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Interactive comment on "Size-selected black carbon mass distributions and mixing state in polluted and clean environments of northern India" *by* Tomi Raatikainen et al.

Anonymous Referee #2

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It is very exciting to see SP2-measured rBC mass loadings and microphysical state information coming out of India. This important yet under-reported region is a critical piece in the "BC puzzle". Raatikainen et al,. present SP2 measurements of mobility-diameter selected aerosol, and have obtained data which, if appropriately quality-assured, will be of high value and interest to the wider community.

The manuscript needs substantial improvement. Most significantly, the quality of the rBC data set should be more carefully assessed: a ~20% bias to a CPC was corrected simply based on the assumption that the CPC was correct. Potentially larger biases than this exist in the data.

Generally, the manuscript should be proof-read for grammar. Imprecise vocabulary

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(e.g. “absorbing particles” for rBC-containing particles) should be revised (there are many light-absorbing particles that may not contain rBC). Background information about the performance of the SP2 should be more clearly summarized in the main text. I also found it difficult to follow how the size-resolved results were combined to reflect the ambient aerosol condition. The results should be more thoughtfully presented (for example, the section about BC volume fractions based on the assumption that the total particle volume is proportional to mobility diameter cubed is followed by a section indicating the lack of value of that analysis after inspection of SP2 LEO fit results).

Specific comments:

1) “Absorbing” and “refractory” are terms that apply to rBC, but not all light-absorbing or refractory particles are either rBC-containing, or detected via incandescence in the SP2. For example, only a small fraction of dust particles incandescence in the SP2, although most of them are refractory. The paper should be made more precise by avoiding un-specific vocabulary. Two examples: 1) page 2 line 13 – the SP2 will not help determine a number fraction of light-absorbing particles; 2) page 3, line 19: the Sp2 does not measure all “refractory absorbing material”. 2) Page 1 line 18: BC mixing state also depends on source. 3) Page 2 Line 11 and page 4, Line 17 and page 9 lines 5 - 11: The light-scattering for rBC-containing particles from the SP2 is a direct measurement (this is essentially a measurement of the particle optical size), and requires no assumptions about particle index of refraction or morphology. However, interpretation of the light scattering in terms of particle composition etc. does require assumptions. 4) Methods: a. Please specify if the aerosol was dried before sampling. b. It is necessary to consider SP2 detection efficiency of rBC, which depends on laser intensity and is influenced by mixing state (see Laborde et al., 2012 – the AIDA intercomparision in AMT, and Schwarz et al., 2010 – Detection efficiency of the SP2 in AMT). A first order estimate of laser intensity can likely be obtained from “YAG power” if Droplet Measurement Technology provided a calibration of this. Note that “YAG power” and laser intensity inside the chamber (where it is critical for rBC detec-

tion) are both temperature sensitive. The statement in the supplement that laser power changes equivalent to 17% in diameter suggests even larger laser intensity changes (depending on the size of the calibration aerosol but likely more than a factor 2; please present the relative change in laser intensity inferred from the ambient measurements and assuming constant index of refraction). c. The supplemental results showing dependencies on detection from the SP2 on chamber temperature (figure S4) for the DMA set at a quite large size (285nm) are quite concerning since the scattering-aerosol detection limit was specified to be 180 nm in the main text. The suggestion that the CPC is a reference instrument and the SP2 is of low quality is not sufficient to simply correct the SP2 concentration without testing any hypothesis as to the origin of the offset (CPC's can undercount!). Over the temperature range specified, the SP2 LFE and Ashcroft differential pressure meter have much smaller temperature sensitivities than the 30% shown in the figure. Hence the basis for a correction, and for establishing the absolute uncertainty of the SP2 measurement is not clear. Note that the good correlation of Aethalometer and Sp2 data is insufficient to rule out potentially large bias in either/both instruments, and does not validate the rBC size distributions, which have much smaller impact on rBC absorption than concentration; the Aethalometer result needs a large scaling factor that depends on total aerosol, and is highly site specific (see, for example: http://www.esrl.noaa.gov/psd/iasoa/node/81). It will be necessary for the authors to build a reasonable basis for evaluating the instrumental uncertainty. As part of this, I recommend including (in the supplemental material) peak-height distributions for the size-selected aerosol from the SP2 from these tests. d. Page 3, line 18: here the SP2 is indicated to detect both rBC-containing and rBC-free aerosol; this lead me to be confused about if the SP2-derived rBC-free concentration was used for anything other than comparison of detection efficiency with the CPC – was Sp2 rBC-concentration used with SP2 rBC-free concentration for the number fraction rBC-containing particls?. Can the data sources for the various parameters be included in a table or clearly summarized in the text? e. Please briefly summarize the inversion for the reader here in the main text. I found the supplemental material also confusing, as

