

Dr. Barbara Ervens  
Editor of Atmospheric Chemistry and Physics  
NOAA

Dear Barbara,

Listed below are our responses to the comments from Referees #1-3. The referees' comments are in bold type and our responses are in italic. We thank the referees for carefully reading our manuscript and for excellent comments! These comments have led to a much better manuscript.

Also, Sonia M. Kreidenweis has provided us with significant input and advice on how to improve the manuscript and address some of the referee's comments. This input and advice warrants co-authorship, so we would like to include her as a co-author in the revised manuscript. We hope this is ok.

Sincerely,

Allan Bertram  
Professor of Chemistry  
University of British Columbia

# Anonymous Referee #1

The authors report their case study from two cloud events in southern California. The study involves an impressive number of instruments and methods and shows that refractory black carbon concentrations are consistent with kappa-Köhler theory.

Although carefully done, I am surprised that the manuscript does not express any science questions or hypotheses. It is not clear whether the authors expected that refractory black carbon concentrations do not conform to kappa-Köhler theory. Moreover, the quantification of the coating of the particles (and their activated fraction) is of high value.

Since I am not an expert in the field who knows all unwritten text on the topic, I would have benefited from a standard manuscript that clearly exposes a science question and hypotheses to address these questions. As is, the manuscript is a simple data publication, which has its value, but remains obscure in the aspects how representative the two cloud events are for average or extreme conditions at the site, and what we can learn out of it. The conclusions sound like “nothing new, but now with more accurate numbers”. To be acceptable for final publication in ACP I recommend that the authors clearly expose the questions they were asking, what hypotheses they established before carrying out their research, and what was learnt besides having more accurate numbers.

*To address the referee’s comments, the introduction will be re-written to more clearly outline the objectives of the study. Also, in the revised manuscript we will try and emphasize better what was learnt in the current study besides having more accurate numbers.*

Details:

- 1) Somewhere in the methods section or the first reporting of data you must specify what the uncertainty presented after the +/- sign actually is. Is it the standard error of the mean? Or standard deviation from the mean? Or the 95% confidence interval? Or something else? I question the assumption of symmetric uncertainties – please check and report on the distribution of your data and use the appropriate statistical parameter to specify uncertainty. Recall that SE and SD are parameters of the normal distribution and as long as it is not established that the distribution are normal, reporting SE or SD are not the key parameters of your statistics.

*To address the referee’s comments, in the updated manuscript we will specify what the uncertainties represent. In addition, in cases where there was ambiguity, we will specify the type of distributions to which the data adhered. Some specific changes that will be made to the document are listed below:*

- a. pg 11458 line 11: “(1) the five minute averaged CVI counterflow was within  $\pm 5\sigma$ ...” will be changed to: “(1) the five minute averaged CVI counterflow was within  $\pm 6\%$  of the mean counterflow (i.e.  $\pm 5\sigma$ , where  $\sigma$  is standard deviation)...”
- b. pg 11458 lines 15-24: all values reported will be converted to median values and all uncertainties reported will be converted to 10<sup>th</sup> and 90<sup>th</sup> percentiles since it was not demonstrated that these data conform to a normal distribution. Furthermore, the following sentence will be added at the end of the paragraph on line 24: “All values reported in this paragraph are reported as the median and the ranges shown in parenthesis represent the 10<sup>th</sup> and 90<sup>th</sup> percentiles
- c. pg 11462 line 1: The first sentence will be changed to: “The median  $AF(D_p)$  for the bulk aerosol and rBC are presented in Fig. 6, where the error bars represent the 10<sup>th</sup> and 90<sup>th</sup> percentiles for each 10 nm size bin.”
- d. pg 11462 line 12: The first sentence of Section 3.5 will be changed to: “Coating thicknesses were determined using a core and shell Mie model and are shown in Fig. 6c and d, where the error bars represent the 10<sup>th</sup> and 90<sup>th</sup> percentiles and the symbols represent the median coating thickness for each size bin.”

