Referee #1

*The manuscript describes a study of new particle formation and growth up to CCN relevant sizes under a clean conditions dominated by biogenic SOA. The findings are presented in a clear and well structured way, and those are within the scope of ACP. Therefore, I recommend publication of this paper after the following comments and questions have been addressed.*

*Specific Comments*

*More than half of the introduction, starting from page 28504 line 1, is focused on the current study. I would recommend condensing and removing details such as describing Figs 1 and 2.*

We realize that it is not common to have two figures in the introduction. However, we feel that the introduction should motivate the questions to be addressed by the paper. We found it difficult to do this motivation without a somewhat detailed discussion of the event.

As a compromise, we have created a new section, "Aerosol event description", that includes the portion of the previous introduction that focused on the details of the WACS2010 study. It falls in between the new, more general, introduction and the methods section, so the text is largely not rearranged.

*It should be possible to have more evidence about the suspected under-counting of <30 nm particles by the Whistler peak SMPS (page 28518, line 12-). This is important for the current interpretations and possible future studies. The SMPS is for on-going measurements, so does it detect <30 nm particles during other time periods? How about long-term correlation with the total number concentration from the UCPC? Would it be possible to do calibration experiments using small particles?*

First, we have revised the SMPS data slightly due to some errors in the original processing. We apologize for this mistake, the outcome of which is to increase the number concentrations of some of the smaller particles. We have updated the size-distribution related figures (although there are only small noticeable differences), and our conclusions regarding survival probabilities and CCN-activity do not change. However, this has not changed the lack of particles smaller than 30 nm. In all cases, we operated the SMPSs using the lowest sample flow to improve the charge distribution. We include at the end of this slides with examples of the comparisons that have been done to demonstrate the quality level of the SMPS data as well as try to address the issue of the low concentrations of particles <30 nm.

In slide one is a summary of SMPS calibrations done in Sept 2011 using PSL particles of nominal sizes 100 nm and 340 nm. Using the appropriate mean free path for Whistler Peak, the PSL primary sizes peak at 109 nm and 346 nm in the SMPS. We can not guarantee the geometric size of the PSLs; it is possible that they are not truly the nominal values or that impurities (below) may have increased their size. From these comparisons, the error SMPS sizing at ≥100 nm is ≤10%. The mode centred near 30 nm that tails down to the lower limit of the SMPS settings (14 nm) is due to impurities in the atomization process. Despite rinsing the atomizer etc several times, the working environment at the site is not conducive to "ultraclean" particle generation. However, the presence of the impurities does demonstrate that particles are measured with the SMPS down to the 14 nm lower limit, but because the mode is centred near 30 nm we can not conclude that there is not some efficiency issue below 30 nm.

In slide two, the number concentrations measured with a 3025 UCPC (nominal detection limit of 3 nm), located in the main site at the Peak, and a 3775 CPC (nominal DL of 4 nm; this is separate from the 3775 CPC used with the SMPS), located in the container at the Peak. The 3025 malfunctioned around July 18, but during the overlap period of 18 days, the two concentrations are virtually indistinguishable, despite being on different inlets separated by about 10 metres.

In slide three, the SMPS total number concentrations are compared with the number

concentrations from the 3025. The two concentrations track each other well, although the 3025 concentrations are mostly higher.

In slide four, the total number concentrations from the SMPS are compared with those from the MSP-WSP (also a differential mobility system) operated in the container at the Peak. Again the covariance is good, but there some places where the MSP totals are significantly below the SMPS. However, the comparison of the totals of dN>100 nm measured with the SMPS and MSP are much closer (see slide five). The SMPS dN>100 nm at the Peak also compares well with the DMT-UHSAS measurement that was in the container (slide six).

In slide seven, the time series of the totals at the Nest and the Peak are compared and the fractions of particles <30 nm and <20 nm are also shown. It is clear that smaller particles were measured at the Nest compared with the Peak. Distributions from both sites (see a few examples in slide eight) show that while smaller particles were observed with the SMPS at the Nest site, there was still no indication of continuous growth from 10-20 nm.

In summary, we find that the SMPS compared well with the other measurements, but we can not rule out an instrumental explanation for the absence of particles <20 nm in the SMPS measurement (e.g. volatilization of these small particles within the SMPS during sampling) or 2) at the Peak site especially there was insufficient oxidized precursor material to grow newly nucleated particles and the numbers nucleated were insufficient for self-coagulation to produce 10-20 nm particles.

Finally, a scanning of the full 2010-2011 timeseries of SMPS measurements at Whistler Peak shows that particles do not ever show up with significant concentrations below 20 nm. This however does not let us determine if this is an instrument issue or if particles always have grown to sizes larger than 20 nm before reaching Whistler Peak.

*Are the new (e.g. 30 nm) particles observed simultaneously at the Peak and the Raven's Nest? Even a relatively small time difference, or lack of that, could tell some- thing about the origin of these particles.*

Response is included in that to the previous comment.

*As described in the Sect. 3.3 (page 28520), it is possible to calculate critical diameters and overall hygroscopicity parameters (κ) from the SMPS size distributions and CCN concentrations. Why was this not done?*

This was done and is included. For 0.1% supersaturation: "...the critical diameter for 0.1% supersaturation must correspond to a diameter very close to 240 nm. This critical diameter at 0.1% supersaturation corresponds to an overall hygroscipicity factor (κ (Petters and Kreidenweis, 2007)) of about 0.11." For 0.2% supersaturation: "Similarly, the CN150 and CCN(0.2%) track each other closely during this same period, also corresponding to an overall κ of 0.11". Since we were not explicit here about the critical diameter, we have changed this to, "Similarly, the CN150 and CCN(0.2%) track each other closely during this same period, which means the critical diameter for 0.2% supersaturation is about 150 nm. This also corresponds to an overall κ of 0.11". The same analysis is done for 0.3% supersaturation, but involved more discussion due to the κ value changing in the middle of the event (see original manuscript).

