Reducing Aerosol Forcing Uncertainty By Combining Models with Satellite and Within-the-Atmosphere Observations: A Three-Way Street

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Key Points:

- Aerosol climate forcing uncertainty is virtually undiminished despite two decades of advances in many aspects of aerosol-climate science
- This review concludes that current and planned aerosol modeling, satellite and ground-based observation programs remain essential
- New, systematic aircraft aerosol particle and cloud process measurements are also needed, along with better model-measurement integration

Abstract

Aerosol forcing uncertainty represents the largest climate forcing uncertainty overall. Its magnitude has remained virtually undiminished over the past 20 years despite considerable advances in understanding most of the key contributing elements. Recent work has produced modest increases only in the confidence of the uncertainty estimate itself. This review summarizes the contributions toward reducing the uncertainty in the aerosol forcing of climate made by satellite observations, measurements taken within the atmosphere, as well as modeling and data assimilation. We adopt a more measurement-oriented perspective than most reviews of the subject in assessing the strengths and limitations of each; gaps and possible ways to fill them are considered. Currently planned programs supporting advanced, global-scale satellite and surface-based aerosol, cloud, and precursor gas observations, climate modeling, and intensive field campaigns aimed at characterizing the underlying physical and chemical processes involved, are all essential. But in addition, new efforts are needed: (1) to obtain systematic aircraft *in situ* measurements capturing the multi-variate probability distribution functions of particle optical, microphysical, and chemical properties (and associated uncertainty estimates), as well as co-variability with meteorology, for the major aerosol airmass types; (2) to conceive, develop, and implement a suborbital (aircraft plus surface-based) program aimed at systematically quantifying the cloud-scale microphysics, cloud optical properties, and cloud-related vertical velocities associated with aerosol-cloud interactions; and (3) to focus much more research on integrating the unique contributions satellite observations, suborbital measurements, and modeling, in order to reduce the uncertainty in aerosol climate forcing.

Plain Language Summary

Aerosols, such as airborne wildfire smoke, desert dust, volcanic and pollution particles, affect Earth's climate by reflecting (some also absorb) sunlight. These aerosol particles also play key roles in cloud formation and evolution, further affecting the planet's energy balance. The magnitudes of these effects, and even the underlying mechanisms, represent the largest uncertainty in climate modeling. Despite two decades of advances in many aspects of aerosol-climate science, aerosol climate forcing uncertainty is virtually undiminished. Yet, reducing this uncertainty is critical for any effort to attribute, mitigate, or predict climate changes. We adopt a

65 measurement-oriented perspective to assess the strengths and limitations of measurement and
66 modeling programs, and conclude that current and planned efforts need to continue. However, in
67 addition, new efforts are needed: (1) to obtain aircraft *in situ* measurements that capture
68 systematically aerosol particle properties for the major aerosol airmass types, globally, (2) to
69 conceive, develop, and implement an aircraft and surface-based program aimed at filling gaps in
70 our understanding of the interactions between aerosol particles and clouds, along with (3)
71 focusing much more research on integrating the unique contributions of satellite observations,
72 suborbital measurements, and modeling, to reduce the uncertainty in our understanding of
73 Earth's changing climate.

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1 Introduction

The confidence with which Earth's present and possible future climate can be simulated depends upon our ability to represent the factors that heat and cool the system, and the processes that mediate the environmental response. In this context, radiative forcing describes the energy fluxes that drive the climate system. Changes in the radiative forcing of climate are caused primarily by perturbations of atmospheric constituents, mainly greenhouse trace gases (GHGs), airborne particles (aerosol particles), and clouds. By convention, positive forcing produces net surface heating, whereas negative forcing produces cooling. Understanding changes in the radiative forcing of climate is critical for any effort to attribute, mitigate, or predict climate change.