- 2) Section 2.9: more details on the operation of the FM-100 should be given. See Spiegel et al. (2012) – there are important issues with respect to facing the instrument towards the incoming airstream. Currently the reader has to assume that everything was done correctly, but the details must be given. The FM-100 does not provide the correct number counts by default, and the Mie-scattering correction suggested by Spiegel et al. (2012) is recommended, unless the authors used size bins that are broad enough to state that this does not affect their data. With 20 channels (of 40 that could be used) it appears that the authors probably used equal size bins which were large. But please be more specific on these details.**

*To address the referee’s comments, the following information on the operation of the FM-100 will be added to Section 2.9:*

*A fog monitor (FM-100, model 100, DMT, Boulder, CO), which is a forward scattering optical spectrometer, was located on top of the container. Details on the operational theory of the FM-100 can be found in Eugster et al., 2006 and Spiegel et al., 2012. Briefly, ambient droplet laden air is pumped through a wind tunnel and carried to a sizing region where droplets pass a laser beam ( $\lambda=658$  nm). Light that is scattered in the forward direction from a droplet crossing the laser beam is collected by photodetectors and the signals measured are used to assign which size bin the droplet falls within. From the measured cloud droplet size distribution both the total cloud droplet number concentration (CDNC) and the amount of liquid water content (LWC) present can be determined (Spiegel et al., 2012). The FM-100 used in this field study collected droplet counts from droplets with diameters from 2-50  $\mu\text{m}$  using the manufacturer’s predefined 20*

size bins. The size bin widths using this configuration were 2  $\mu\text{m}$  for droplets  $<20 \mu\text{m}$  and 3  $\mu\text{m}$  for droplets  $>20 \mu\text{m}$ .

During this study the FM-100 was located approximately 50 cm above the top of the container, approximately 1 m from the residual inlet, and 1.5 m from the total inlet. The instrument was mounted on a freely rotating board allowing it to be turned into the direction of the wind. The wind speeds encountered during the two cloud events sampled were in general light ( $< 2 \text{ m s}^{-1}$ ). When the wind direction was obvious, an operator ensured the FM-100 was turned into the wind. Calibrations of the FM-100 were performed by Droplet Measurement Technologies prior to installation.

- 3) Turning the FM-100 into the wind direction is very important for Cloud 2 where a  $\pm 90$  degree variability is given on p. 11458, l. 18. It is unclear that this actually means (if it is SD then it basically means that the wind was from all directions, although the mean was from 190 degree), but it must express a very unsteady wind vector during Cloud 2 and hence it is imperative to document proper orientation of the sensor under such conditions.

*See response to comment #2 above.*

- 4) 11454, 24: correct wording (and work flow) is to fit a function to the data, not to fit the data to a function!

*The wording in Section 2.5 will be changed as suggested.*

## Anonymous Referee #2

The manuscript presents total and cloud-residual aerosol measurements from two cloud events. There is a strong focus on BC aerosol, with interpretation of the ratio of total to cloud-droplet residual concentrations for activated fraction, and of “coating thickness” on BC for consistency with kappa-Koehler theory for CCN activation. This is an interesting “pool to play in”, and the observations are interesting; however, more discussion and analysis need to be included to present and support hypotheses about the implications and science to be gleaned from the two case studies included. Two specific areas that need more attention are the SP2 analysis, and inclusion of some discussion about the implicit assumption that BC is involved in the particles that activate as CCN before a droplet forms.

For the SP2 analysis, my concern is focused on two points.