*The Raven's Nest SMPS detects particles smaller than 685 nm, but there could be larger particles having an effect on CCN concentrations. Are these seen at the Peak where size distributions are measured up to 10 μm?*

The concentration of particles larger than 650 nm at the peak was less than 1 $cm^{-3}$ throughout the event. We have added the following text: "However, the MSP instrument at the Peak measured concentrations lower than 1 $cm^{-3}$ for particles with sizes between 650 nm and 10 μm during this time period. Unless

there were significantly more large particles at Raven's Nest than the Peak, the low bias in the SMPS concentrations was small."

*Why does Fig. 11 shows signals instead of mass concentrations? Is the sulfate signal larger than noise? If the highest sulfate concentrations are larger than the noise, what about 100 nm and smaller particles; is it possible to detect concentration changes in 100 nm particles (page 28521, lines 4-6)?*

The signal is proportional to the mass but must be normalized to the mass determined in the MS mode of operation in order to be quantitatively comparable. To avoid uncertainties associated with this normalization and to remain a qualitative analysis, signal units were used.

We agree with the reviewer that it may be difficult to detect concentration changes in sub-100 nm particles. During the study we performed daily 15-30 min particle filtered measurements. From these filtered measurements we estimate that the average deviation in the PToF signal across the size distribution is approximately 0.0019 Hz and have added this to the Fig 11 caption. This implies that we are able to reliably detect concentration changes for particles in the range of approximately 100 - 600 nm (vacuum aerodynamic diameter, though this should be shifted to somewhat smaller sizes for geometric diameters) and that the SO4 signal change observed in Fig 11 between 100 and 300 nm is valid.

The Figure 11 caption now reads: "Size-resolved sulfate and organic signal from the AMS at Raven's Nest for the morning of July 7 and the evening of July 8. The x-axis represents geometric diameter (converted from vacuum aerodynamic diameter assuming a density of 1300 kg $m^{-3}$). The average deviation in the PToF signal across the size distribution is approximately 0.0019 Hz, which implies that changes in organics throughout the entire size range and sulfate from 100-600 nm may be reliably detected (at least qualitatively). Organics have increased in size and in total mass. The total mass of sulfate has remained essentially constant, but it has grown to larger sizes due to condensation of organics. While there was sulfate signal for 100-150 nm particles above the detection limit during the early time, there was no significant sulfate signal in this size range during the later time. The timing of this shift in sulfate sizes corresponds to the lowering of kappa in the 100-nm particles."

*In the instrument description section (page 28507, line 14) it is mentioned that the AMS detects 100–700 nm (vacuum aerodynamic diameter) particles, but why is the size range wider in Fig. 11?*

We have added text to be more specific about the meaning of 100-700 nm, "these values correspond to ~50% cut points based on Liu et al. (2010), but quantitative measurements are made outside of this range."

*The conclusions based on Fig. 12 are not clear (page 28521, line 7-). The AMS detects 100–700 nm particle mass, so it does not tell much about the composition (hygroscopicity) of 100 nm particles.*

We agree that the mass as measured by the AMS is dominated by particles greater than 100 nm. The Org44/total ratio (and mz43) in Fig 12 implies that the CCN activity reduction in 100 nm particles is due to decreases in oxygenated components if the organic composition is similar across the size distribution. Analysis of PToF data for m/z 44 and m/z 43 indicate that that the 44/43 is approximately similar from 100-600 nm. We have added a clarifying point in the text.

*It could be possible see the changes in organic oxidation state by plotting size-resolved organic m/z 43 and 44 signals.*

*Technical corrections*

*Page 28506, line 25: from 15 to 10 mm?*

Fixed. Changed to µm

*Page 28507, line 25: Wrong year for the Roberts and Nenes paper?*

Fixed. Changed to 2005.

*Page 28526, line 4: m missing from microphysical*

Fixed.

*Page 28535, Fig.3 caption: Petjata et al. (2009)?*

Fixed. Changed to Petäjä.

Referee #2

*This manuscript describes a case study of aerosol and CCN formation from mainsly biogenic precursors at a pristine site in Whistler, Canada. The study is carefully made and the results are presented i n a clear and comprehensive manner. This is a nice, although relatively detailed, case study and fits well within the scope ACP. I therefore recomment publication in ACP after the following concerns have been addressed.*

*General comments:*

*1. I agree with the first reviewer that the comparability of the two SMPS systems deserves considerably more discussion in the paper. It is well known that as the particle size gets smaller, the charging probabilities before size classification and counting efficiencies in the CPCs also get lower, and considerably more uncertain. Have the instrumentation used in the different locations been quantitatively compared with well-defined aerosol populations? Have the detection efficiency curves of the individual CPCs been compared and calibrated with the same aerosol subject to similar losses? How about the DMAs - have they been compared?*

Included in the response to reviewer 1.

