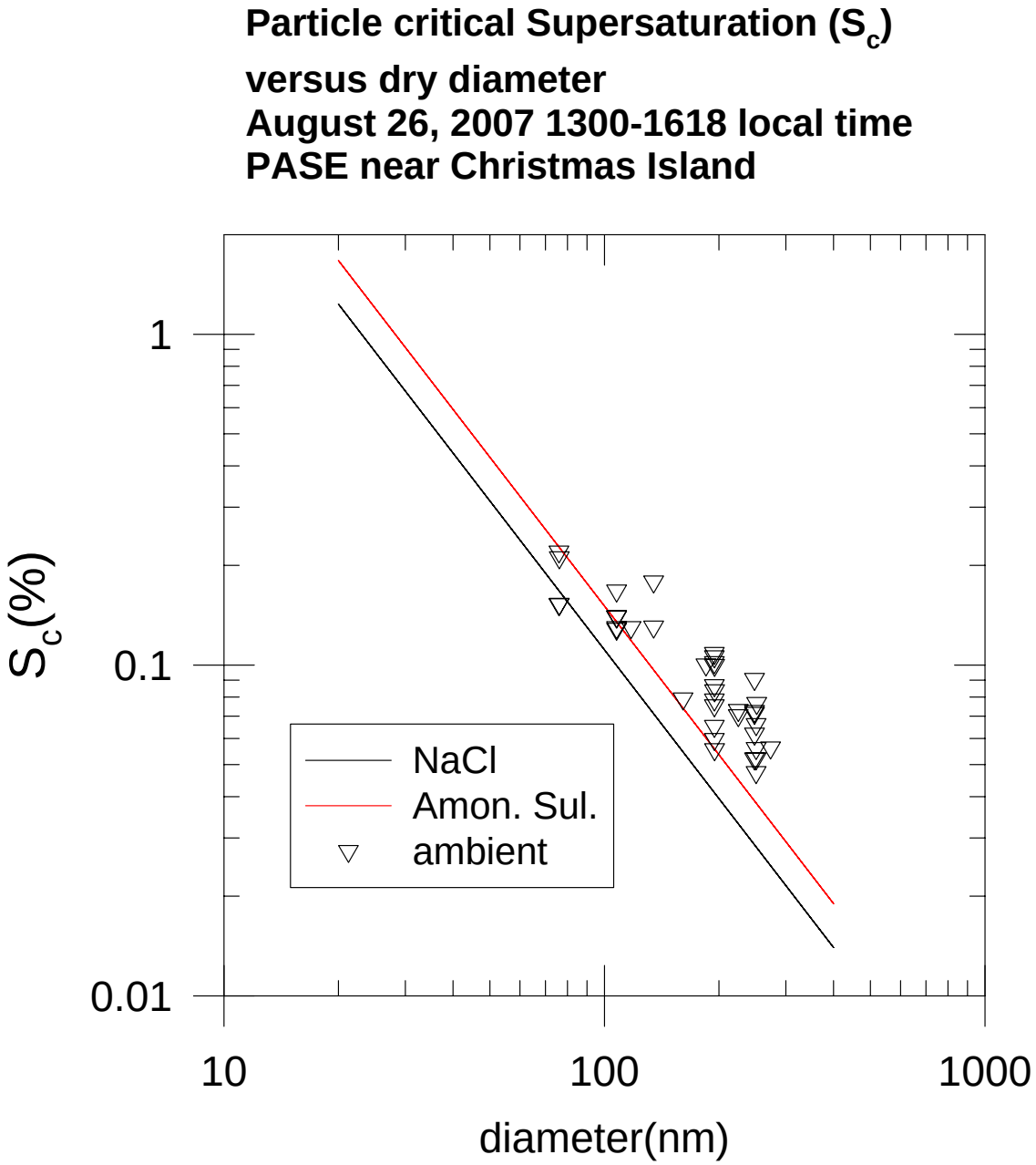


Figure 8. As Fig. 3 for a different flight.

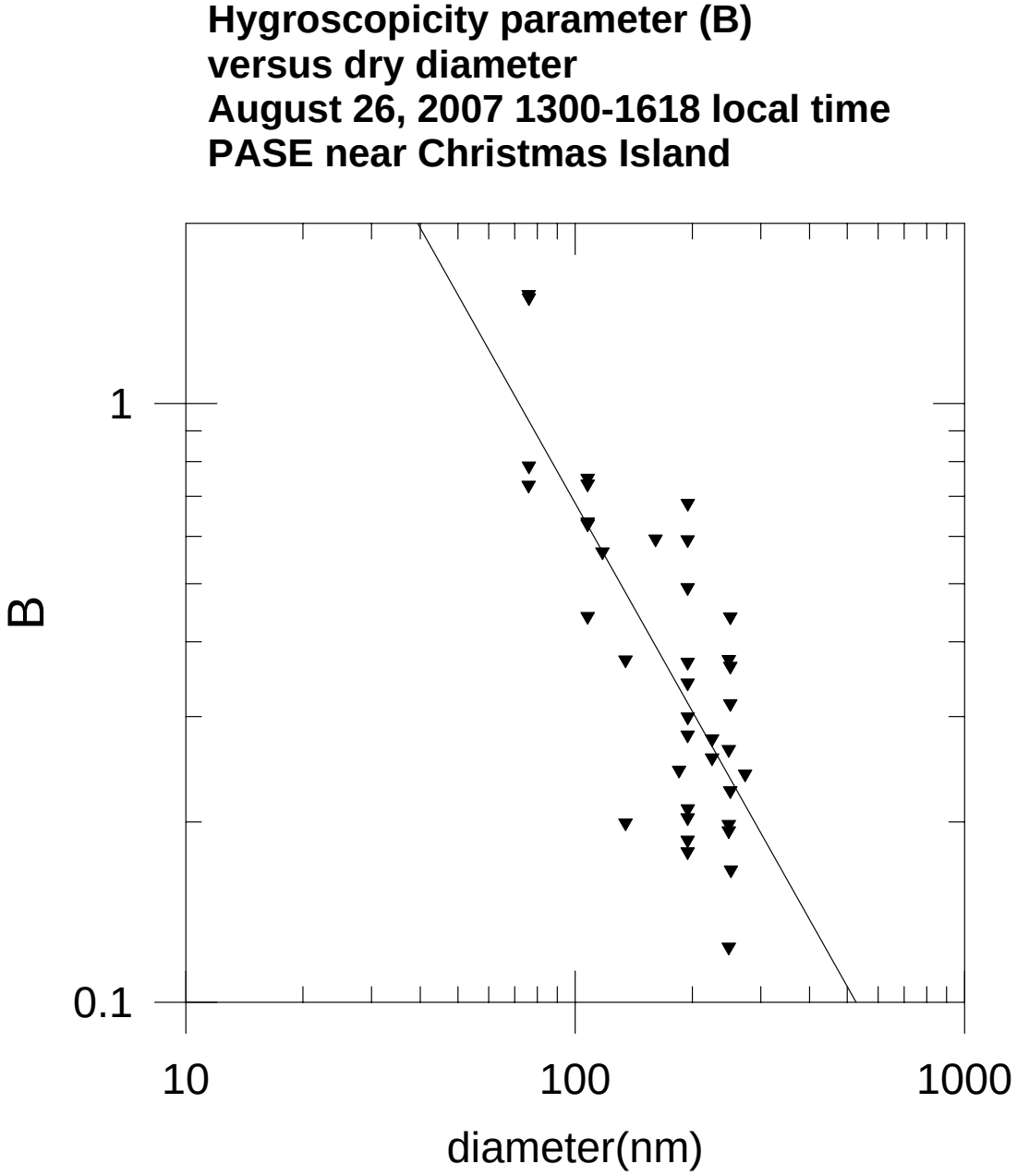


Figure 9. As Fig. 5 but for data displayed in Fig. 8.

