

## General comments

The manuscript by Barnard et al. evaluates light absorption by Mexico City's pollution particles sampled during MILAGRO. Single scattering albedo (SSA) at 870 nm is calculated from a size distribution derived from a sunphotometer, mass of several important chemical species measured with a Sunset Lab OC-EC instrument and ion chromatography, and a literature value of refractive index for each component. The results of this calculation, done in two different methods, are compared with an SSA measured using a photoacoustic technique.

I do not recommend publication of this manuscript in its current form, for two reasons. First, the text is unclear and verbose. It took me hours just to be clear on the framework of closure analysis (what is compared against what). One can remove many words from the text while maintaining or enhancing the digestibility of entire text.

Second, there is little scientific merit in the content, as far as I can tell. The SSA closure at a single wavelength at a single location does not contribute a statistically significant input to aerosol models. The multiple sources (measurements, assumed refractive index and morphology) of considerable uncertainties do not help; they make it very difficult for the reader to gain insight as to what factors contribute most to the inconsistency found among the derived SSA values.

## Specific comments

Abstract. Shorten it. For example, "based on a modal approach" is not descriptive. Delete it or rephrase. Be concise throughout the text.

The second paragraph of Section 2.2. The measurement uncertainties (10% for absorption, 15% for scattering) are rather high, particularly for the ground-based measurements with long averaging time interval. This seriously hampers the authors' attempt to evaluate the physical verisimilitude of the WRF-Chem model. Also, explain why the uncertainty in the resulting SSA is about 6%, smaller than those of the underlying measurements. If the individual measurements are subject to systematic errors that compensate for each other, describe them.

The fourth paragraph of Section 2.3, and Figure 3. The use of term "fine mode dust" needs to be justified. This component, which is really a collection of unidentified species, is substantial, sometimes exceeding half of the measured PM<sub>2.5</sub> mass. Unless accompanied by plausible evidence that this mass is indeed dust, this high degree of uncertainty in chemical composition makes it hard to justify the subsequent optical calculation, particularly if a non-zero imaginary component of refractive index is assigned to this unidentified component.

Table 2. Clarke et al.'s study is cited as a reference to the dust density and refractive index. I find this inappropriate. Clarke et al. concluded an imaginary part of refractive index being  $0.0006 \pm 0.0001$ , significantly lower than the authors choice of 0.004. Clarke et al.'s study was on Asian dust, which may be different from Mexico's in mineral composition, and was at 530 nm, not 870 nm. Also, Clarke et al. assumed a density of  $2.5 \text{ g/cm}^3$ , but never established it based on evidence, making the reference paper

an ineffective support for the authors' choice of density 2.6 g/cm<sup>3</sup>. Suggest removing that reference, or using their choice of refractive index.

The paragraph after equation (1). The neglect of coarse mode needs better justification. There is a gap in the upper size of "fine mode" between Dubovik et al.'s (1.2 μm) and the authors (2 - 3 μm aerodynamic which is higher than 1.2 μm geometric for most particles). My own calculation shows that a 2 μm aerodynamic diameter is equivalent to 1.5 μm geometric for a density of 2.6 g/cm<sup>3</sup>, and to 1.7 μm geometric for 2.0 g/cm<sup>3</sup>. The particle mass between the two size cuts may indeed be significant. I suggest showing an estimate of neglected mass and its ratio to the total PM<sub>2.5</sub> mass.

Sections 3.2, 3.3 and Figure 6. The unimpressive agreement between the model and observations certainly need explanations. Yet I failed to find a satisfactorily quantitative one in the subsequent section. In particular, the uncertainty analysis in terms of aerosol size is utterly missing. Even if the mass of each component was perfectly correct, an error in size alone could explain tens of percentage point differences in absorption (see, for example, Figure 4 of *Bond and Bergstrom [AS&T 2005]*), larger than +/- 10% uncertainty discussed in relation to a choice of density.

Table 3. Explain how the refractive index of non-absorbing components,  $n(\text{OM})$  and  $n(\text{inorganic})$ , affects the calculated absorption ( $\Delta\text{Babs}$ ).

The last sentence of Section 3.3. Please clarify how this sentence is consistent with "at the T1 site, dust appears to be the major factor that influences scattering", a statement in Section 3.2.

The second paragraph of Section 3.5. It is not clear to me how the refractive index for fine mode was determined to be  $1.55+0.04i$ . Please clarify.

The last paragraph of Section 3.6. To avoid over-generalization among the readers, it should be noted here that the conclusion is valid for the measurements at a single wavelength as a single location. The validity of the model calculation at other wavelengths or in other locations remains to be established. It should also be noted that the CTOM agreed with the photoacoustic observations on a daily average basis, not generally at higher temporal resolution.

### **Technical corrections**

Section 1.1. The term "editorially" is misused.

The second paragraph of Section 2.4. "a least" should read "at least".

The sentence starting with "Uncertainties, such as those for BC and OM are obtained". Insert a comma after "OM".