Although GHGs contribute most of the positive radiative forcing, its magnitude is tightly constrained, so uncertainty in the climate forcing by aerosol particles dominates the uncertainty in forcing changes overall (e.g., Forster et al., 2021; Watson-Parris & Smith, 2022). Globally, aerosol forcing is generally negative. As one illustration of the importance of reducing aerosol forcing uncertainty, decreases in aerosol amount in some regions can lead to increased surface heating (Jenkins et al., 2022; Quaas et al., 2022). Thus, understanding the factors driving climate change, and improving our ability to predict climate changes under different future scenarios, requires a reduction in the uncertainty in aerosol climate forcing. This includes both the direct radiative forcing due to light scattering and absorption by airborne particles, as well as indirect effects due to the interactions between aerosol particles and clouds (ACI).

There are fundamental reasons why aerosol radiative forcing is more difficult to quantify than forcing from GHGs. First, present-day aerosol forcing is often assessed relative to assumed pre-industrial conditions, which are presumed to be largely unaffected by human influences (e.g., IPCC, 2013). Yet, there are few observational constraints on the pre-industrial aerosol state (e.g., Carslaw et al., 2017). For example, for aerosol amount there is no analog to the ice core data that defines pre-industrial GHG concentrations, and unlike gases, aerosol microphysical properties vary greatly. Further, even for the present day, the distinction between natural and anthropogenic aerosol is often ambiguous. Is wind-blown dust to be considered natural or anthropogenic when it is emitted in regions experiencing desertification due to increased use of water resources, over-grazing and other farming practices, or some combination of larger-scale environmental changes

of uncertain natural or anthropogenic origin? The same question applies to smoke aerosol from lightning-ignited wildfires that might be greatly intensified by anthropogenically enhanced warming, drying, or forest management practices. Also, in practical terms, remote-sensing data alone cannot be interpreted precisely enough in most cases even to distinguish natural aerosol from aerosol that are unambiguously anthropogenic when directly sampled (see Section 2 below). These issues with historical data and the attribution of today's emissions contribute to large uncertainty in the difference between aerosol forcing under both pristine and present-day conditions (Carslaw et al., 2013; Hamilton et al., 2018).

Note that the total direct radiative forcing by aerosol includes natural sources, representing the dominant contribution to the global atmospheric aerosol mass load, plus anthropogenic aerosol. The Intergovernmental Panel on Climate Change (IPCC) calls the total quantity the aerosol direct radiative effect, and refers to the anthropogenic component as the aerosol direct forcing. Given the ambiguities from an observational perspective, we use the term "aerosol forcing" to include anthropogenic and natural direct and indirect forcings and adjustments; we specify direct or indirect where needed.

A second reasons why aerosol radiative forcing is more difficult to quantify: aerosol particles are vastly more spatially and temporally heterogeneous than GHGs, due to relatively short atmospheric lifetime, sources that are highly non-uniform in space and time, and particle physical and chemical evolution that varies with atmospheric conditions. As such, frequent, global observation of aerosol distribution and key properties must be provided to capture this heterogeneity. Not only is aerosol amount highly variable at different locations and times, aerosol light scattering, light absorption, and cloud nucleating properties vary considerably with source, composition, and particle evolution. As we discuss in Section 2, these factors cannot be determined adequately from remote sensing alone.

Finally, aerosol particles produce relatively large adjustments to forcing compared to GHGs. That is, aerosol particles have secondary climate impacts, such as changing the atmospheric stability profile or altering cloud radiative properties, lifetime, and precipitation. These semi-direct (ambient heating by light-absorbing particles), indirect (e.g., increased particle concentration causing additional cloud droplet or ice crystal formation), and other effects

represent aerosol-induced changes in the atmosphere that can cause changes in surface temperature indirectly. For example, vertical redistribution of aerosol extinction can alter the atmospheric thermodynamic structure, and atmospheric adjustments can even change the sign of black carbon forcing in the upper atmosphere compared to the surface (e.g., Lau et al., 2008; Samset et al., 2013), whereas for major GHGs such as CO_2, adjustments are estimated to represent much smaller fractional contributions (Forster et al., 2021). A combination of measurements and modeling is required to constrain adjusted forcing.