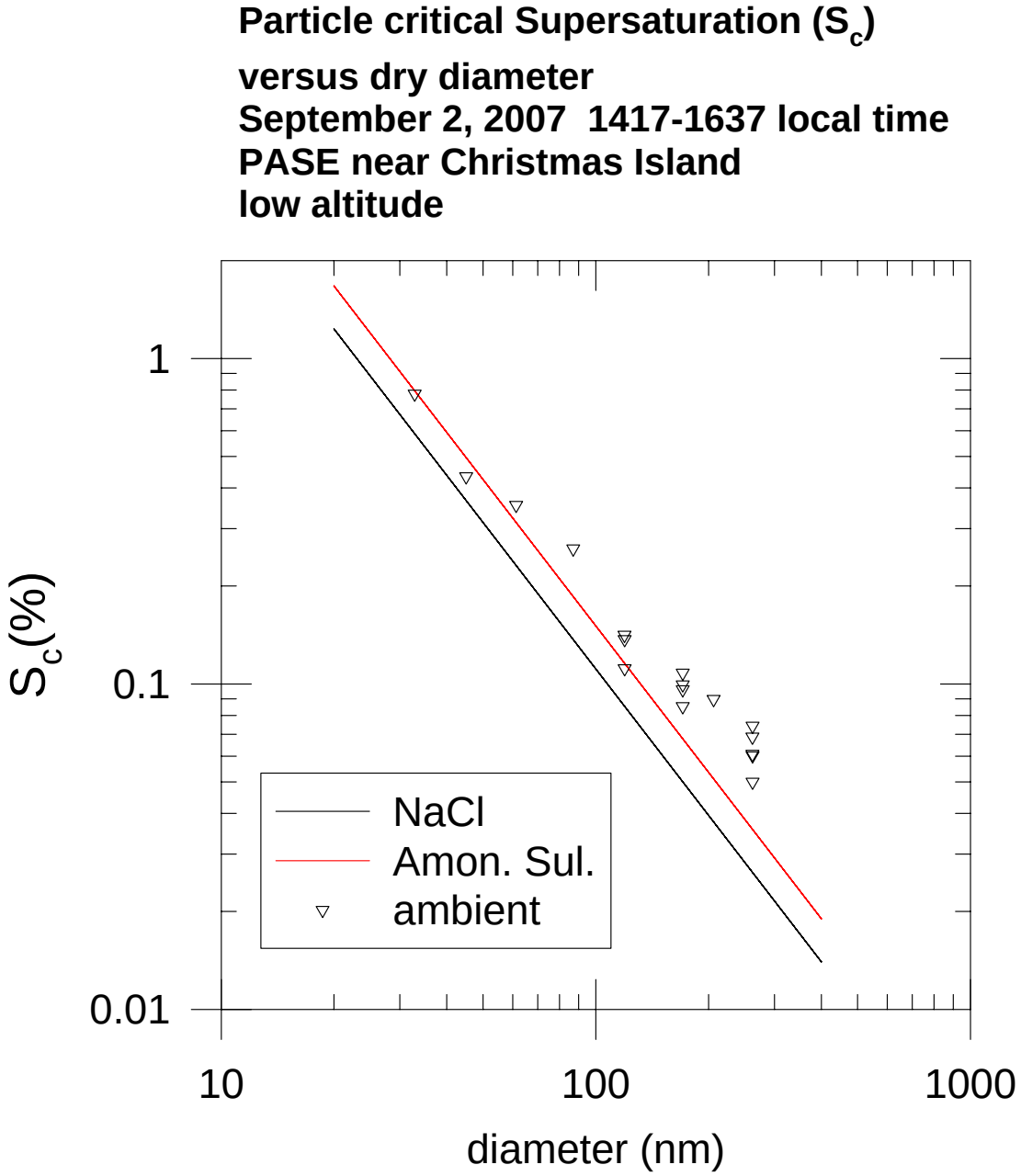


Figure 10. As Fig. 3 for a different flight.

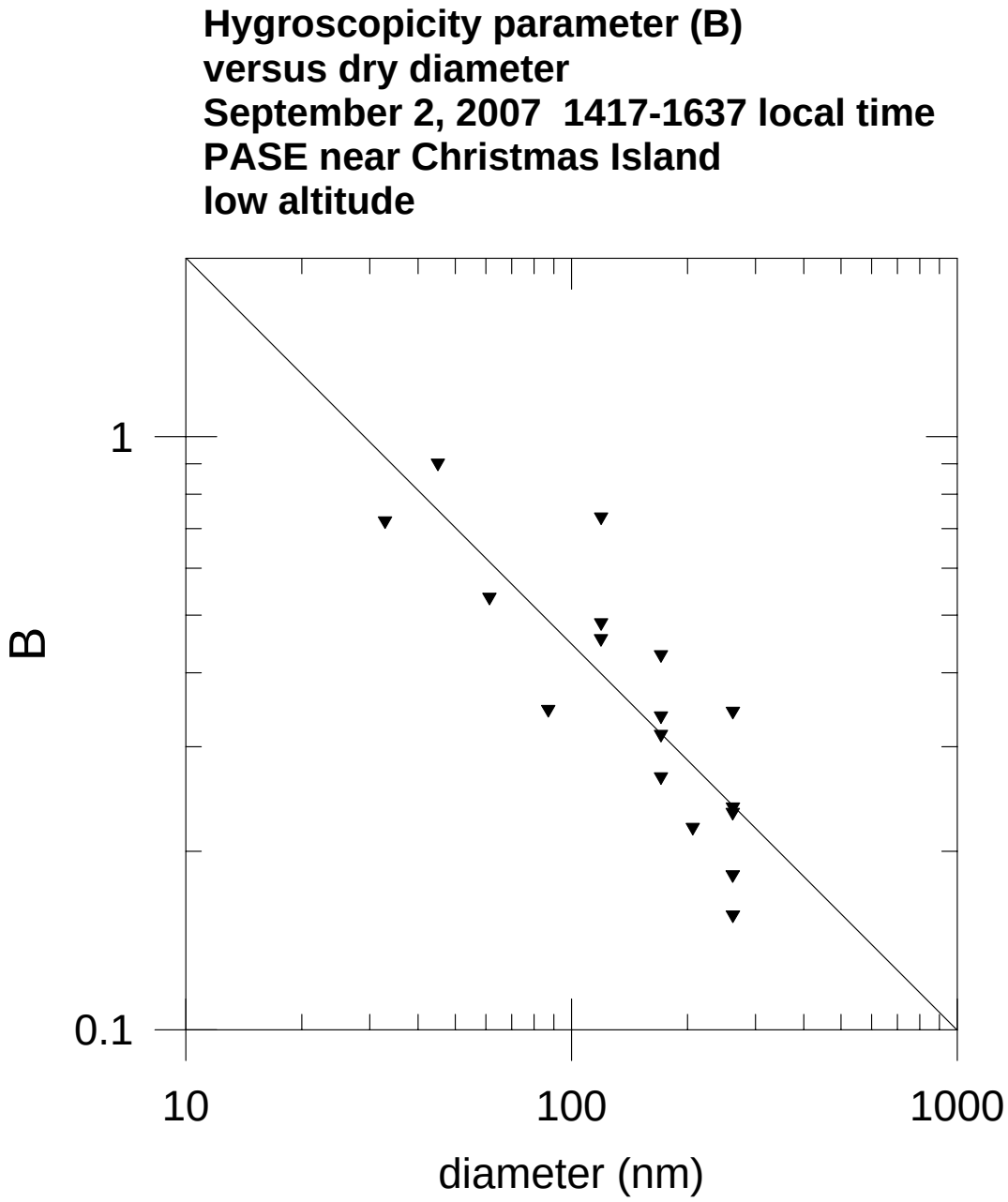


Figure 11. As Fig. 5 but for data displayed in Fig. 10.