*It is problematic to try to get information on the free tropospheric influence on particle formation from the differences between the intruments - before their agreement using the same aerosol population has beenh demonstrated.*

*2. It is naturally a little disappointing that it is practically impossible to confirm the role of vertical transport and boundary layer meteorology in the observed particle formation based on the available data. I think it is in line with some previous observations from the Finnish site in Hyytiälä (e.g. Nilsson et al. 2001; or Lauros et al. Atmos. Chem. Phys., 11, 5591–5601, 2011). It would be nice if the authors could add a little discussion on what kind of experimental data and data analysis tools would be needed to potentially solve this problem. I assume it would have to do on things like e.g. the time resolution of the instruments as compared with the time scale of vertical mixing. Anyway, some guidance for future studies would be nice.*

This is a good idea. We have added the following text: “Although we had measurements at two elevations on Whistler mountain, we did not notice a difference in the timing of the onset of the

particles between the two locations (to within ~10 minutes), which would have given clues to the source location. This negligible difference is likely due to fast mixing and upslope flows during the daytime boundary layer. Future studies would require particle flux measurements (e.g. Nilsson et al., 2001), balloon-borne CPC measurements (e.g. Laakso et al., 2007; Lauros et al., 2011) or aircraft measurements (e.g. O'Dowd et al., 2009) to gain more insight into the nature of the new particles."

*3. I think that Fig. 1 deserves some more discussion. The good agreement between temperature and OA concentration practically implies that the OA concentration is limited rather by VOC emissions than their condensation (or at least the thermodynamics of the net condensation), right? I think this might link nicely to the topic touched upon in the conclusion section - namely the biogenic influence on aerosol loadings and what kind of temperature response one might expect from organic aerosol loadings.*

The stronger temperature dependence of SOA on VOC emissions than thermodynamic partitioning has been studied in detail at Whistler and another Canadian Location in a recent paper by Richard Leaitch (Leaitch, W. R., et al.: Temperature response of the submicron organic aerosol from temperate forests, Atmos. Env., 45, 6696-6704, 2011.). We have added the following discussion to the introduction. "The effect of temperature on the SOA-precursor emissions (increasing emissions with increasing temperature) is stronger than the thermodynamic effect of temperature on SOA partitioning (decreasing SOA with increasing temperature). Thus, temperature and SOA are positively correlated, consistent with Leaitch et al., (2011), which explored the the temperature dependence of SOA at Whistler and Egbert (another forested Canadian location)."

*Specific comments:*

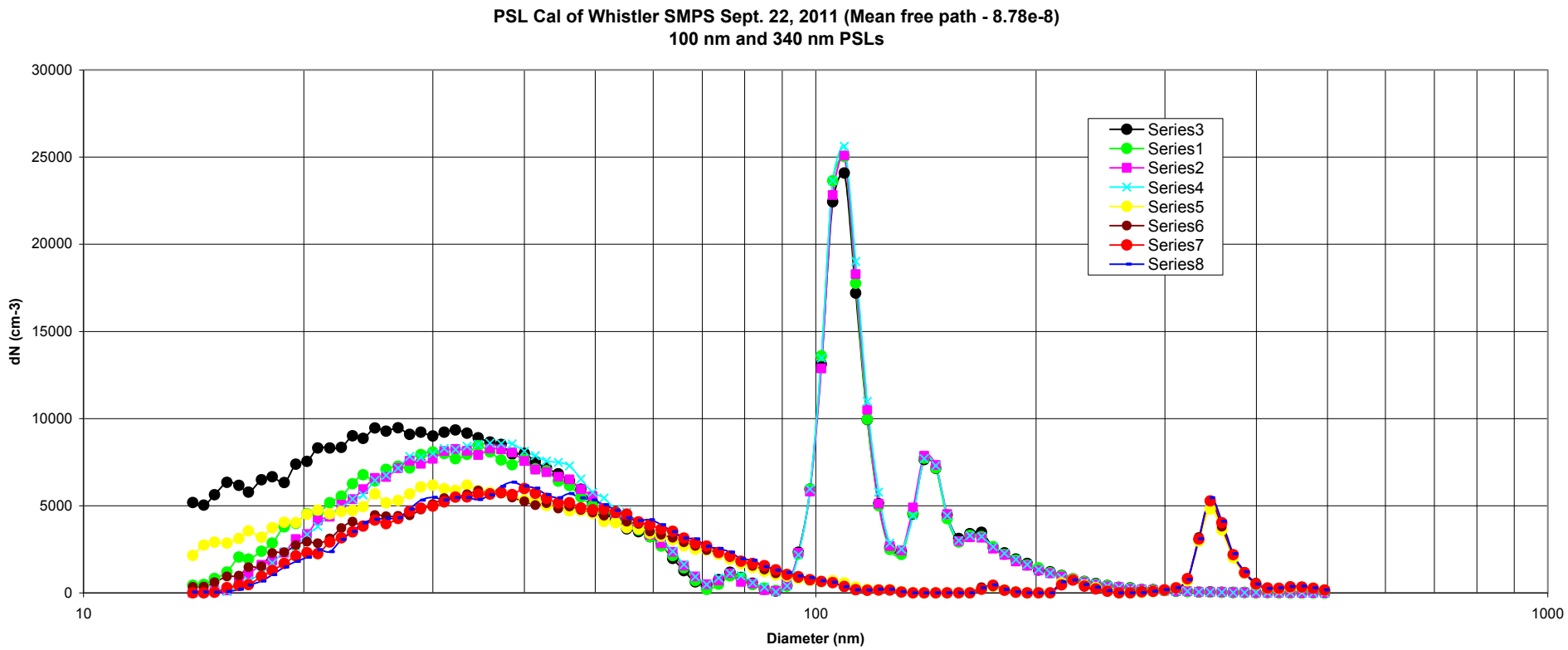
*4. p. 28506, line 25: I assume "15 to 10 mm in diameter" should be "15 nm to 10 um in diameter"*

Fixed.