- 1) **First, the detection efficiency of the SP2 is strongly dependent on laser intensity, the mass of BC in the particles, and, for small BC, the amount of non-BC material internally mixed with BC (Laborde et al., AMT 2012, Atmos. Meas. Tech., 5, 3077–3097, 2012 [www.atmosmeas-tech.net/5/3077/2012/doi:10.5194/amt-5-3077-2012](http://www.atmosmeas-tech.net/5/3077/2012/doi:10.5194/amt-5-3077-2012)).**
  - a. **This may be influencing the sharp reduction in apparent BC activated fraction at small sizes (Figure 6). The authors should include more information about SP2 laser power, and the level of agreement observed between the SP2s in cloud-free sampling.**

*The referee raises an important point. As the referee mentioned, the detection efficiency of the SP2 decreases when the diameter of rBC cores become small (i.e. less than approximately 70-90 nm) (Laborde et al., 2012, Schwartz et al 2010).*

*Since the main conclusions about black carbon in this manuscript are based on relative measurements taken with the two SP2 instruments (i.e. the residual SP2 and the total SP2) the detection sensitivities of the two instruments as a function of size need to be similar. To ensure that this was the case, we carried out the following: First we measured the rBC size distributions with both single particle soot photometers during cloud free sampling conditions and when the CVI was switched off. Not surprisingly, the two instruments gave slightly different results. Then we applied a size dependent correction factor to the SP2 connected to the total inlet, to bring the two results in agreement. This correction factor has been applied to all the results presented in the initial manuscript. To address the referee's comments this information will be added to the manuscript. We will also include in the manuscript, the size distributions measured with the two SP2s in cloud free conditions before and after applying the size-dependent correction factors to the total SP2. We will also include the size dependent correction factor applied to the total SP2 results.*

- b. **Note that cloud processed BC will naturally have thicker coatings than unactivated BC independent of the mechanism by which rBC became associated with the cloud droplets. This potential bias could affect the conclusion about the size distribution of rBC in the cloud residuals compared to total rBC.**

*Here the referee is pointing out that rBC particles can obtain coatings by in-cloud processes, such as aqueous-phase chemistry. To address the referee's comment, in the revised manuscript, we will discuss in-cloud*

*processes and the implications for the conclusions reached in revised manuscript.*

- c. **On another note, the influence of fresh rBC that likely is not thickly coated from the recent city overpass should also be explicitly considered.**

*See response to comment #3 below.*

- 2) **Secondly, not enough information is given about the potential biases associated with coating thickness determinations for the rBC-containing residual particles; I am concerned that the statement about increasing coating thickness with decreasing rBC core diameter could merely be due to an artifact (although it is expected a priori from our understanding of CCN), and encourage the authors to consider possible bias in their interpretation. At the heart of this issue the point that the SP2 is typically unable to optically size all rBC below an optical size roughly equivalent to that of a 220 nm PSL particle, which I think is roughly equivalent to the optical size of a bare rBC particle of volume equivalent diameter  $\sim 160$ nm. If one only sizes rBC-containing particles with at least this optical size, one will never capture the contribution to coating thickness of small bare or thinly coated rBC masses, but will only size small rBC masses with sufficient internally mixed material to reach this optical size threshold.**

- a. **I suggest that the authors carefully look at and present the statistics of LEO success in different rBC mass ranges to explore this (i.e. in your nomenclature, how what fraction of failed LEO fits is attributed to lack of elastic scattering signal at the 5% of peak laser power point, and how does this fraction change with rBC core mass?). I think that you need more analysis to evaluate whether the biases incurred from 50% LEO failure is affecting your interpretation of coating thickness trends.**

*This referee as well as Referee #3 brings up the issue of potential biases associated with the coating thickness analysis. We thank these referees for bring up this important issue. To address the referees' comments we will: 1) explain better why we observed LEO failure in some cases, 2) present in much more detail the statistics of the LEO failure and 3) directly address the issues of potential biases raised by Referees #2 and #3. Also, note in the original manuscript we reported an approximate 50 % LEO failure. In the revised manuscript, this failure rate will be slightly improved since we have found solutions to these failures in some cases.*

- b. Finally, I don't understand how your failure "b" of scattering at time zero (i.e at the center of the laser beam?) is relevant to the LEO fit. These issues affect figures 6 and 7.**

