Some responses from A. Clarke to “Reply to Comments” from C. Brock et al..

I appreciate the authors consideration of my earlier comments and indications of changes to be made in the final version. I will just clarify a few points further here with reference to original line numbers identified. New comments in ***Bold Italic*** follow earlier comment (**Bold**) and authors response (*italics*).

L98 Please specify that “….an inlet..” is actually the “ shrouded solid diffusor inlet designed by Clarke (University of Hawaii) and evaluated by McNaughton et al., 2007.”

We will make this change. The origin of the inlet and sampling using it is described in more detail in Brock et al. (2019), which focuses on the aerosol sampling methodology

OK. As an inlet affects all the aerosol data collected I think it needs to be clearly identified in a paper of this scope. Moreover, it should be noted that this inlet must be operated under isokinetic conditions as failure to do so will particularly impact the relative collection of aerosol components associated with the larger sizes.

L220 The treatment of all components as externally mixed sizes would benefit from additional discussion of when this may or may not be a representative approach.

We will amplify this point, but in the interest of space will largely direct the reader to Froyd et al. (2019), who provide both the methodology and an examination of the mixing state of the aerosol during ATom. The PALMS data show that there is always an external mixture present; for example, most of the coarse mode particles are always different in composition than the accumulation mode. There are particles with dust in them and particles with no dust; the same with sea salt, biomass burning, BC-containing particles, etc. There are many internally mixed organic/sulfate particles as well, but they are present externally from particles of these other types.

Yes, I think all these points are well understood. However, perhaps some clarification is needed as Froyd (section 3.2) states “The most abundant classes under most tropospheric environments are the sulfate–organic–nitrate internal mixtures and biomass burning (BB) particles……..” **and goes on to say** “All particle types acquire secondary material such as sulfate, ammonium, organics, and nitrate during atmospheric transport and aging. This secondary accumulation does not change particle assignments, except that heavy coatings may partially obscure unique signatures, resulting in a particle classified as “Other”. For example, a mineral dust particle that contains secondary sulfate, nitrate, and organic material

will still be classified as mineral dust, and the derived dust mass includes the secondary material…….

Such mixtures of aerosol components can alter the optical, humidification response, nucleating ability etc. relative to treating components as being in external mixtures.

L522 Far more robust comparisons with ambient extinction and AOD exist in the literature. Given the numerous and sometimes subtle considerations (Fig 2) for calculated extinction discussed here, I do not see how the agreement or lack thereof in Fig. 6 actually "……..indicates the methodology to calculate ambient aerosol optical properties is sound." It may be sound but better agreements with simpler assumptions exist. This data set is not designed to get AOD closure or even challenge many sources of uncertainty. One worthwhile objective would be to determine what are the most important measurements needed to characterize AOD within a specified uncertainty. Or how well do we need to know all properties to reduce uncertainties to an acceptable level. Assessing the global role of intensive aerosol properties measured would appear better suited to the ATom measurement strategy.

We agree that there are much better ways to perform AOD closure; that was not a goal of the ATom measurements. We believe that our comparisons with the AERONET observations have value nonetheless. Certainly if there were no correlation between the AOD calculated from the ATom slantwise profiles and the AOD measured by "nearby" AERONET sites, this would be a major cause for concern. Only when we calculate the aerosol hygroscopicity and add the coarse mode measurements from the underwing probe does the AOD from the profiles show consistency with the AERONET observations. Again, the purpose of the AOD comparison is to demonstrate that we've properly accounted for the key features of the aerosol that contribute to ambient extinction. We certainly agree that the ATom dataset provides measurements that can be used to evaluate the sensitivity of climate to aerosol properties; that is the intent of providing this dataset for broader use by the community. We are currently working with modeling and remote sensing groups to diagnose discrepancies between remote sensing and in situ measurements at low AOD values (see below) and to evaluate assumptions underlying retrievals and models.

The main purpose is stated above *…"Again, the purpose of the AOD comparison is to demonstrate that we've properly accounted for the key features of the aerosol that contribute to ambient extinction."*
Simply getting "ballpark" agreement of AOD with Aeronet in higher AOD cases does not indicate what has been properly accounted for.
What are the "key features" referred to and how are they "properly accounted for"?

It would be helpful to take a profile and show the contributions of the various measured aerosol components (and associated water) to the calculated aircraft AOD so the reader has a sense of what is being "properly accounted" for and which data is most important.
Does the calculated spectral AOD agree with Aeronet spectral AOD? – a key feature that is not extensive.
Regarding low AOD comparisons, it is probably worth noting that AOD contribution from above aircraft max altitude can often be 0.01 (or more).

L800-803 "To our knowledge this is the first….."----- This claim is not correct! …Etc.

The authors include the following in their response.
This dataset gives constraints for global models that have not previously existed. For example, we provide a size distribution for dust particles that can be directly compared with that carried in models (in most models dust mass is predicted and the size distribution prescribed). We provide an estimate of the contribution of these dust particles to ambient extinction and AOD; again, this can be compared directly with models. We do the same for sea salt, biomass burning particles, sulfate/organic mixtures, and even meteoric particles of stratospheric origin. No other data set does this; the comprehensive, self-consistent description of the size-and-composition-resolved aerosol properties in ATom is absolutely unique.

I believe these assertions still appear to be an overstatement.
For example, the size distributions of dust and black carbon are similar to those described previously for use by modelers along with their optical properties (*See Clarke et al., Size distributions and mixtures of dust and black carbon aerosol in Asian outflow: Physio-chemistry and optical properties. JGR, 109, 2004 and references therein)*

Also, sea salt size distributions and size-resolved fluxes have been characterized for modelers in Clarke et al. *"An ultrafine sea-salt flux from breaking waves: Implications for cloud condensation nuclei in the remote marine atmosphere [https://doi.org/10.1029/2005JD006565]* ***and references therein as well as other papers by various authors.***

Internal mixing is also common and well documented for most species with varying contributions as a function of size. The Froyd 2019 paper discusses a lot of assocated features of the data. However, some direct comparisons that reveal the differences and or improvements afforded by the ATom data sets over previous data should be presented that support the claims above. A proper comparison and assessment would probably require a separate paper focused on this topic.