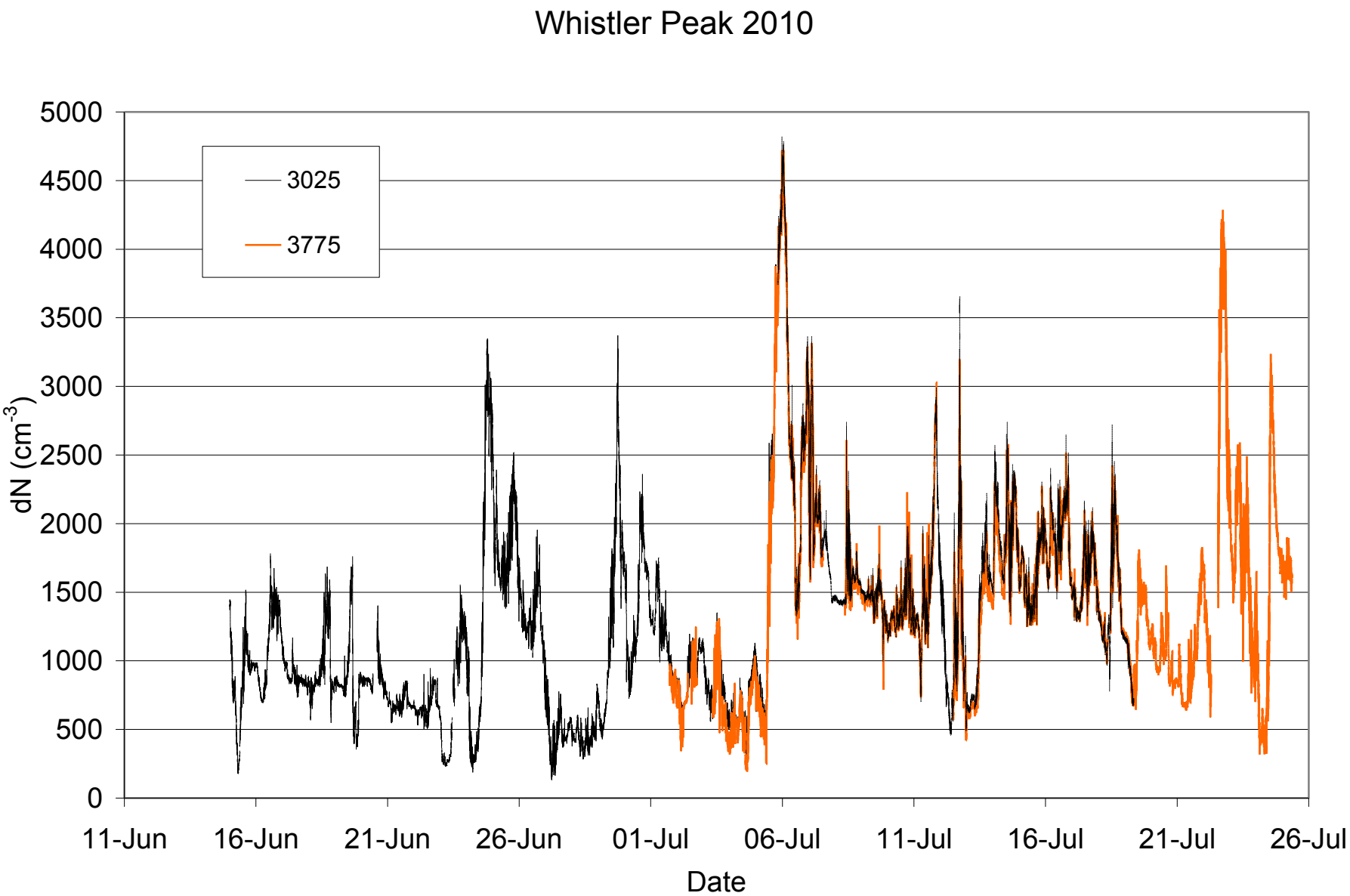
*5. p. 28535, line 3 from above in the caption for Fig. 3: "Petjata" should be "Petäjä"*

Fixed.