*In the revised manuscript we will explain better why we observed LEO failure.*

- 3) There needs more discussion of the microphysical route that rBC takes to get incorporated into the cloud droplets. What is the working hypothesis about the interaction?**

**Do you believe that BC are internally mixed with other materials in CCN before activation?**

- a. What is the coagulation rate for the freshly emitted rBC from the city with existing cloud drops?**

**What do these measurements potentially teach us about BC interactions with cloud droplets?**

*Two possible mechanisms exist for incorporating rBC containing particles into cloud droplets: nucleation scavenging and coagulation between the rBC containing particles and cloud droplets. Results and calculations suggest the dominate mechanism for incorporating rBC particles into the cloud droplets studied is nucleation scavenging. First, the fraction of rBC containing particles activated into cloud droplets increases as the size of the rBC core increases (See Figure 6 A and B in the original manuscript). If coagulation dominated we would expect to see an opposite trend. Second, calculated coagulation rates together with estimated lifetimes of the cloud droplets, cannot explain the fraction of rBC containing particles activated into the cloud droplets – the calculated coagulation rates are too small. To address the referee's comments this discussion will be incorporated into the revised manuscript, including calculations of coagulation rates.*

- 4) How do these results bear on the different approaches to modeling included in the introduction? What is learned from having observed two clouds? The statement that "rBC contributes to CCN" suggests (to me) an active role, what evidence supports this?**

*In the revised manuscript we will try and emphasize better what was learnt in the current study besides having more accurate numbers. In addition, see response to Referee #2, comment #3.*

**Specific comments:**

- 5) **Introduction: please clarify the sentence including “we measured BC as a function of size: : compared to BC as a function of size.”**

*To address the referee’s comment, the sentence on pg 11450 line 20 will be modified to the following:*

*“In this study the size resolved activated fractions of rBC and the total aerosol were investigated at a marine boundary layer site (251m a.m.s.l.) in La Jolla, CA.”*

- 6) **Section 2.4: Aquadag causes a higher SP2 response per unit mass than ambient rBC. For aquadag calibrations, it is recommended that the Aquadag signal used in the calibration be downward scaled by 0.75 for a better calibration (see Baumgardner et al.: Atmos. Meas. Tech., 5, 1869-1887, 2012 [www.atmos-meas-tech.net/5/1869/2012/doi:10.5194/amt-5-1869-2012](http://www.atmos-meas-tech.net/5/1869/2012/doi:10.5194/amt-5-1869-2012)). It does not appear that this was done here.**

*Thank you for pointing this out. To address the referee’s comments in the modified document the measured peak heights of the Aquadag calibrations will be scaled down by a factor of 0.75 and all calculations for rBC will be recalculated with the new calibration equations. In addition, in Section 2.4, this downward scaling will be discussed. Specifically, we will add the following sentence to Section 2.4:*

*“Using the recommendation by Baumgardner et al., 2012 the average peak heights determined for each of the Aquadag® sizes were scaled downward by 0.75 since Aquadag has been shown to cause a higher SP2 incandescence signal response per unit mass than ambient BC.”*

- 7) **Section 2.5: Please explicitly note that the coating thickness has not been validated experimentally, and merely provides consistency between observed optical scattering and mie theory. Of course having noted this, I’ll suggest that you extrapolate even further, to coating volume, which is the more relevant quantity for kappa-Koehler.**

*To address the referee’s comments we will point out the caveat raised by the referee in Section 2.5. Specifically the following sentence will be added:  
“It should be noted that the coating thicknesses have not been validated experimentally, but merely provide consistency between the observed optical scattering and Mie Theory.”*

*In addition, we will add four panels to Figure 6 that will show the coating volume as a function of the rBC core diameter and the volume fraction of the coating as a function of the rBC core diameter.*

- 8) **Non-rBC particles passing through the SP2 can be used to constrain peak position and full width half maximum at times concurrent with the BC**