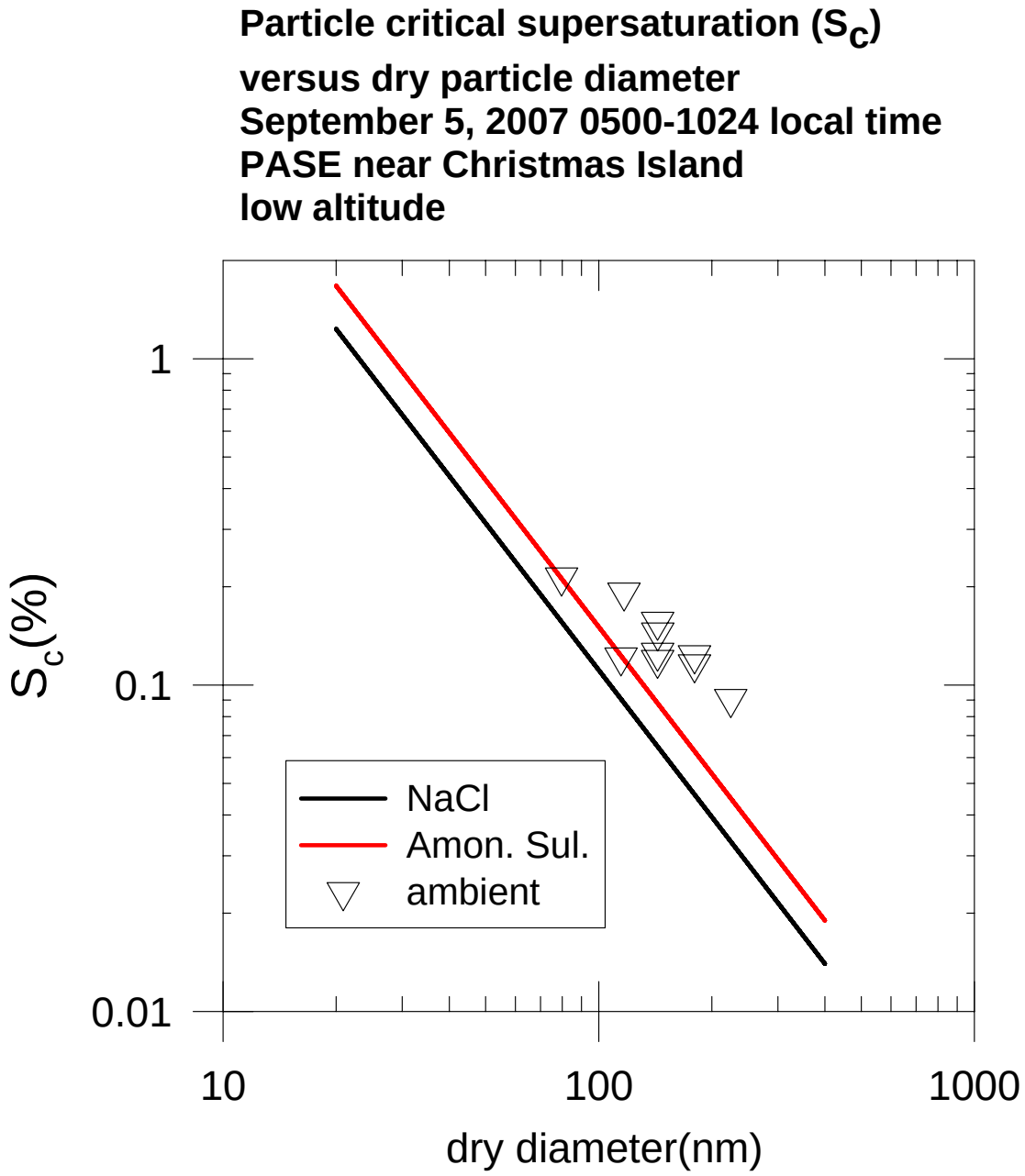


Figure 12. As Fig. 3 for a different flight.