# PSL calibrations at Whistler Peak

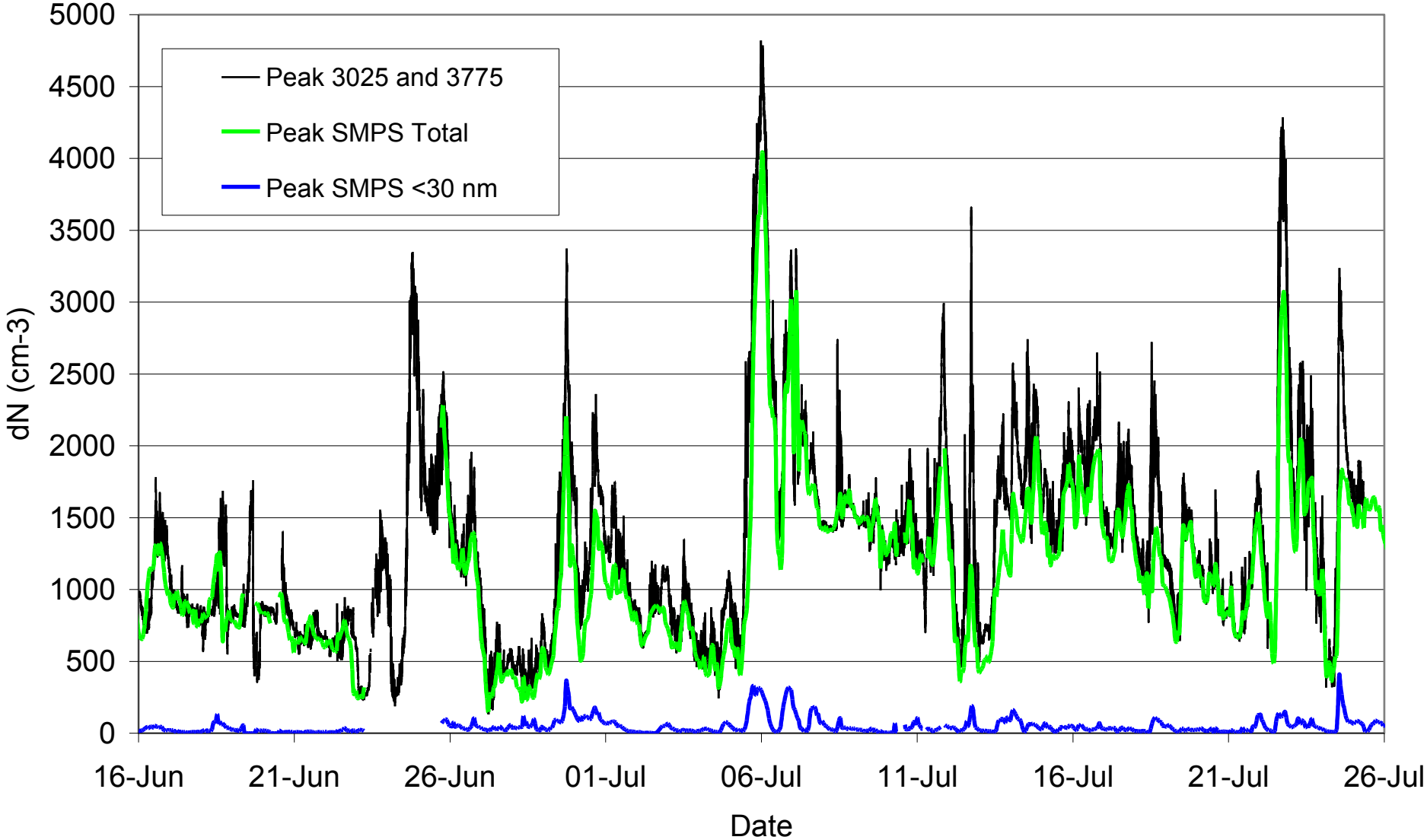


# CPC comparison

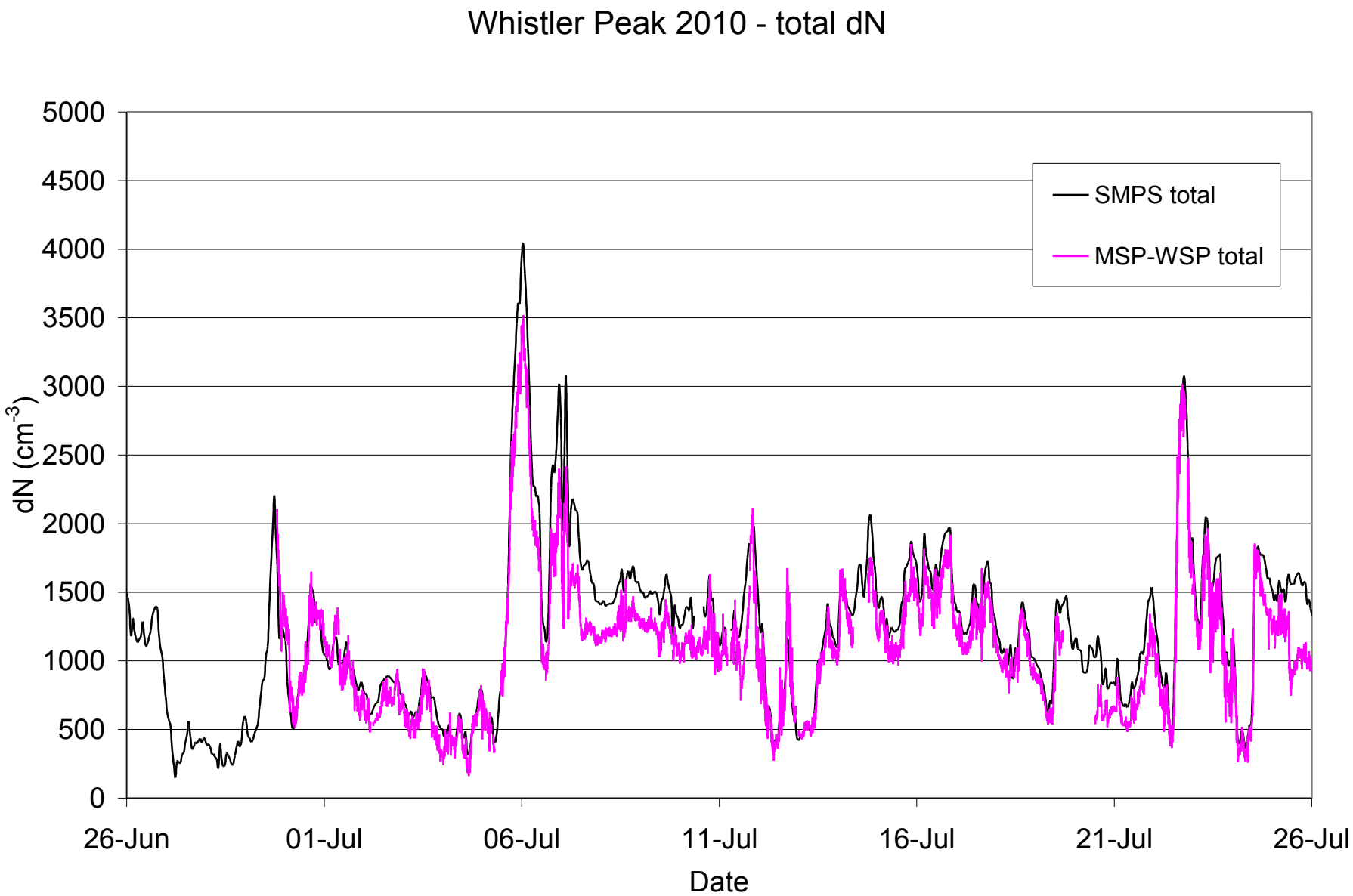