**observations. These parameters can drift, so I suggest that you check variability in this way, and update the parameters on a reasonable time scale if necessary.**

*Non-rBC particles were used to validate the laser peak position and FWHM throughout the sampling periods. We will add discussion on this point to the revised manuscript to address the referee's comments. Also, we will add two new figures to the revised manuscript that will compare the full Gaussian fit amplitudes to the leading-edge only fit amplitudes for purely scattering ambient particles measured during the field campaign. These figures illustrate that the peak position and full width half maximum did not drift significantly during the field campaign.*

- 9) Note that for the SP2 measurement at 1064 nm, there are better estimates for the index of refraction of rBC – see: Method to measure refractive indices of small nonspherical particles: Application to black carbon particle by Nobuhiro Moteki, Yutaka Kondo, Shinichi Nakamura, 2010, in JAS. It is necessary to include the density of the rBC assumed in converting SP2 mass to mie-diameter.**

*Thanks very much for binging this to our attention. The coating calculations will be redone using a refractive index of  $2.26-1.26i$  (Moteki, Kondo, & Nakamura, 2010) and the following text will be added to Section 2.5:*

*“The complex index of refraction used for the core was  $2.26-1.26i$  (Moteki et al., 2010b, Taylor et al., 2014D) and for the shell was  $1.5-0.0i$ , which is consistent with that of dry sulfate or sodium chloride (Metcalf et al., 2012, Schwarz et al., 2008a, Schwarz et al., 2008b). An rBC density of  $1.8 \text{ g cm}^{-3}$  was assumed.”*

- 10) Section 2.6: SMPS and SEMS agree to 4% in what? Total count? Count at each size?**

*The text in Section 2.6 (pg11456 line 7) will be modified to:*

*“During cloud free sampling periods the total number concentrations of particles with sizes between 70 and 400 nm measured with the SMPS and SEMS agreed to within 4%”*

- 11) Section 3.2: +/- 5 sigma does not give the reader much information. Can you present in a more useful way as, e.g. stability in CVI-D50, or % flow?**

*Please see the response to Referee #1, comment #1.*

- 12) Figure 3 does not do a good job in showing the narrated relationship between droplet number and LWC. Perhaps a scatter plot would get this point across more clearly and strongly.**

*This is a reasonable suggestion from the Referee, but the relationship between droplet number and LWC was only included as a qualitative observation. Since there are no major conclusions drawn from this relationship and since we would prefer to keep the number of plots to a minimum, we would prefer not to include a new plot to show the relationship between droplet number and LWC, unless the referee strongly objects.*

- 13)Section 3.3.2: Please comment on the likelihood of droplet condensation on affecting bulk residual size distributions; it is not clear that the CCN that activated have not been affected in the cloud.**

*See response to Referee #2, comment #1b.*

- 14)It is not clear if the residual rBC size distributions have been biased by low laser power/size dependent SP2 detection efficiency. The Detection Efficiency of the Single Particle Soot Photometer (Schwarz et al 2010) discusses these issues. If there is concern about this biasing rBC results, an easy fix is to remove the smallest rBC cores (e.g. below 100 nm or so? ) from the analysis.**

*Please see the response to Referee #2, comment #1a.*

- 15)Section 3.5: These results should be examined for possible bias as discussed above.**

*Please see the response to Referee #2, comment #1a.*

- 16)Section 3.6: As the rBC-containing particle size distribution in the ambient is different than that of bulk aerosol, it is not clear to me what Figure 7 is meant to show in terms of the comparison of rBC and bulk aerosol.**

*Figure 7 was meant to show the role of total diameter (i.e. rBC core diameter + 2\*coating thickness) in cloud droplet activation. The discussion on this figure will be modified in the revised manuscript to make this point more clearly. If we are unable to make this point more clearly, then we will remove the figure from the manuscript.*

- 17)Section 3.7: “rBC cores have thick coatings which lead to overall particle diameters > 100 nm”..This assumes that rBC was present before activation. Please add discuss/support.**

*See response to Referee #2, comment #3.*

- 18) The relevant parameters for kappa-Koehler are the volume and kappa of non-BC material. Figure 9 essentially hides the fact that this is independent of BC content. I suggest explicitly discussing this, and attempting to show this in the context of both Figures 7 & 9, by plotting the horizontal axis as a diameter of non-BC materials (i.e. the diameter of only relevant materials for kappa-Koehler theory).