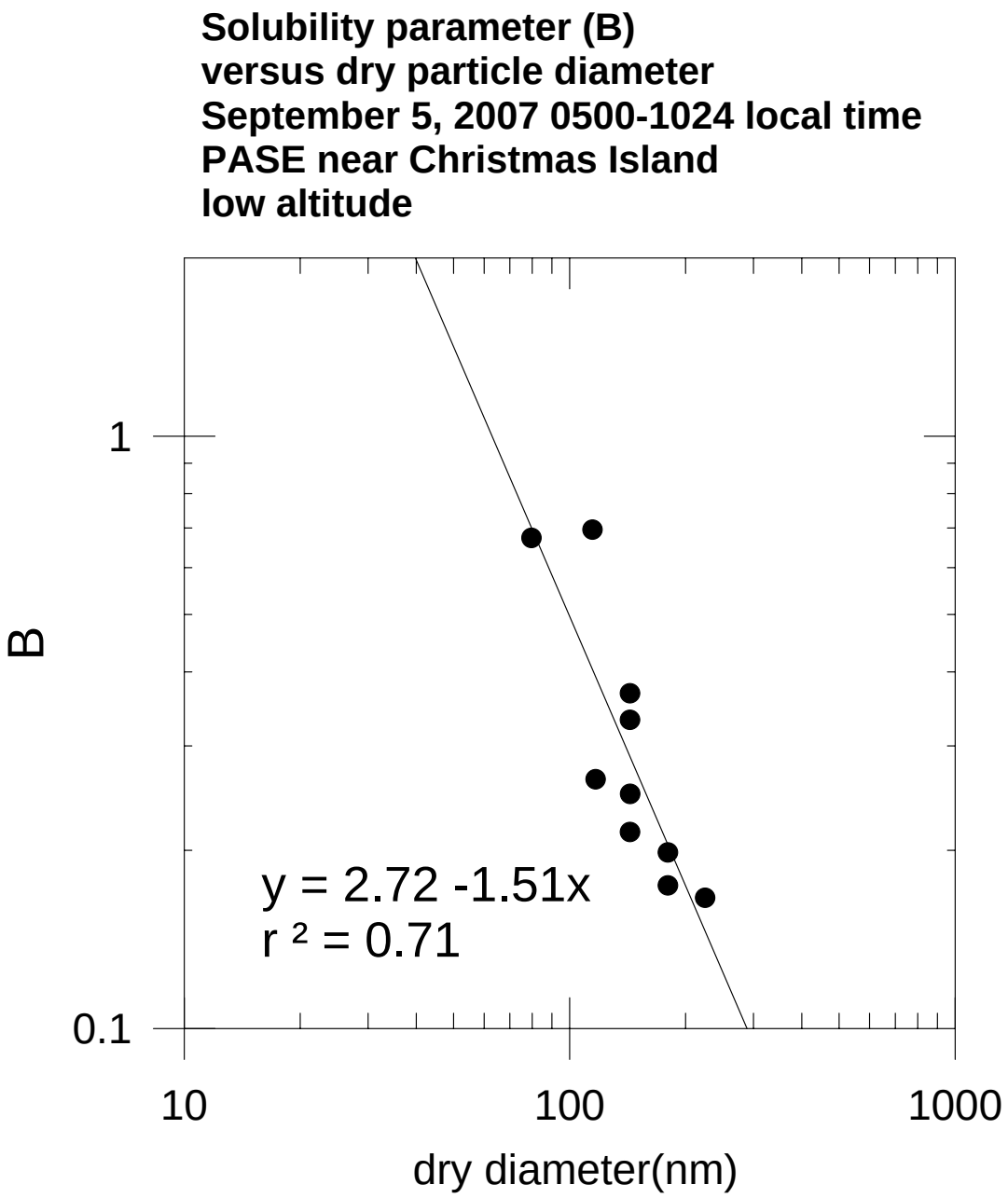


Figure 13. As Fig. 5 but for data displayed in Fig. 12.

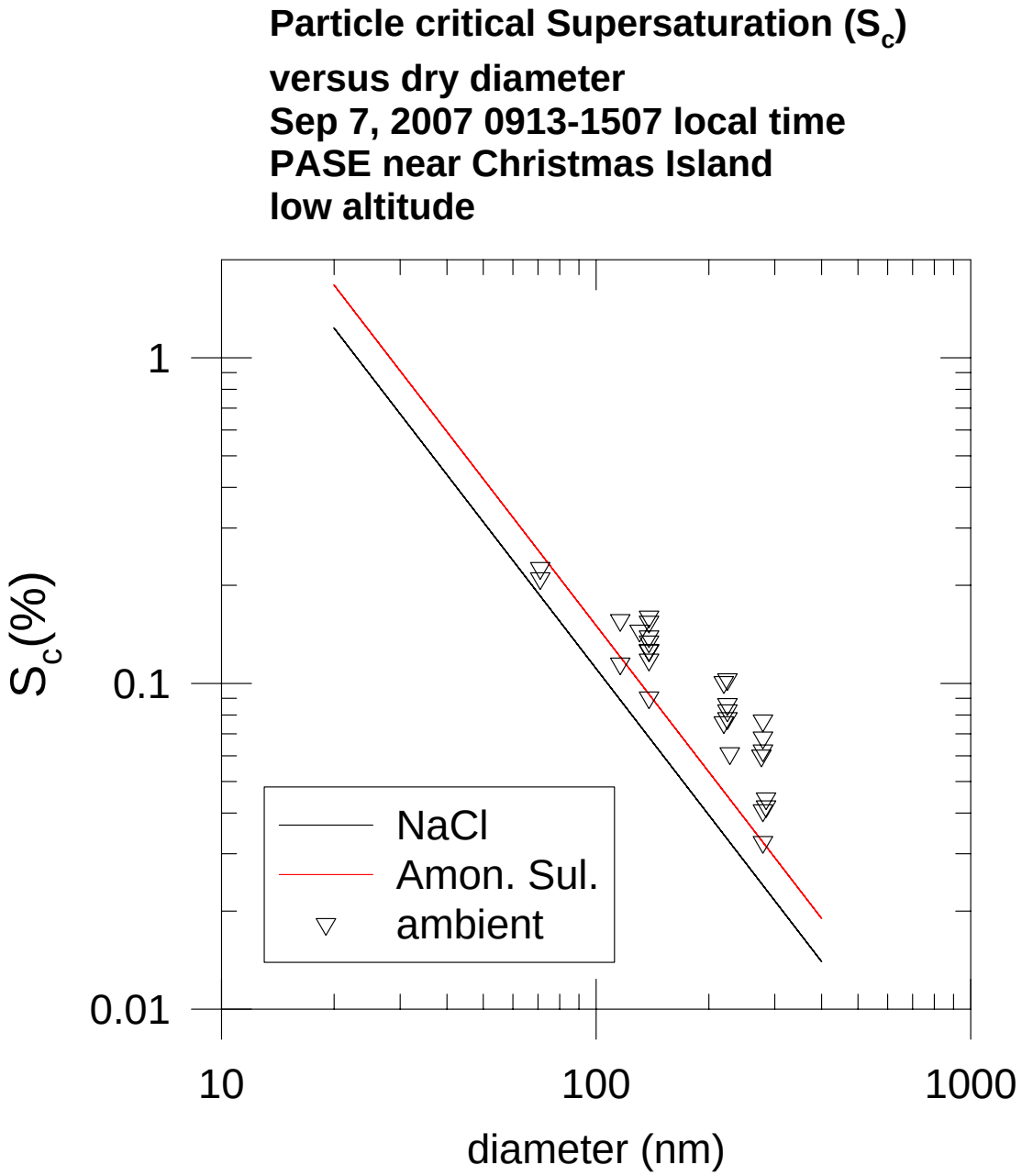


Figure 14. As Fig. 3 for a different flight.