# CPC (3025) and SMPS total number comparison

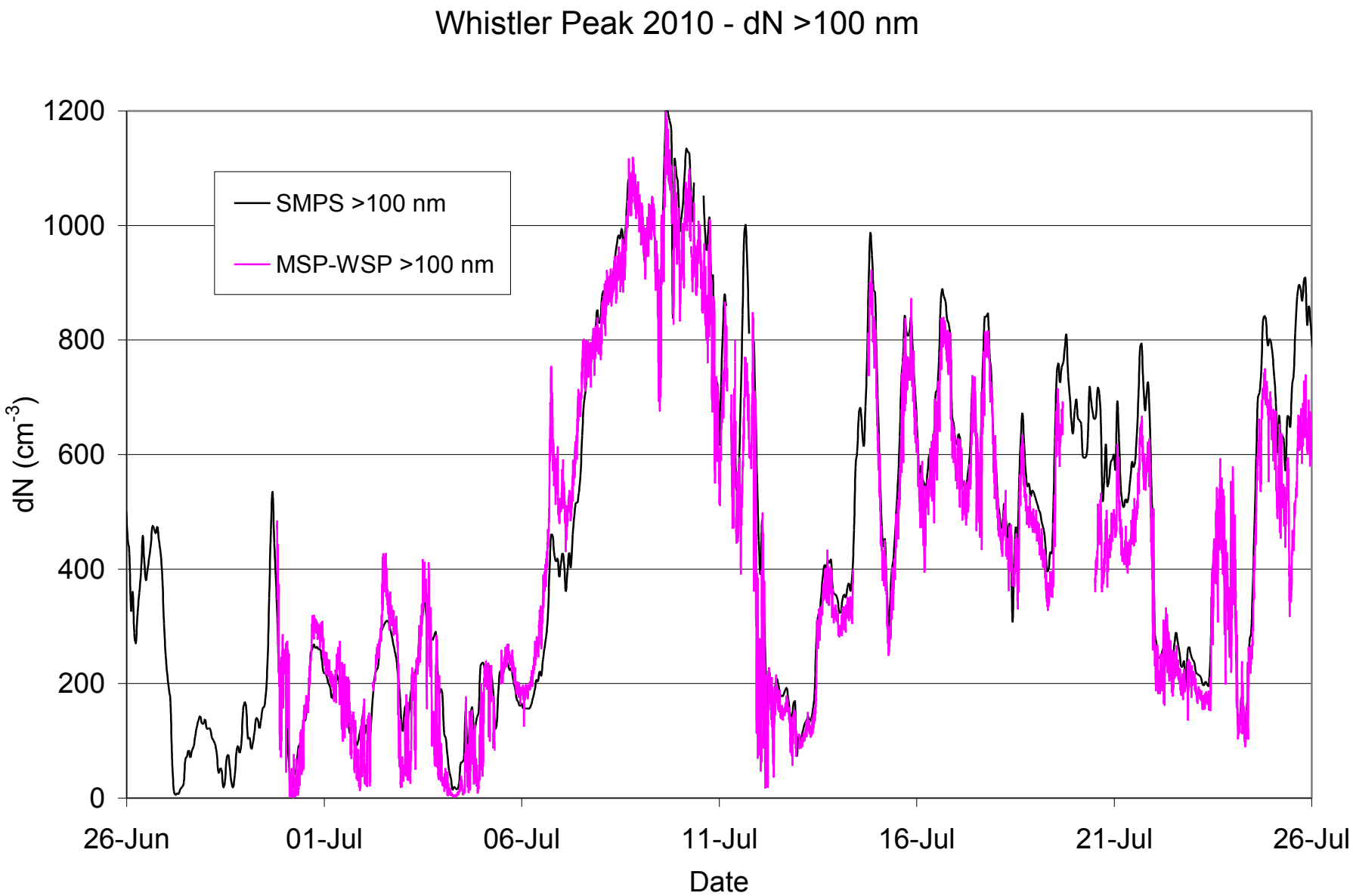
Whistler 2010