*See response to Referee #2, point 7. In addition, a new panel will be added to Figure 8, where  $Sc$  will be plotted as a function of the volume fraction of the coating at different rBC core diameters.*

## **Anonymous Referee #3**

This MS gives data collected at two out of three cloud events during a field campaign at Mt. Soledad, California. It contains a wealth of data on scavenging of rBC, which could, however, benefit from a more detailed analysis. The authors are very clear about problems experienced during sampling. Actually, the problems that occurred are the usual ones in field studies but are seldom mentioned.

The data are valuable, but the study needs more work and more discussions of implicit and explicit assumptions, as well as a clearer description of the measurement set-up and the measurements themselves. I'll go through the points in the order they appear in the text.

**Abstract:**

- 1) The abstract mentions the total inlet and the CVI (lines 4 and 5), but it is unclear from the abstract alone that the CVI actually is the residual inlet.

*The abstract will be modified to make this clear.*

- 2) Giving percentages of cloud droplets sampled by the CVI without mentioning briefly how total droplet concentrations were obtained is rather confusing.

*The abstract will be modified to try and clear up this confusion.*

- 3) The main results given in the abstract, which are also major results of the study, refer to coating thicknesses of rBC cores of different diameters. The point of coating thickness and core size is one of the most critical ones of the whole study and needs further discussion and investigation (see

below) could the fact that small rBC cores were found to have thick coatings and large cores thin coatings be an artefact of the upper size cut of the SP2 (220 nm) as larger particles (core+shell) could not be detected?

*See response to Referee #2, comment #2a.*

**4) section 2.2 inlets:**

**Figure 1 is insufficient in its present form. It doesn't show how the droplet residuals were moved to the different instruments - were the particles passed into a sampling manifold where the instruments sampled in parallel or was there a common sampling line with the instruments connected sequentially?**

*To address the referee's comments Figure 1 and the corresponding discussion of the figure will be modified to make it clear how the droplet residuals were moved to the different instruments.*

**5) Where was the FM-100? As percentages of droplets are given: what was the spatial distance between the aerosol inlet(s) and the FM-100?**

*See response to Referee #1, comment #2.*

**6) How about the homogeneity of the cloud? Patchiness could give a huge effect if the total droplet concentrations and CVI droplet concentrations were not measured at the same spot (and even then patchiness can give problems)**

*During this study the FM-100 was located approximately 50 cm above the top of the container, approximately 1 m from the residual inlet, and 1.5 m from the total inlet. This information will be added to the revised manuscript to address the referee's comments. In addition, data were classified as in-cloud and included for analysis if the five minute averaged LWC was greater than  $0.05(\text{g m}^{-3})$  to remove periods of entrainment, or "patchy" regions of the cloud as much as possible (Cozic et al, 2007).*

**7) Section 2.4**

- a. refractory black carbon mass measurements / Section 2.5 coating thickness: how accurate is the calibration with Aquadaq soot? How reliable is the lower cut size of 70 nm? SP2's have their problems, and more discussion of the accuracy of the data is needed.**

*To address the referee's comments more discussion on the accuracy of the data from the SP2 will be included. Specifically, the following*

*sentence will be added to Section 2.4: “The uncertainty in the rBC mass (at the 95% confidence limits) stemming from uncertainty in the fit of the calibration data was 1-16% for SP2<sub>Res</sub> (depending on particle mass) and 3-25% for SP2<sub>TotL</sub> (depending on particle mass)” With regards to the lower cut size of 70nm, we believe the referee is referring to the detection efficiency of the SP2. Please see response 1a for referee #2 for further details on discussion of the SP2 detection efficiency.*