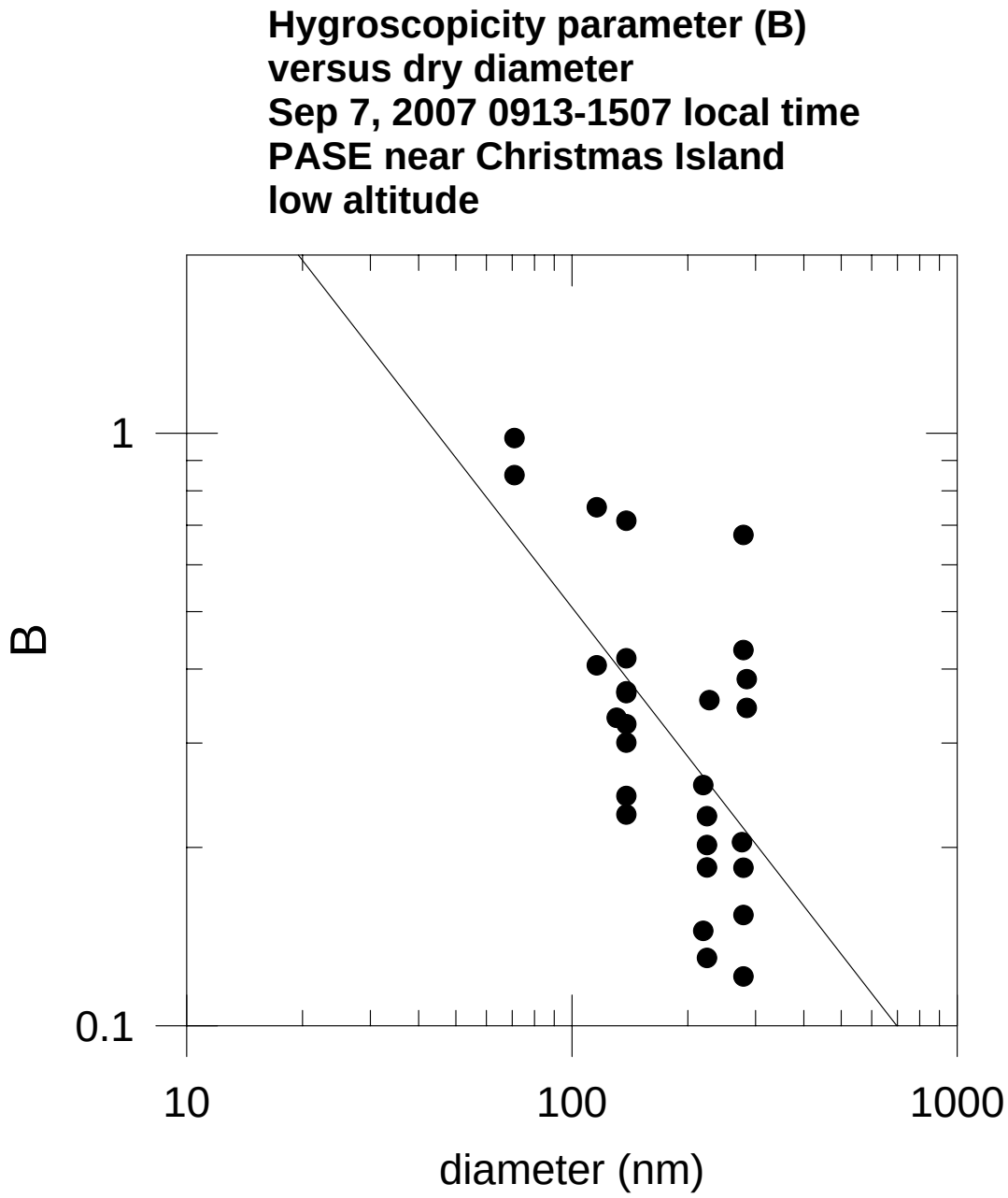


Figure 15. As Fig. 5 but for data displayed in Fig. 14.

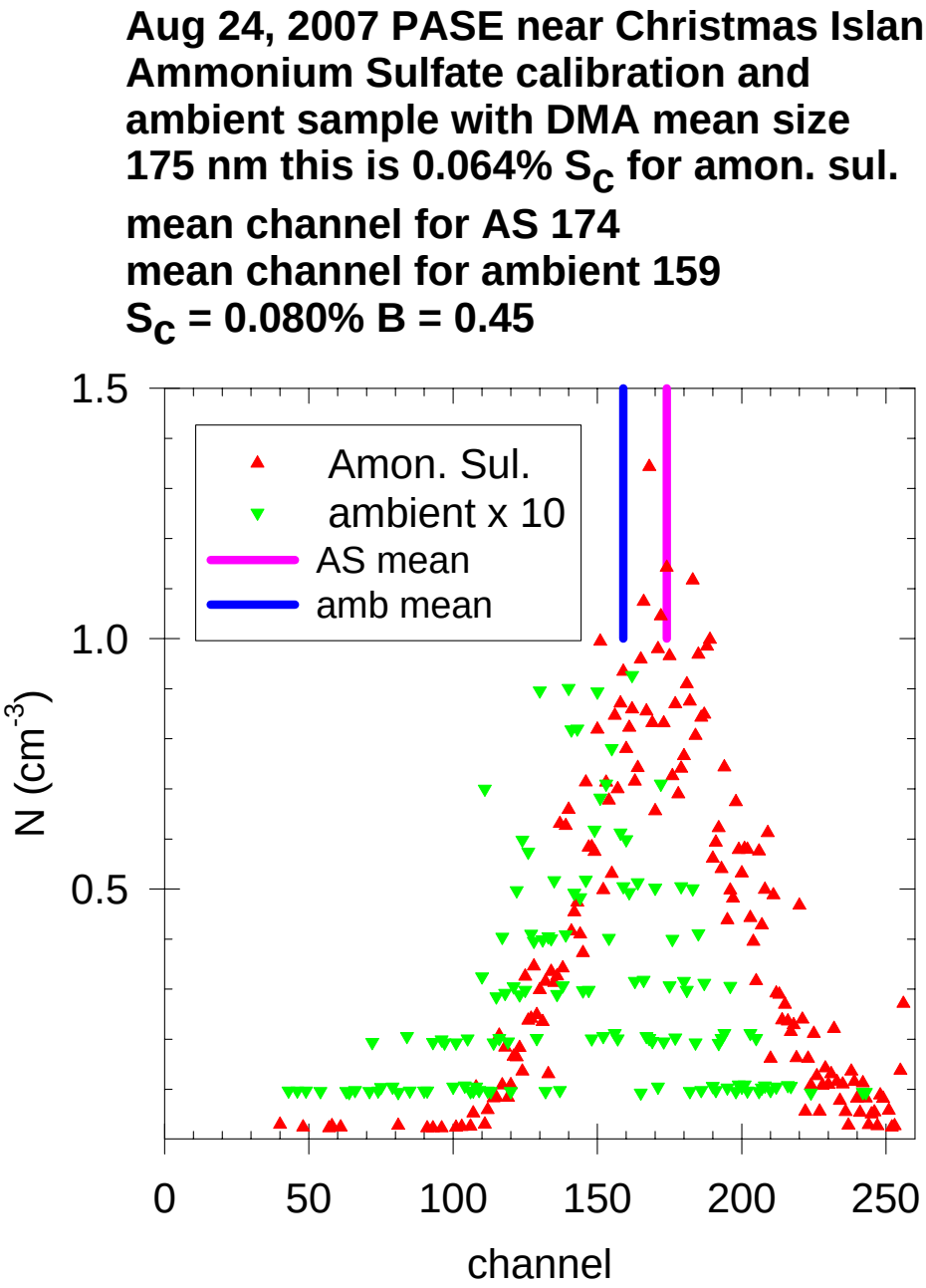


Figure 16. Plots of cloud chamber number concentrations versus channel number, which is related to droplet size, for ambient aerosol and ammonium sulfate calibration particles of the same sizes from the DMA.