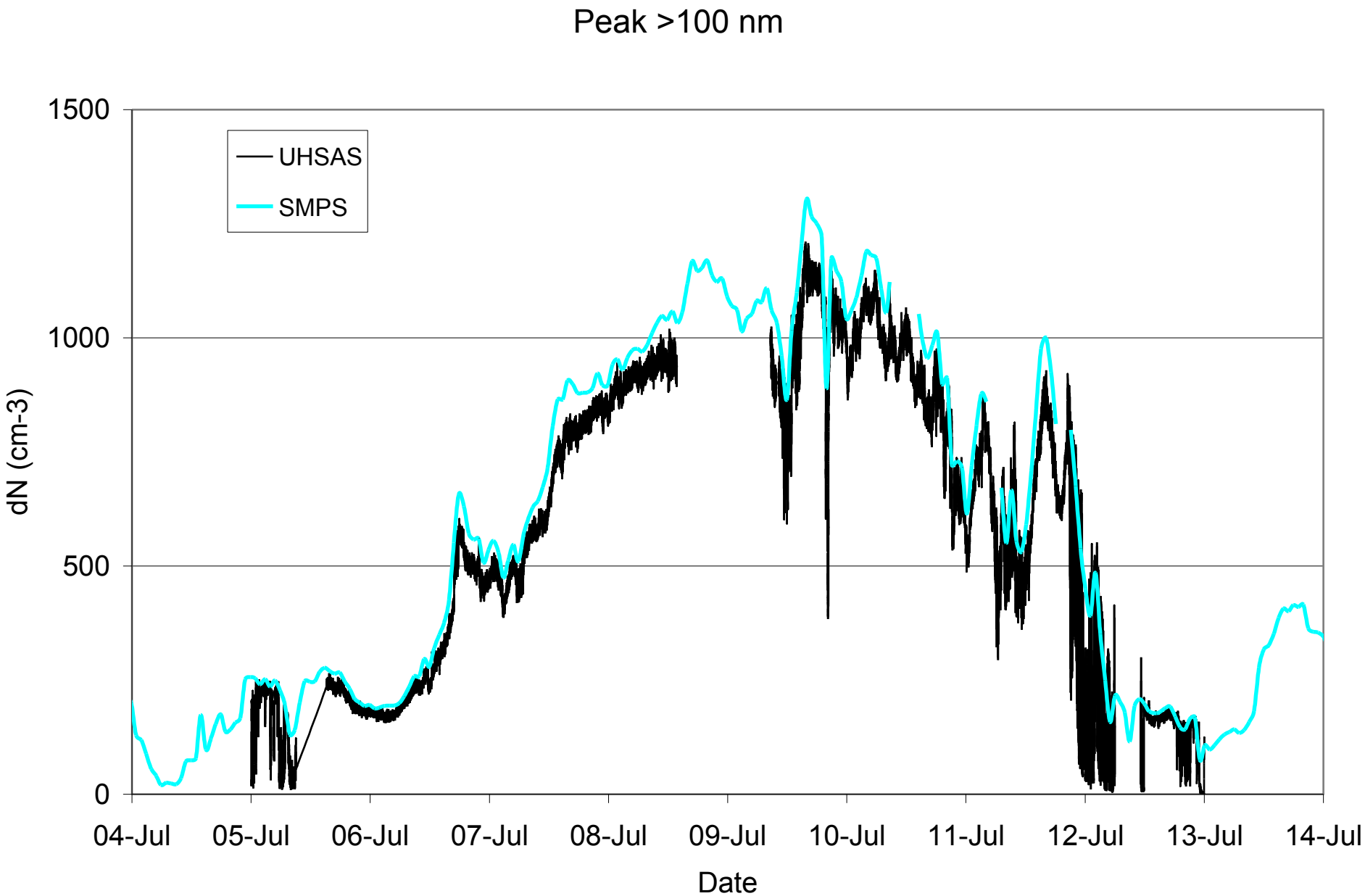


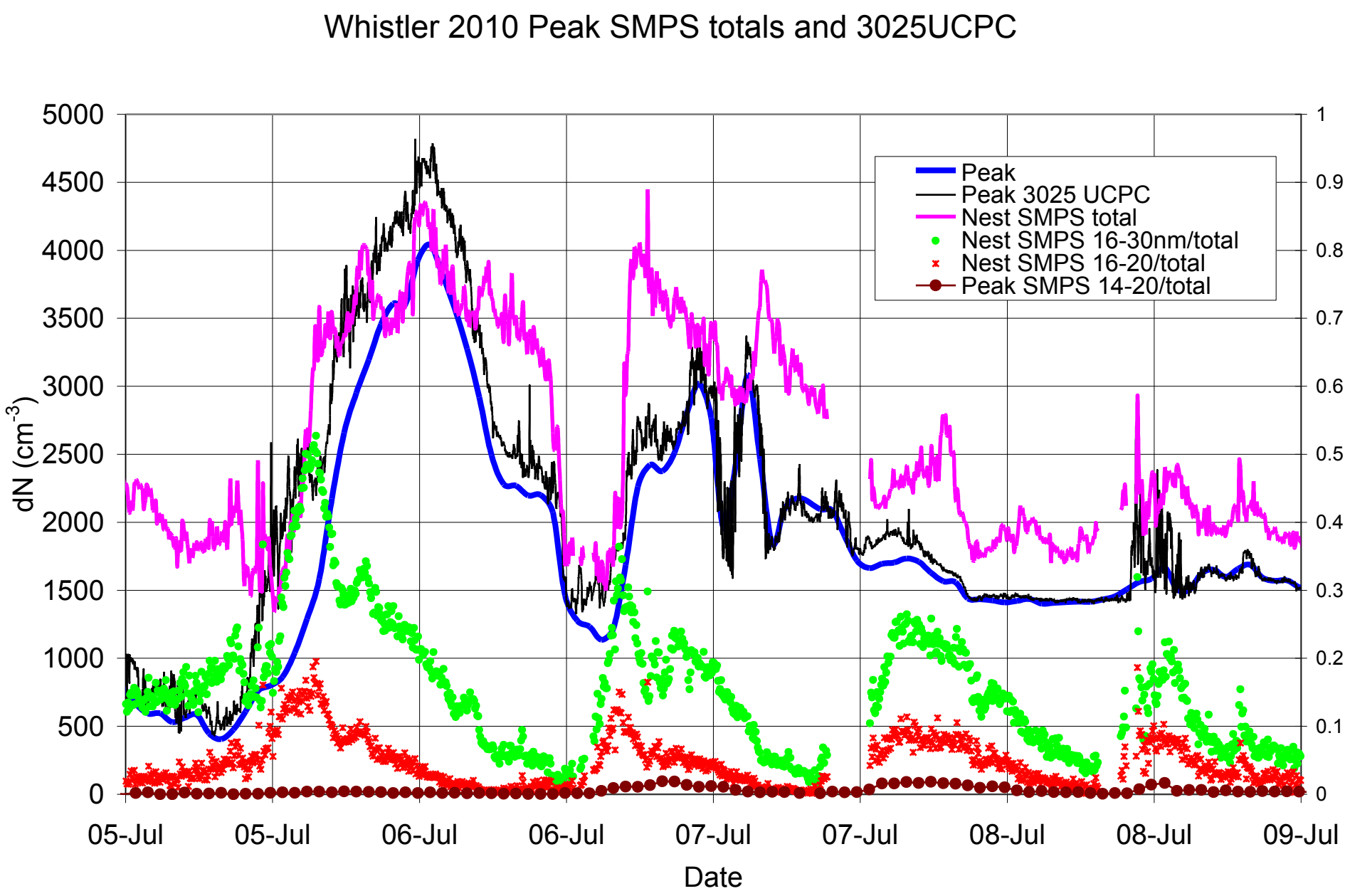
# SMPS and MSP total number cn comparison

