We thank the reviewers for their constructive comments and have made changes, where deemed appropriate. Specific responses to each of the comments are provided below (reviews' comments in black and our responses in red).

Anonymous Referee #1:

General comments:

This paper presents new data on size resolved aerosol absorption measurements by extraction of size-resolved and bulk aerosol samples into different solvents, followed by spectrophotometric measurements of the solutions. These authors have published several papers already on bulk aerosol absorption measurements in different locations (Los Angeles, the Southeast U.S.). This paper extends the prior measurements by extraction from a size resolved collector at three different sites in the Atlanta, GA area.

Measurements are compared with the previously reported bulk sampling method, together with standard filter-based aerosol optical properties (e.g., wavelength resolve Aetholometer). Although the time resolution of the size resolved measurements is lower (48 hours rather than 15 minutes), the size resolved measurements provide new data on the mass absorption efficiency and complex refractive index of ambient particles as a function of both wavelength and size. The size resolved data also allow for extraction into two different solvents, demonstrating that organic solvents more efficiently extract absorbing chromophores than does water. This observation implies that prior water extract measurements underestimate brown carbon absorption. Comparisons between the three different measurement sites, which span the regime from near roadway urban, to non-roadway urban to rural, points to interesting differences in the aerosol absorption characteristics, with an indication of a relatively larger brown carbon aerosol source at the rural site. The quantification of wavelength dependent brown carbon absorption relative to black carbon absorption is nicely placed in the context of previous measurements by other methods. Absorption by brown carbon is an important current topic, and this paper advances the exiting database for ambient measurements and analysis.

Generally, the paper is well written, methods are clearly defined, the conclusions interesting and well justified. There are three general comments that could improve the overall manuscript. First, the authors don't highlight the specific value added by the size resolution in either the abstract or the conclusions. Since size resolution differentiates these measurements prior ones, this lack of emphasis is somewhat surprising. Does the size resolution simply enable the Mie calculations, or does it

inform our understanding of the sources of brown carbon? Second, there is little discussion of the role of relative humidity and hygroscopic growth on aerosol absorption determinations (see more specific comments below). Third, some of the data in the tables would be more effective if plotted graphically, and would make the manuscript easier for the more casual reader. These suggestions are listed as "optional" below.

The Abstract has been modified to emphasis the importance of the size resolved BrC data. The other two comments are addressed below.

All of these comments, and the more specific comments below, fall in the category of "minor." I recommend publication after minor revisions.

Specific comments:

1) Abstract, page 18234, line 7. Although it is clear in the text, in the abstract, the meaning of "central site" is not clear, nor how that would be differentiated from a "road site near the city center."

We have modified the text, changing "central site" to "representative urban site" (Page 2, Line 39).

2) Page 18237, line 16-18: "light absorption measured from liquid extracts does not suffer from the interference by BC or other absorbers, since thy can be isolated by dissolution .." Is BC known to be totally insoluble in water or methanol? Is there a quantitative limit for the solubility of BC, especially in an organic solvent?

It is generally assumed that BC is insoluble in water and organic solvents [eg., Andreae and Gelencsér, 2006]. The strong dependence of absorption on wavelength for the filtered extracts (i.e., Absorption Angstrom Exponents between 5 and 7) in contrast to a known much weaker dependence for BC supports the view that BC is not soluble. Furthermore, BC absorbs strongly over all wavelengths and so the process of zeroing the spectra by subtracting all absorption measurements by the value at 700nm, will remove any BC absorption, if it existed.

Reference:

Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing 5 carbonaceous aerosols, Atmos. Chem. Phys., 6, 3131–3148, doi:10.5194/acp-6-3131-2006,2006.

3) Page 18242, line 6: Check for grammatical errors.

The sentence was modified as follows:

As ~~noted,~~ the MAAP corrects for scattering, ~~and thus the~~ absorption (bap_MAAP) was determined directly from the instrument's reported BC concentration using the fixed mass absorption coefficient of 6.6m2 g−1 at 670 nm.

4) Section 2.4.2 and Mie Calculations. There is no discussion of the effect of relative humidity here or in the data analysis that follows. Particles are stated to have been collected at ambient RH by the impactor, such that the particle size is that of the ambient RH rather than dry particles. The mass in equation (9) is a dry mass (correct?), with an assumed density of 1.5, such that the number of particles in equation (8) may have been overestimated in the transformation to equation (9). The authors should comment on the role of hygroscopic growth relative to dry particles in the Mie calculations and the subsequent comparisons.

The Mie calculations predict the light absorption of a single particle based on the complex part of the refractive index, which is determined from the solution light absorption. The calculation requires a particle mass and density (eg, see Eq 6 and 7). To estimate light absorption by all particles of that size the total number of particles must be estimated, which again requires a mass and particle density (eg, see Eq 9). As long as a consistent mass and density are used throughout the calculation the final results is, for the most part, insensitive to the choice of mass or density (see uncertainty analysis). One could use a mass and density of aerosol that includes water or just the mass of organic material and overall aerosol density, as we did, either way the results are similar.

5) Page 18243, line 22 (and Table 7, nomenclature): Text should specify, in addition to the table, that "absorption", A, is unitless. Presumably this quantity is equal to –log10(I/I0)? Perhaps this is stated elsewhere? In any case, this term is also commonly referred to as "absorbance," which the authors may wish to note for clarity.

The text is modified (Page 15, Line 309-310, absorption->absorbance, A_λ->A_λ, unitless; Table 7, Line of A(λ): light attenuation->light absorbance, A(λ)-> A_λ).

6) Section 3.2 and Table 3: An optional suggestion to the authors: The information in the table is important and interesting, but would be more effectively presented as a figure plotting the various quantities as a function of wavelength and overlaid with one another (e.g., six graphs, with JST, YRK, RS results for each quantity). This format would allow presentation of more wavelength information (higher resolution) than shown in the table.

Section 3.3 and Table 4: Same (optional) comment. Plots of the wavelength dependence of k for each size (with sizes overlaid on the same plot) would allow presentation of more wavelength data (which the authors presumably have) and would be much easier for readers to interpret at a glance (especially with respect to the rather fine print in Table 4).

We agree with the referee that a plot would be qualitatively easier for readers, but considering the large amount of information included in the table, we believe showing specific numbers is more straight-forward and quantitative, making it more suitable for any future comparisons/analysis.

7) Page 18252, line 23: "suggesting a source in the rural region" – presumably meaning a brown carbon source? Can the authors go further in speculating what such a source might be? Does this imply brown carbon from biogenic VOC oxidation?

With the low level of EC, relatively high concentrations of brown carbon suggested that there is a brown carbon source other than primary emission in the rural region. The brown carbon could be from biogenic VOC oxidation (ie, new SOA mass) or further enhanced with chemical aging (ie, chemical transformation), but we do not have direct evidence from this study to support either. Further investigations, including both in-lab and field observations, on the brown carbon formation process and key tracers, would help answer this specific question.