- b. Same for the measurements of the coating thickness. Why could only 50% of the particles be fitted with the LEO procedure? Please give reasons. This could be a severe limitation of the results of the study.**

*See response to Referee #2, comment #2a.*

- 8) Section 2.6: which condensation particle counters exactly were used with the SEMS and SMPS?**

*Information on the exact condensation particle counters used will be added to the revised manuscript.*

- 9) Section 2.9: this section is much too short. How was the FM-100 operated? Positioning with regard to wind direction? Operation principle (just a short description), calibration, accuracy etc.? This is crucial for the comparison with the CVI, especially as the droplet sizes seem to be quite small (and the LWC of cloud 2 very low)**

*See response to Referee #1, comment #2.*

- 10) Section 3.3.1: if there is evidence of bimodality in the BulkAero\_tot distributions, why perform the size distribution fit only with a single mode lognormal distribution?**

*To address the referee’s comments in the revised manuscript we will make it clearer why we used a single mode to fit the data.*

- 11) Section 3.3.2 / 3.5: last par. of 3.3.2: most rBC cores are rather near the lower cut size of the SP2. The comparison of the rBC<sub>res</sub> and the rBC<sub>tot</sub> shows that the droplets contain larger rBC cores than the unactivated aerosol, and that the coating thickness (section 3.5) of larger cores are thinner than those of smaller cores. As the coating thickness could only be determined for the residual inlet, and as the SP2 there had actually a very narrow size range (70 - 220 nm), the result that large particles have thin**

coatings and small particles have thick ones could be an artefact of the upper cut size of the SP2?

*See response to Referee #2, comment #2a.*

**12) If the particles have core-shell structure, of course they are internally mixed, but maybe this should be explicitly stated. Internal mixture can already have occurred in the aerosol entrained into the cloud, and new droplet activation of these particles, as well as through collision of unactivated particles with droplets. Any way to estimate this?**

*See response to Referee #2, comment #3.*

**13) Section 3.7 / Conclusions There seems to be an implicit assumption that all activated particles have to have diameters > 100nm - why?**

**14) We did not intend to imply that all activated particles have to have diameters > 100nm. We are sorry for the confusion. In the revised manuscript we will adjust these sections to try and remove this confusion. Do you have meteorology based estimates of the supersaturation history in the clouds (apart from the calculations with kappa-Koehler theory)?**

*We do not have meteorology based estimates of the supersaturation history, but during the field campaign CCN instruments were connected downstream of the CVI and used to derive upper limits to the cloud supersaturation (Modini et al. 2014, in preparation). The upper limits determined from the CCN instruments was 0.1%, which is consistent with the estimated Sc values reported in the current study. This information will be added to the revised manuscript.*

**15) Please discuss why are there no large rBC cores with thick coatings? Is this caused by the failure of the LEO fitting procedure? If it is, then this point has to be discussed in depth. The main results are the sizes and coating thicknesses of rBC incorporated in cloud droplets, and if there are severe limitations to the analysis procedure, the limitations have to be stated at every mention of the results. If it is due to the upper cut size of the SP2, then it should also be stated explicitly**

*Please see response to Referee #2, comment #2a.*

**16) Very minor points:**

**a. section 3.7.1 Crosier (2007) is missing from the list of references**

*The reference the referee is referring to is Gysel and Crosier (2007). Due to the spacing of the posted discussion article the reference "Gysel and*

*Crosier (2007)" were split over two pages making it appear that there was a reference to just Crosier (2007).*

- b. section 3.7.3 better use molecular mass instead of molecular weight - it really is a mass and not a weight, even though "weight" is often used.**

*As suggested, molecular weight will be changed to molecular mass.*