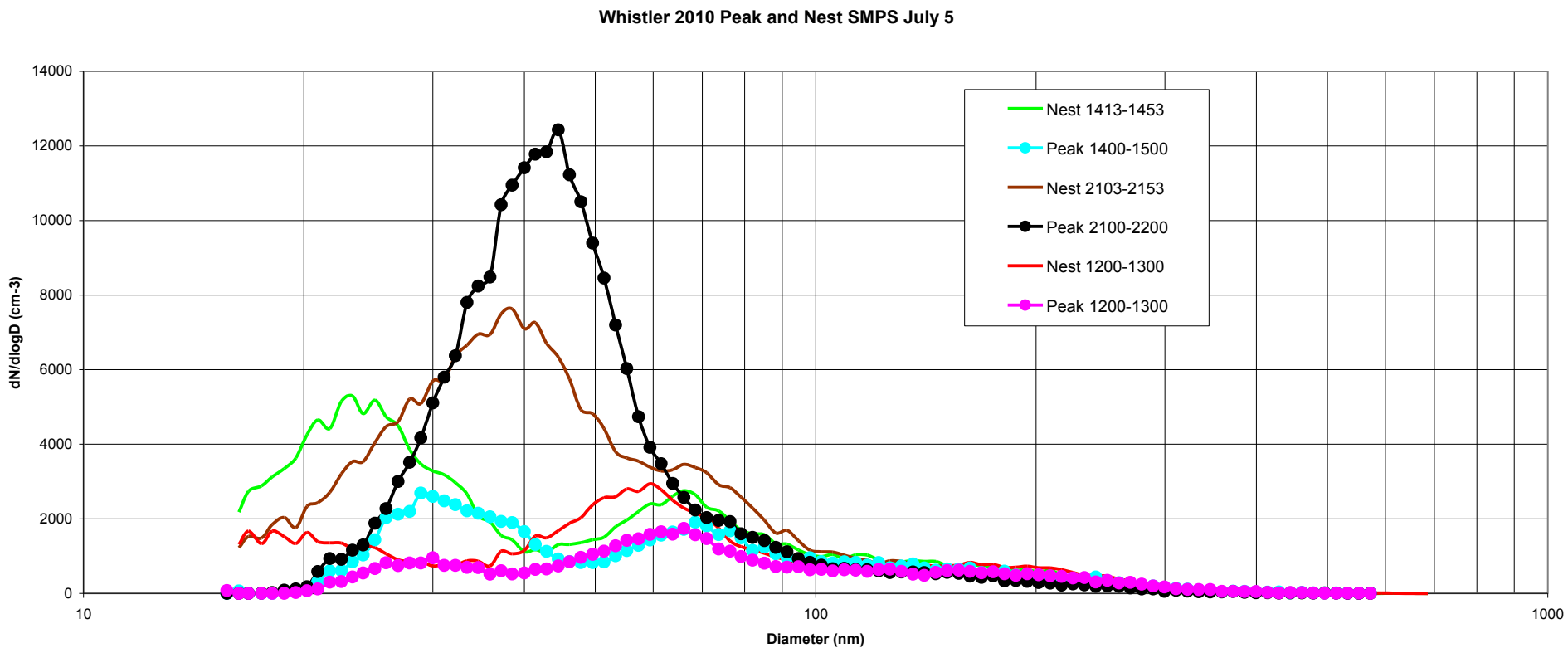


# SMPS and MSP dN>100 nm comparison




Peak >100 nm
UHSAS
SMPS
1500
1000
500
0
dN (cm-3)
04-Jul
05-Jul
06-Jul
07-Jul
08-Jul
09-Jul
10-Jul
11-Jul
12-Jul
13-Jul
14-Jul
Date


Whistler 2010 Peak SMPS totals and 3025UCPC
dN (cm-3)
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
05-Jul
05-Jul
06-Jul
06-Jul
07-Jul
07-Jul
08-Jul
08-Jul
09-Jul
Date
Peak
Peak 3025 UCPC
Nest SMPS total
Nest SMPS 16-30nm/total
Nest SMPS 16-20/total
Peak SMPS 14-20/total


Whistler 2010 Peak and Nest SMPS July 5
14000
12000
10000
8000
6000
4000
2000
0
dN/dlogD (cm-3)
10
100
1000
Diameter (nm)
Nest 1413-1453
Peak 1400-1500
Nest 2103-2153
Peak 2100-2200
Nest 1200-1300
Peak 1200-1300