Interactive comment on “Photophoretic spectroscopy in atmospheric chemistry – high sensitivity measurements of light absorption by a single particle” by Nir Bluvshtein et al.

Anonymous Referee #1

Received and published: 18 March 2020

The authors report a photophoretic technique as applied to levitated droplets in a double-ring electrodynamic balance. They show that the droplet position changes slightly when slightly light-absorbing molecules are present in the sample, due to the indirect photophoretic effect. By relating the change in DC balance voltage to the change in position, the authors are able to determine the complex part of the refractive index (cRI). The authors demonstrate the retrieval of the cRI of carminic acid in a PEG400 solution at 0.23wt% and report agreement within error to their bulk measurements.

This is a nice new technique and demonstrates another method that the cRI can be established from measurements on levitated droplets. However, it appears to me that
the application of this method to light-absorbing properties of atmospheric sample may be limited. Overall, the paper is well-written and falls within the scope of the journal. Clear answers to the following questions/comments would alleviate my concerns over the applicability of this technique:

1. What range of k is this method appropriate for? An upper and lower limit should be estimated to allow readers to gauge the effectiveness of the method in characterizing brown carbon samples.

2. Would this method be equally applicable to molecules that absorb less strongly but that are present at higher concentration?

3. What range of samples/conditions (particularly RH) can this method be applied to? I would assume that any water vapor in equilibrium with the droplet would be affected by light-absorption and thus add additional complexity to the interpretation of the droplet movement with the laser illumination.

4. Does this measurement require the sample be dissolved in a solution with PEG400 (or similar) as a solvent? How could this technique be expanded outside of this solvent system given the need for well-characterized thermal accommodation coefficients and thermal conductivity?

5. How does $\Delta U/U_0$ vary with a non-absorbing sample (pure PEG400 for example)? This is a necessary benchmark to show that there is no measurable effect when no absorption occurs.

6. The main reason to make measurements on levitated droplets rather than bulk samples is that the super-saturated and super-cooled states accessible to aerosol in the atmosphere can be reproduced in a well-controlled lab environment. I would like the authors expand on the applicability of their method to these kinds of samples.

7. What causes the oscillation in the Mie resonance spectra compared to the simulation? Is this from the laser or related to the movement of the droplet with the heating
beam?

8. Please show a figure (or SI figure) of the DC voltage as a function of time as the heating laser cycles on/off.

Thank you for presenting a very interesting new method!