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Reply to comment by Prof. Tony Clarke

Charles A. Brock et al.

Author comment on "Ambient aerosol properties in the remote atmosphere from global-scale in-situ measurements" by Charles A. Brock et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-173-AC1, 2021

The authors thank Prof. Clarke for his constructive comments on the manuscript. His comments are in **bold**; our responses are in *italics*.

L49 Please specify the exact "…database commonly used in global models…"

We will change the Abstract to identify the OPAC database against which we compare our observations.

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.

L184 Can authors clarify what is understood as "significant concentrations" criteria for cloud free air as stated "..absence of significant concentrations of droplet or precipitation size particles."

In the revised manuscript we will state the criteria used to identify cloud particle contamination. This methodology is the topic of a manuscript in preparation (Dollner et al.).

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.

L232 "We substitute negative AMS values with zeros when calculating optical or hygroscopic properties." Does this have the effect of reducing uncertainties in these calculations or other consequences?

When AMS mass concentrations for species are very low, as is the case for organic aerosol (OA) for much of the remote free troposphere during ATom, negative mass values can be recorded (e.g., Hodzic et al., 2020). These values represent noise in the measurement. It is appropriate to include these negative values when calculating averages of some extensive parameters such as mass. However, for the purpose of estimating some intensive parameters such as refractive index or the hygroscopicity parameter (kappa) it is not possible to include negative values. Since negative values indicate individual observations of concentrations statistically indistinguishable from zero, for parameters where it is necessary we substitute zeros. In these cases, the refractive index and kappa value would be governed by other species that are present in significant concentrations (e.g., sulfate and ammonium).

L273-275 Provide reference for "few percent" under typical aged plume conditions as seen by ATom and common coating thickness.

Coating thicknesses for BC are provided in Table S5 in the supplemental materials, along with geometric mean diameter and standard deviation for different airmass types. This table will become S6 in the revised manuscript. Globally averaged, the number fraction of BC-containing particles (for accumulation-mode particles with diameters >60 nm) was 3.1% outside of biomass burning plumes and 8.2% within identified plumes. We will include this information in the revised manuscript, possibly in a supplemental table sorted by region, which could be useful.

L473-479 Perhaps an examples of such profile should be included that illustrates application of these requirements. Some additional information would be useful

We feel that the requirements are clear as stated in the text. Adding another figure would only lengthen an already long manuscript.

Approximately what fraction of profiles are excluded by imposing these conditions?

About 26% (162 of 625) profiles were excluded. We will add this information to the text in the revised manuscript. (Note that the total number of profiles is revised to 625 from 640; some of the profiles counted in the submitted manuscript were over continental North America and will be excluded in revision.)

Are there some regions that meet these criteria more commonly than others?

We will create a new table in the Supplemental Materials that provides this information. The table is reproduced below. The Arctic and the Southern Ocean had the lowest fraction of profiles that met the criteria, primarily due to inability to descend the lowest 1 km layer because of clouds.

Table S5. Total number of profiles/number of profiles meeting AOD criteria[1].

Region	*ATom-1*	*ATom-2*	*ATom-2*	*ATom-4*
Arctic	*24/14*	*26/18*	*25/12*	*27/20*

Pacific N. Midlatitude	*9/7*	*31/22*	*33/20*	*35/26*
Pacific Tropics	*39/30*	*32/30*	*29/24*	*22/21*
Pacific S. Midlatitudes	*21/17*	*16/13*	*23/11*	*22/19*
Antarctic/S. Ocean	*5/4*	*6/3*	*21/10*	*17/9*
Atlantic S. Midlatitudes	*10/10*	*4/3*	*11/10*	*17/11*
Atlantic Tropics	*17/16*	*16/12*	*20/17*	*15/15*
Atlantic N. Midlatitudes	*8/5*	*11/9*	*20/15*	*13/10*

[1]Total number of profiles includes only those profiles over the ocean extending from the boundary layer to at least 8 km of depth.

There is also an implicit assumption that any layering in AOD is constant over the horizontal extent of a profile (not very common) but there is no lidar data etc. to confirm that. For example even in clean Southern Hemisphere regions and AOD dominated by sea-salt and water there is appreciable variability in meso-scale AOD (Shinozuka et al., JGR, 2004) and wind speed (and fetch) driving the sea-salt and water component.