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the DMPS number size distribution comes from the DMA + the CPC: how is the ratio of "DMPS number size distribution to CPC concentration" particularly meaningful? The basic point is that SP2 concentrations are corrected to account for the number fraction of particles in a particular size bin with other than 1 electron charge? Please expand to explain how multiply charge particles are dealt with; this is likely a question that is in readers minds when looking at figure 3. This is at the heart of interpreting the "basic" SP2 data of rBC concentration and size distribution. f. Section 3.1: g. The Supplemental figure S1 should be updated to show that the Aethalometer sampled in parallel with the DMA. 5) Results: a. Page 4, line 25: What is the basis for this assumption? Every SP2 user sees rBC associated with singly and doubly charged (at least) peaks when calibrating with Aquadag or other materials (this is the bare rBC case). In the case of size-selected internally mixed particles, why would a narrow rBC size distribution be assumed? b. Section 32. Please address multiply-charged particles in the DMA. Page 5, line 18 – As these particles appear to be mostly bare, fractal rBC, how do you know that the ∼210 nm mode are not doubly-charged particles? This analysis would be helped by including the information from LEO fitting which was done. A prioriy one would expect that, unless there were dramatic changes in fractal dimension (which the authors rule out), a single size selected aerosol population will contain a continuum in which more massive rBC cores are associated with lesser amounts of non-rBC material, and smaller cores are associated with larger amounts of non-rBC. c. Page 6, line 1: note that rBC mass is a better first order proxy for the absorption cross-section in each size bin. d. Page 6, line 22 – 27: what are the values and total uncertainties observed here Page 6, line 30: Secondary rBC modes at larger sizes have been observed in China, these should be cited here: Wang et al., Shen, Black carbon aerosol characterization in a remote area of Qinghai–Tibetan Plateau, western China, Science of The Total Environment, V.479–480:151-158 (2014) and Huang et al., Black carbon measurements in the Pearl River Delta region of China, Journal of Geophysical Research, 116(D12208), doi:10.1029/2010JD014933, 2011. Is anything similar observed here? e. Page 7, line 6 and line 25: the volume ratio goes as the

ratio of diameters cubed, not as the linear ratio. This should be corrected throughout the paper. Line 21: The low ratio of rBC to total volume is in fact not distinctly different from previously published SP2 results based on LEO interpretation with Mie shell-and-core model. The assumption of this analysis (that Dm provides a route to total particle volume even for rBC-containing particles) is shown to be false in the next section; the results should be more clearly presented. f. Page 9 – why aren't any of the LEO results shown? What was the average scattering cross-sections? What fraction of rBC-containing particles were successfully fit? If the particles are bare, the scattering cross-section to rBC mass relationship should match expectations for material with the index of refraction measured by Moteki et al., Method to measure refractive indices of small non-spherical particles: Application to black carbon particles, J. Aero. Sci., 2010. The analysis of page 10 requires the LEO data, or analysis of the evolution of scattering signal for individual rBC particles, to support the assumption that the rBC particles are bare. 6) Conclusions: Please include quantitative values with uncertainties in this section. 7) Table 1: the $\pm$ values are standard deviations? This should be stated in the caption. Please include absolute uncertainties for rBC concentrations at least. 8) Figure 2: The time series does not seem to be extensively referenced in the text, perhaps this figure should be moved to supplemental material? 9) Figure 5: DMA data down to 20nm was taken. Why is the full range not shown if the clearly biased data below 200 nm is shown? I would prefer to see the whole range. Here it seems that the CPC data was not used for total particle number – why not? And if not, how is rBC number fraction $\sim$50% down to 100 nm when the SP2 scattering particle detection limit is specified to be 180 nm?

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