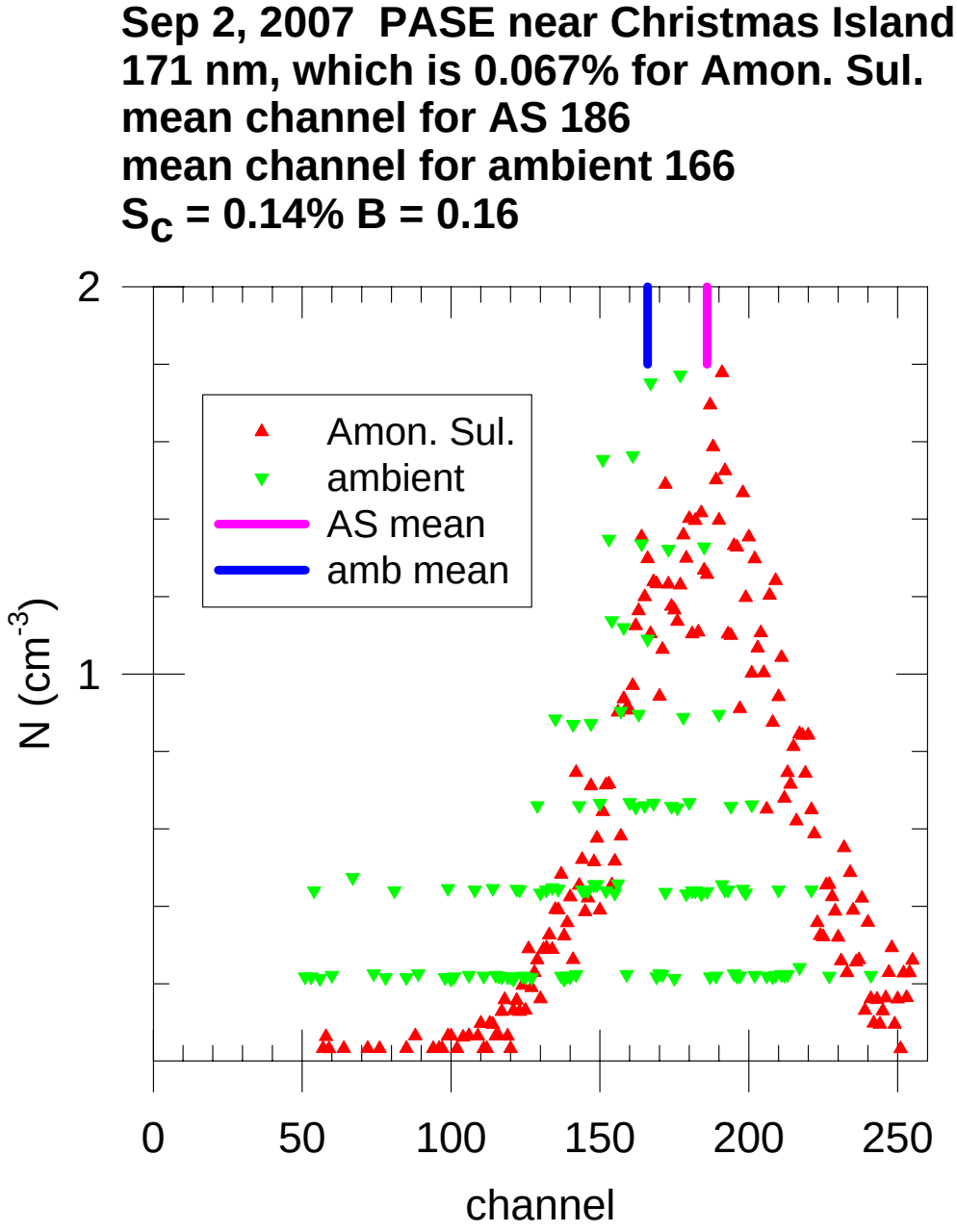


Figure 17. As Fig. 16 but for a different flight.

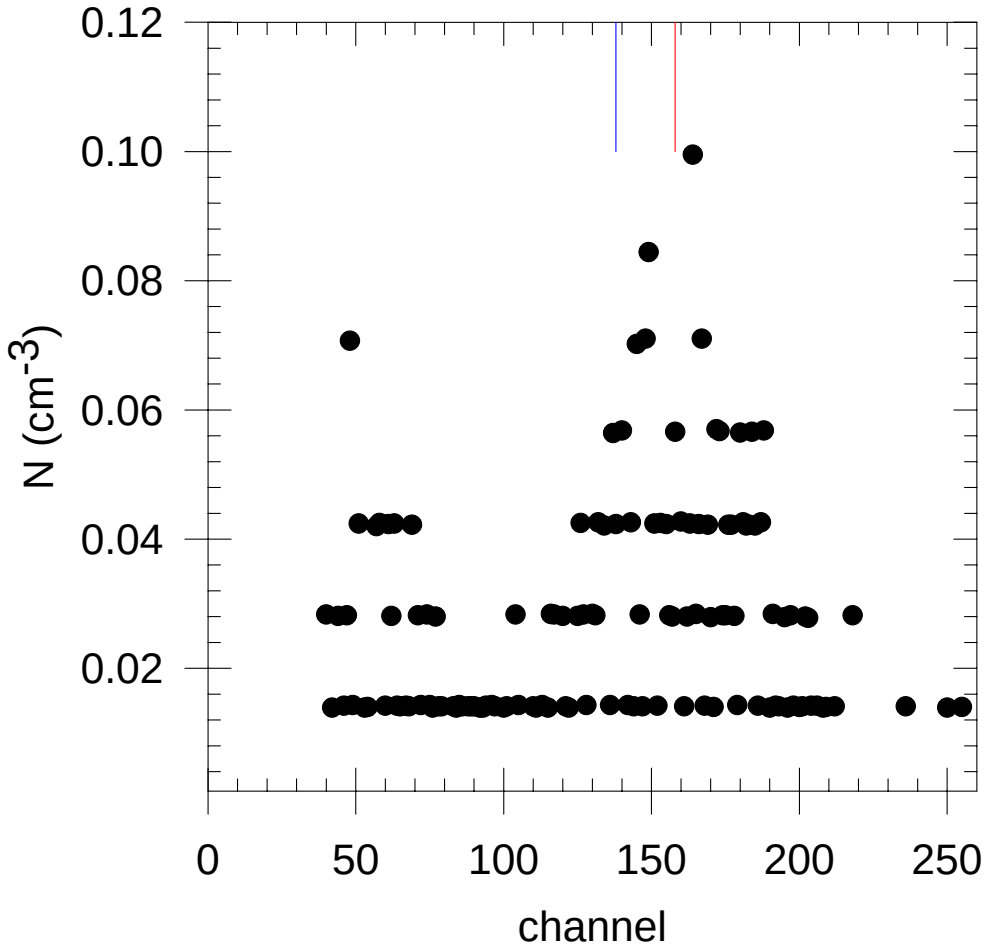


Figure 18. As Fig. 16 but for surface measurerments of a diesel plume and ambient aerosol showing a bimodal distribution.