The en-route vertical profiles made during ATom were not intended to determine AOD. Spiraling profiles, bounded by column integrated measurements using sunphotometers and lidars (e.g., as performed during DISCOVER-AQ), are necessary to do a proper job of column radiative closure given the mesoscale variability in aerosol properties in the boundary layer, the presence of discontinuous aerosol layers, and other effects. However, the AERONET sites nearest the ATom measurements provide an opportunity for a "sanity check" of the many calculations used to derive ambient aerosol optical properties. Further, with hundreds of profiles, biases in the ATom dataset relative to the AERONET observations should become evident. The slope of the regression, and the surprisingly high correlation coefficient (given the spatial complexity suggested by Shinozuka et al.), indicate that the ambient aerosol optical properties are being calculated without substantial errors (except for a possible bias at low AOD values discussed below). We will discuss these limitations and assumptions in the revised manuscript.

Regarding lidar, the DC-8 was quite full. All available space was occupied by aircraft spares, aircrew seats, compressed gas cylinders, instrument spares, and whole-air-sampler flasks. We definitely agree that any future global-scale mission with a stronger focus on aerosol optical properties and processes should operate a high spectral resolution lidar as a key component of the payload. Ambient, open-path optical properties

(instruments just now being developed) would also be very valuable.

L510+ Comparison with AERONET if profile is within 300 km is generally not going to be accurate due to commonly observed atmospheric variability over these scales. Needs more discussion regarding strategy here in the section labeled "Limitations of the Atoms Data Set."

We agree that we should discuss the issue of how the slantwise vertical profiling can affect comparisons with remote sensing measurements of AOD, adding noise but not sampling bias (see response to preceding comment). We were actually expecting worse agreement given the distance and the spatial inhomogeneity you mentioned. We will amplify this discussion in the revised manuscript.

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.

L523+ and Fig. 6

Fig. 6 Regression line should not be forced through zero (or at least include and discuss both forced and non forced regressions) and the few high AOD cases here are "the tail that wags the dog". For the majority of cases (AOD < 0.1) there are large disagreements with some greater than a factor of two. Aircraft uncertainty bars seem larger than expected.

In the revised manuscript we will provide a regression line that is not forced through zero, as well as a log-log plot. You are correct that there are biases between the ATom and AERONET data at low AOD values. These biases at low AODs also appear in comparisons we are now undertaking with satellite-based AOD data products. We are investigating the potential causes of these biases; it's not yet clear if they lie with the ATom data or the remote sensing methods. Regarding the aircraft uncertainty bars, they are appropriate estimates based on evaluations of specific cases. We take number size distributions, map

bulk and single particle compositions to them, calculate hygroscopicity, add underwing probe data, and use Mie theory to calculate an ambient extinction, then coarsely integrate it vertically using slantwise profiles. So naturally the uncertainties are fairly large! A proper accounting of uncertainties for all data points would require Monte Carlo simulations of the full range of variations of the uncertainties in each of these parameters, which is far too computationally expensive to perform on each of >24,000 data points.

Prior careful Southern Hemisphere clean region profile comparisons to AOD (and AERONET) highlight importance of meso-scale variability and windspeed etc. (see Shinozuka et al, JGR, 2004).

We fully agree that AOD closure experiments must be done much more carefully. ATom is not an AOD closure study; we have merely performed a "sanity check" using the closest available AERONET sites.

Fig 7. These data points need uncertainty whiskers added. Given uncertainty in Fig. 6 it is hard to know what to make of the variability in this data shown on a log scale. Some discussion and comparisons to other references appear warranted here.

We will modify Fig. 7 to include uncertainty estimates, and expand discussion and comparisons.

L635+ and Fig. 12. Please note the actual number of profiles used (and excluded) that were used to generate each of the panels shown in Fig. 12.

We will note the number of data points in each altitude range of each panel. These regional averages were not produced by averaging together the separate profiles that were used to calculate AOD; rather, all data falling in a 1-km altitude range--even those recorded in level flight--were averaged together for that airmass type. We will clarify this in the revised manuscript.

It should be noted that a single 30min profile flying at 450m/s covers about 800km on the ground. The tropics are not a closed system. Hence, horizontal advection in multiple layers below the aircraft is the norm on such scales and usually varies with altitude while advecting at rates far greater than subsidence. Hence, it is problematic to interpret subtle changes in mean values for multiple profiles as an indication growth without other data that can support it.

We are attempting to show that the general features of smaller particles at high altitudes and increasing diameters towards the surface are present in the dataset, as one would expect. These general features are anticipated based on the work of Clarke et al. (1992) and many subsequent observational papers by Clarke and others, as well as modeling studies (e.g., Yu et al., 2010) that show new particle formation associated with convective outflow and subsequent condensational and coagulational growth in descending dry air. This is all complicated by horizontal transport from various sources, as you indicate here. But averaged over many profiles over four seasons, the expected general features are present in the median fitted lognormal diameters, if more subtly than expected. We absolutely agree that more analysis is needed to quantitatively evaluate any of these processes, and this was done for Williamson et al. (2018) and Kupc et al. (2019); the big picture is the focus here. We will add a caveat to that effect where Fig. 12 is discussed.

