

Response to reviewer Helmuth Horvath

The authors thank Dr. Helmuth Horvath for his useful comments (highlighted in yellow). The point-by-point responses for individual comments are given below.

Reviewer general comment #1

This is a very well designed study and gives good insight into the role of light absorption and its interdependence on other variables such as shape or coating.

Unfortunately I could not find information on the optical setup of the Droplet Measurement Technology photo-acoustic spectrometer, but I doubt that it is optimized to determine the scattering coefficient. Most likely the photomultiplier will give a signal proportional to a weighted integral of the scattering function. This means that depending on the particle size a certain fraction of the scattered light is omitted. Usually, the larger the particles, the more scattered light is truncated. If this is the case it is recommended to estimate, which fraction of scattered light is lost.

Response to general comment #1

The nephelometer used in the Droplet Measurement Technology is a “reciprocal nephelometer” as the light source and the detector are reversed. The detector in the instrument has a cosine response and captures scattered light between 5° and 175°.

The reviewer has made a valid point regarding the loss of scattered light due to truncation of the detection angle. The nephelometer in the DMT photoacoustic spectrometer is not able to capture forward and backward scattered light. With the help from the manufacturer, this loss due to truncation of the detection angle is calculated, based on Mie theory, to be about 1% for particles of 400 nm in diameter and increases to ~4% at about 1000 nm, ~8% at 2000 nm, and ~14% at 2500 nm.

Particles observed at the Toronto and the two Ottawa sites are expected to be contributed largely by vehicle emissions. Relatively speaking, particles emitted from gasoline vehicles are smaller (<30 nm) than that from diesel vehicles (~ 60 nm), but both are small. These particles are too small to scatter enough light. Based on the above calculations, the losses in scattering signal should be about 1%, i.e., negligible. In the case if samples are mixed with aged background particles, the losses in scattering signal will be much larger.

Current information reported in other BAQS-Met papers and the Windsor AMS data indicate that Windsor measurements were largely influenced by aged particles. In this case the particle diameters can extend beyond 600 nm due to the presence of condensable materials and the effect of atmospheric processing. Depending on the history of the air mass, some particles may have diameter larger than even 1000 nm although size distribution data are not available to prove this. Comparing all the field measurements, we believe the light scattering measurements made in Windsor are less accurate and it may be under-estimated by up to 10% or more. As scattering data is never a major component in this manuscript, the authors decided to remove all scattering data and focus the analysis on the absorption data only.

Reviewer general comment #2

The specific absorption coefficient exhibits a considerable variation for the different types of aerosols, the value of 22.5 m²/g in the suburban station is reasonable. I have my doubts, that the value of 2.6 m²/g in Windsor is not too low. This would mean that the air contains pure carbon spheres with a diameter of at least 0.5 micrometers. For internal mixtures or other shapes the particles even need to be bigger.

Response to general comment #2

The authors appreciate this comment regarding the low SAC values for the Windsor data. Our response here has been informed by our approach to address an important issue raised by reviewer #2 regarding internally vs. externally mixed particles. To gain more insight into this issue we examined individual measurements at the three study areas more closely in order to determine an approach that would best yield information about mixing state and its impact on our results. In the process, we discovered that the data were affected by an improper internal instrument setting that was changed prior to the study when the manufacturer was making repairs and conducting routine maintenance of the photoacoustic spectrometer. When this setting was corrected for in our data there were some changes in the measurements, essentially shifting results towards larger SAC values. Thus, this led to a different range of B_{abs} and SAC values, but relative comparisons, which were the main thrust of our work, were not affected. Due to low BC levels this had the greatest impact on the Windsor data. With these modified measurements, the low mean value referred to above by the reviewer, has been raised, generally ranging from 5-20 m² g⁻¹ depending upon the time period (i.e., the nature of the particles and their corresponding non-refractory mass to black carbon mass ratio). When the non-refractory to BC ratio, which is related to particle coating thickness, increases to its highest values, the resulting SAC also increases to approach a value of 30 m² g⁻¹. These results are more inline with general observations of the enhancement due to non-refractory materials depositing on the BC particles. All the measurements will be updated in the revised manuscript. .

Reviewer general comment #3

When reading this paper I was confronted with a huge quantity of abbreviations, LI, PA, AMS, PPS, SAC, SVF, AMS, just to name a few. I had to make table of abbreviations in order to read the paper. Obviously the authors use these abbreviations in their lab, but this does not mean that all readers are acquainted to it. With a little more compact writing it should be possible to write the full words. If I would not be asked to read the paper, but just looked at it as an interested reader, instead of preparing the table I would have skipped the paper.

Response to general comment #3

The authors have now included a list of all the abbreviations used in the revised manuscript.