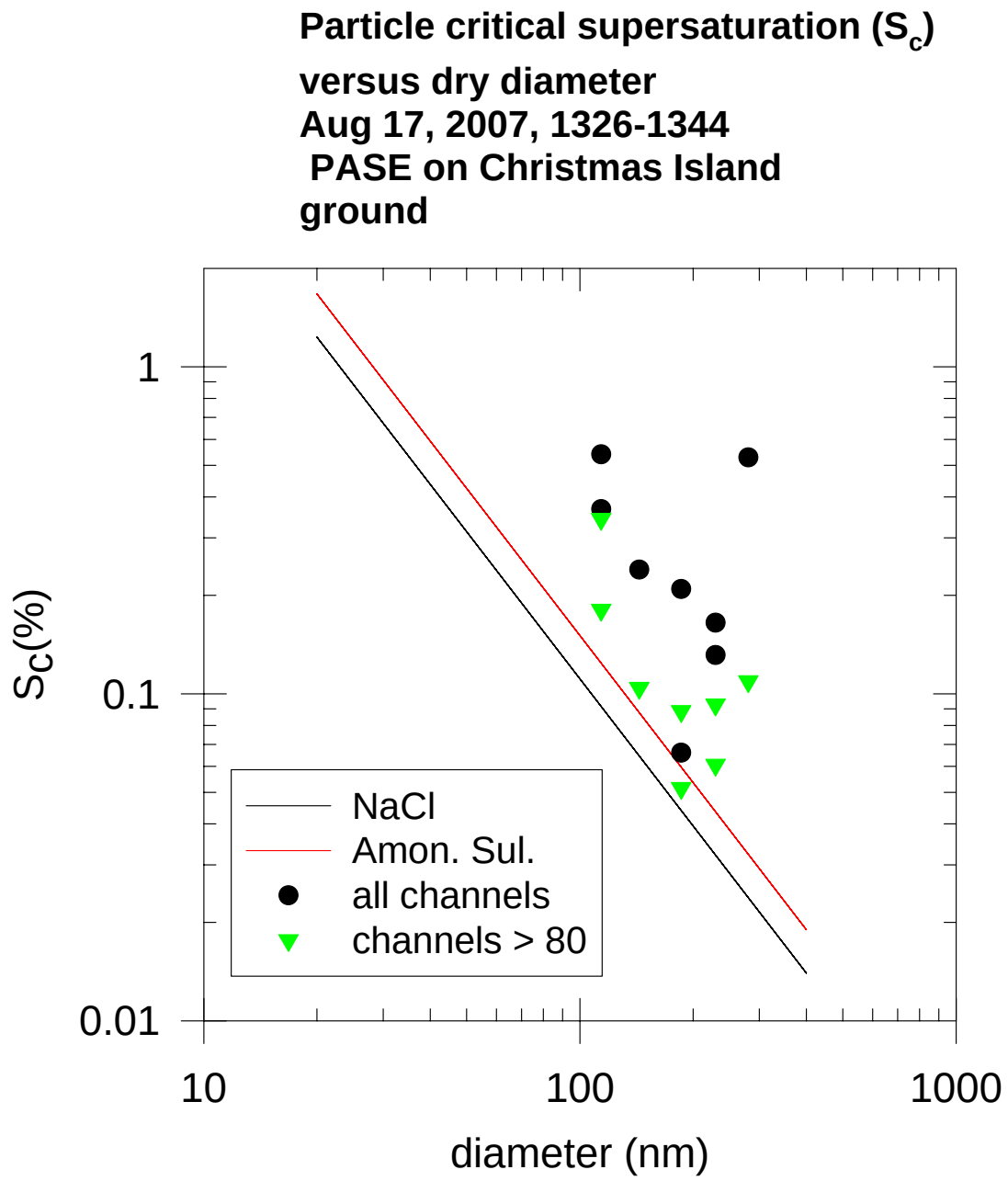


Figure 19. As Fig. 3 for the situation noted in Fig. 18. Considering different concentrations for different threshold channels, which result in different S_c.

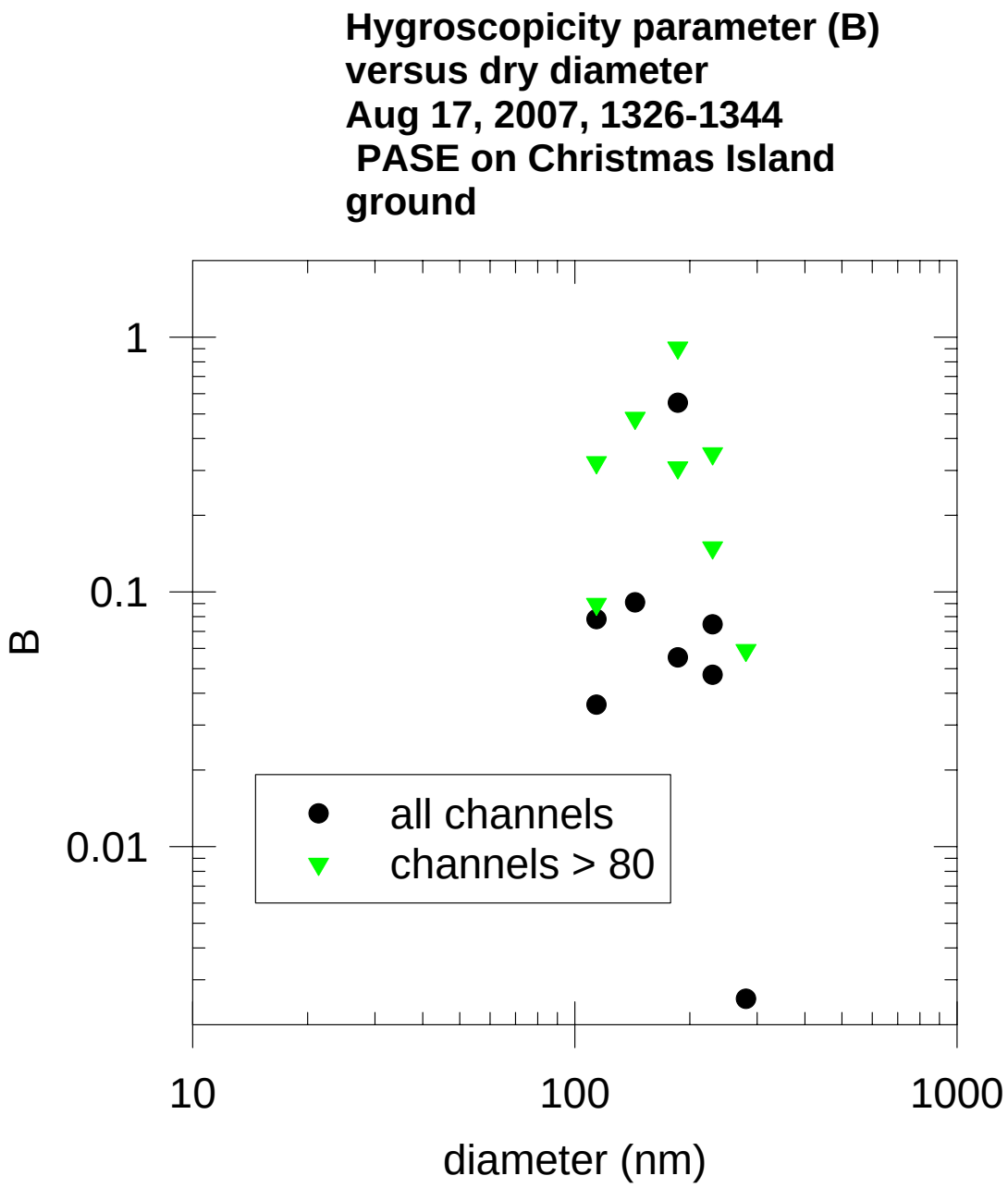


Figure 20. As Fig. 5 but for data displayed in Fig. 19.