L665+ I am not clear on what argument or process is being claimed here. "This decrease……..with increasing altitude."

See response to the preceding comment above. We will clarify in the revised manuscript.

L682+ Yes, it would be difficult in this paper to try and compare ATom data to "the extensive literature on global aerosol microphysics….". A comparison to OPAC makes some sense since it is widely used. However, a comparison to shipboard data does not seem particularly useful.

For most of ATom, the column-integrated aerosol properties are dominated by aerosol characteristics in the marine boundary layer. The shipborne measurements highlighted by the Quinn et al. paper, covering much of the same latitude range as the ATom measurements over the Pacific, Southern, and Atlantic Oceans, are thus extremely relevant to the ATom dataset. We dipped into the MBL repeatedly every few degrees of latitude, spending ~5 minutes in a MBL "run" between vertical profiles. We are also trying to sample coarse particles using a combination of in-cabin measurements behind the Clarke inlet and an underwing probe, so it's especially relevant to compare the coarse-mode properties we derive with the more extensive measurements reported by Quinn et al. Consistency between the shipborne measurements and those made on ATom are thus extremely useful in evaluating the representativeness of the airborne data in this challenging environment. This consistency also highlights the discrepancy between these observations and the OPAC database; we're not the only ones to see this discrepancy. There's a problem here with model assumptions that needs to be addressed.

Admittedly there are a large number of comparisons to other data sets that might be made for various purposes. However, there is the extensive aerosol profile data by Clarke and Kapustin (Science, 2010) for many similar regions sampled by ATom. Much of this is flown on the same DC-8 platform, using the same inlet system and with similar instrumentation. That paper synthesizes eleven global campaigns and about 1000 vertical profiles to address issues of aerosol size, nucleation, optical properties, CCN etc. and include the objective of providing input for modelers etc. (see supplementary material). Some reference to this work should be made and possible selected comparisons could be considered.

We agree and will reference and compare to relevant findings from the Clarke and Kapustin paper. This comparison and discussion will be limited given the length of the manuscript and its introductory nature.

L726 I think the heading "Limitations of the ATom dataset" sounds more prejudicial then necessary. The data is what it is. Perhaps something like "Use and Application of the ATom data set" would work with a lot more effort spent in the text on guiding modelers and others in its effective use.

This very good suggestion will be incorporated in the revised manuscript. More explicit guidance to modelers is definitely warranted.

L770-775 I recognize that detailed uncertainty analysis is beyond the scope of this paper. I assume it will be a part of other papers analyzing the data. However, Figures 6 and 7 raise some concerns noted above. Here it is claimed that Fig. 6 suggests that accumulated errors in ambient extinction are <30% but it appears that a significant number of points would fall outside a 30% deviation from the regressed line. More discussion of this is warranted.

We agree that more discussion of uncertainties is warranted. We are working on Monte Carlo simulations of uncertainties for specific, representative cases. It is challenging to independently verify the uncertainties however. The AOD comparisons are really the only data available to work with, and as you have pointed out, it's difficult to make direct evaluations of accuracy based on these given the distance and spatial inhomogeneities. We appear to be underpredicting AOD at low values of AOD. However, remote sensing

experts we have spoken with are less confident in their own measurements at low AOD, so it's not clear where the biases lie. We are actively working with partners to identify the source of these evident biases (whether in the ATom dataset or in the remote sensing data). We will highlight these issues but they won't be resolved in this manuscript.

L796++ The initial findings described here are a disappointment given the potential capabilities described for the instrumentation. These are all basic observations that have been well established in numerous global measurements and will be common knowledge for most readers interested in this paper. These "findings" are not a justification for a program of this scale and a greater focus on the characterization of the intensive aerosol properties seems warranted and of interest.

ATom had a number of objectives; the aerosol objectives were considered secondary and did not motivate the conception or execution of the project. The purpose of this manuscript is to describe the methodology, give a basic overview of the observations, and provide an entry point to the aerosol dataset. The manuscript is already 66 pages long, plus supplemental materials. It is the starting point for further analysis. In addition, we dispute that these "basic observations have been well established". For example, Fig. 5 provides a detailed breakdown of the contribution of different aerosol types to ambient extinction, on a global-scale basis. This is extremely useful new information for modeling groups attempting to calculate direct radiative effects in different environments. Further, the mapping of particle compositional information to size distributions to provide a complete description of the size-dependent composition of the aerosol from 10s of nm to 4 μm in diameter is absolutely unique; the technique was introduced by Froyd et al (2019).

L800-803 "To our knowledge this is the first….."----- This claim is not correct! The Clarke and Kapustin 2010 Science paper mentioned above synthesizes eleven airborne campaigns of very self-consistent global airborne profile data for use by scientific community (see supplementary data.) Moreover, in addition to the synthesis in that paper, the archived NASA and NSF data sets it references are all available and merged with gas, aerosol size and composition, radiative properties, meteorological and aircraft data etc.. Many also include continuous lidar (up/down) that allows visualization of the 2-D atmospheric curtain (eg. aerosol, ozone) that the plane is sampling.

We do not dispute that the synthesis of the extensive measurement campaigns described in Clarke and Kapustin (2010) is an extremely valuable and useful global-scale dataset. But what we have done here is something different: we have combined multiple instruments into a single, comprehensive description of the aerosol (e.g., Fig. 11). That is, we combine the composition and size distribution measurements together to calculate all the relevant properties of the aerosol, from CCN concentrations at arbitrary supersaturations to the contribution of biomass burning particles to optical extinction. (The only portion of the dataset that is not fully integrated with these composition-resolved size distributions is the black carbon and brown carbon measurements. We simply cannot add absorbing components to the composition-resolved size distributions that, when integrated, provide the BC mass and absorption and the BrC absorption uniquely. It is badly underconstrained.)

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.

Further, the ATom dataset does this in deployments executed over relatively short intervals. The advantage of this approach is that models can use prescribed or nudged meteorology to directly sample the model domain for the locations and time periods over which the ATom dataset was obtained. Values that are averaged over multiple campaigns and years, as in Clarke and Kapustin (2010), can only be compared by models using climatologically representative meteorology. Individual campaigns can certainly be compared with model runs using prescribed meteorology, but these individual campaigns are not global in scale (other than HIPPO, which had only black carbon aerosol measurements).

So we stand by our claim that this ATom dataset is unique in nature. We will expand on why this is the case, and how they differ from previous datasets such as those by Clarke and Kapustin, in the revised manuscript. Again, we don't dispute that these earlier datasets are extremely valuable and extensive; this ATom aerosol dataset is just a different beast, designed from the start to be optimally comparable to models by integrating measurements from multiple instruments into a single, composition-resolved description of the externally mixed aerosol.

L803-806 I do not think "Snapshots" really describes the nature of the data and no aircraft campaign can really provide a climatology. Only the synthesis of model and satellite data can do that. Aircraft measurements can help ensure the interpretation of these products is consistent with observations. Greater effort describing how to use ATom data for that objective is warranted.

We agree that it's difficult to describe how these data represent a specific state of the atmosphere. They are not a climatology, but rather the condition of the global atmosphere over the ~23 day period it took to make a circuit. "Atmospheric Tomography" certainly describes the goal of the project; a cross-sectional picture of the state of the remote oceanic atmosphere in a given season. Given the "tomography" analogy, we feel that "snapshot" represents the objective of ATom, even though it's not a truly instantaneous picture. And this terminology emphasizes that a comparison with model climatologies is generally not warranted; specific meteorology must be invoked. We definitely concur that a synthesis of model and satellite data is necessary to place the measurements in their proper context. This manuscript is only the starting point for much more extensive analysis.

Concluding comments:

This provides a good overview of ATom data. Suggesting and discussing strategies for using the ATom data would be helpful for modelers and others. The authors do suggest modelers "nudge times" to coincide with the data sets and suggest "....the model domain be sampled along the flight track". This is probably essential for any direct comparison of aerosol "extensive" data to models but is far more demanding than summarizing mean profile values etc. An example on the scale of ATom data is the comparison of DC-8 NASA PEMT mission aerosol data over the Pacific (Clarke et al., JGR, 2001) with the MATCH assimilation chemical transport model (Collins et al., 2001).

Thank you very much for your constructive comments on the manuscript. We appreciate your expertise and global perspective. We expect that this dataset, together with already-developed datasets such as those provided in your earlier work and others, some of which

are compiled by the GASSP program (Reddington et al., 2017), will be useful in understanding atmospheric composition and constraining key sources and processes within global models. This manuscript represents the starting point for much more extensive and detailed analysis along the lines you suggest. We will amplify the need for further analysis in the revised manuscript.

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