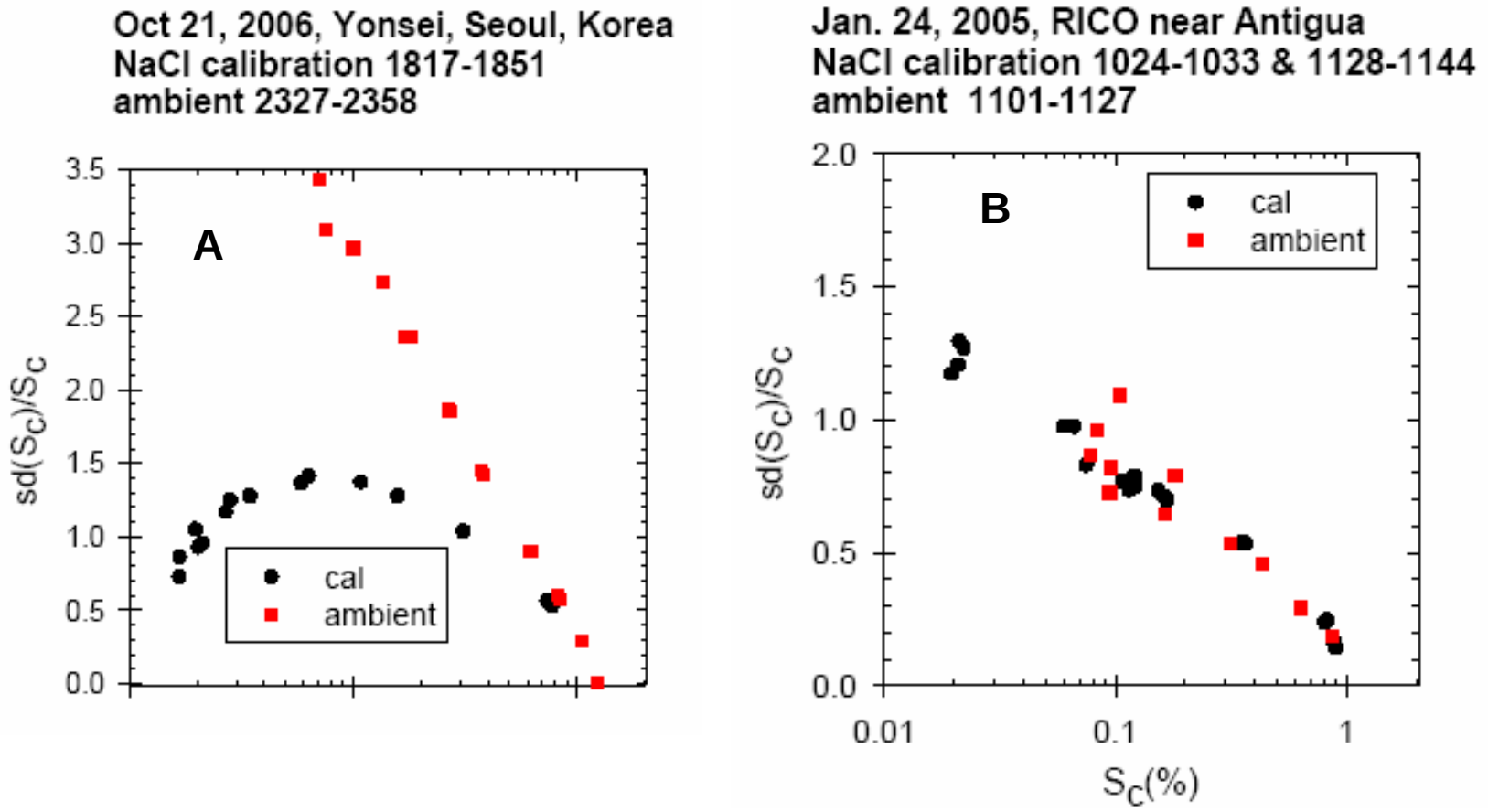


Figure 21. Relative variability (standard deviation; sd) of S_c for dirty air (panel A) and clean air (panel B). The ambient is much wider than the calibration in panel A and similar to the narrow calibration aerosol in panel B. This suggests the inadequacy of only using the mean values of S_c to express solubility (B).

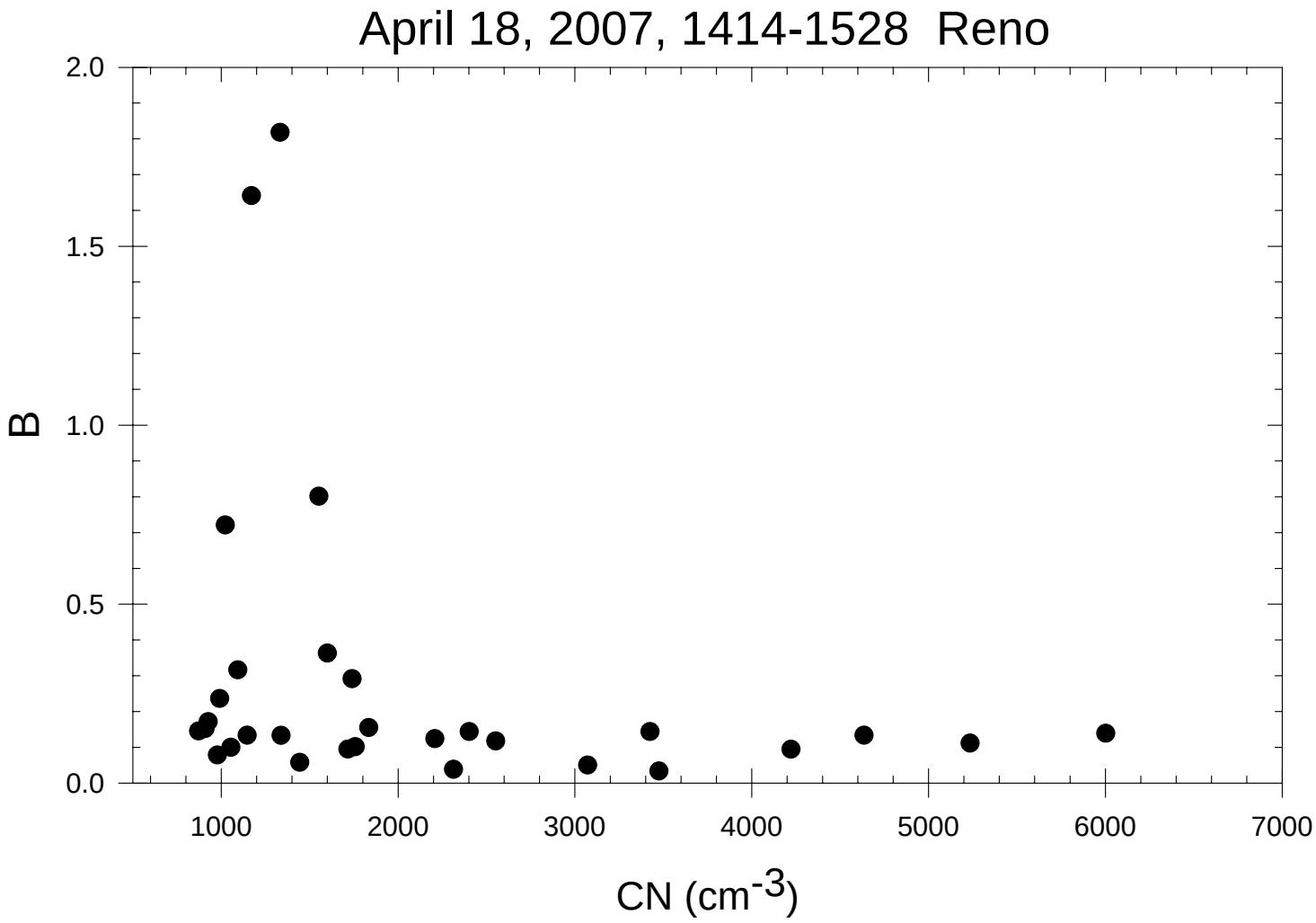


Figure 22. Particle solubility (B) from size versus Sc measurements lotted against total particle concentrations (CN). Notable is the great deal of variability of B for the same CN concentrations between 1000 and 1500 cm-3.