Reducing the magnitude of aerosol forcing uncertainty to a value comparable to that of CO_2 and other long-lived GHGs (estimated at ± 0.35 W/m^2 and ± 0.23 W/m^2, respectively; IPCC, 2013) will require greater observational constraints than near-future satellite measurements alone are likely to provide. A lower bound on the uncertainty range of just the direct component of the total global-mean all-sky aerosol forcing (i.e., that due to scattering and absorption by airborne particles but not their indirect effects on clouds) is estimated as ±1.1 W/m^2 when considering anticipated data from a planned next-generation NASA satellite mission aimed at addressing global climate change issues (1σ; Thorsen et al., 2021). (This number represents 30% or more of the global all-sky aerosol forcing by most estimates.) Systematically incorporating prior information beyond that available from current or near-future large-scale observations is necessary to reduce uncertainties in the role aerosol particles play in climate forcing. Thorsen et al. (2021) suggest that incorporating additional particle property constraints might approximately halve the direct forcing uncertainty estimate, though the even larger uncertainty associated with the indirect effects of aerosol particles on clouds is more difficult to reduce or even to estimate. Thus, to reduce climate modeling uncertainties, there is a crucial need for better particle optical, microphysical, and chemical property information than can be provided by satellite measurements alone. This persistent uncertainty is reflected in part by the enormously diverse, yet often simplistic, particle property assumptions made in different models (e.g., CCSP, 2009). Such diversity is also true of the aerosol optical model priors (i.e., initial guesses) or climatologies adopted as constraints in leading satellite aerosol retrieval algorithms (e.g., Remer et al., 2005; Levy et al., 2007; Kahn & Gaitley, 2015; Kim et al., 2018; Holzer-Popp et al., 2013). Further, the uncertainties in aerosol-cloud interactions must be reduced as well.

Previous reviews of aerosol climate forcing tend to adopt a model-centric perspective, appropriately, as models offer the ability to make climate predictions. Such reviews can provide formal estimates of aerosol forcing and its uncertainties based on current modeling, often also taking account of the limited constraints provided by available measurements (e.g., Bellouin et al., 2020). Here we take a more measurement-focused perspective, reviewing where we stand at present on the contributions measurements can make and their relationships with models, allowing us to also identify gaps that need to be filled.

Figure 1 provides a framework for discussing research activities aimed at reducing uncertainty in aerosol forcing, and also helps organize the material presented in subsequent sections. It illustrates the unique and essential roles played by satellite observations, measurements made within the atmosphere (often termed "suborbital" measurements when taking a perspective that includes global satellite measurements), as well as aerosol and climate modeling, in closing the gap in aerosol forcing uncertainty relative to that of GHGs. The arrows and associated, color-coded annotations in Figure 1 highlight the required *exchange of information* between these three elements. (The common expression "a two-way street" means A affects B, but B also affects A. Here, A affects B and C, B affects A and C, and C affects A and B; hence, a three-way street.) From the synthesis provided by this review, we conclude that satellite measurement and modeling efforts and plans for addressing aerosol climate forcing are relatively mature. However, the uncertainty in aerosol impacts on climate directly, and indirectly through their interactions with clouds, could be significantly reduced through specific enhancements to the suborbital component, along with greater synergy among the three elements. As such, we review the satellite and modeling contributions first (Sections 2 and 3, below), then consider the suborbital element in Section 4, and provide a synthesis in Section 5 and the Conclusions. The view presented here is informed in part by discussions in the AeroCom and AeroSat communities over the past decade (https://aerocom.met.no and https://aero-sat.org, respectively; last accessed: March 2023), the work of the Systematic Aircraft Measurements to Characterize Aerosol Air Masses (SAM-CAAM) Science Definition Team (Kahn et al., 2017), and the collective insights of the authors of the current paper.

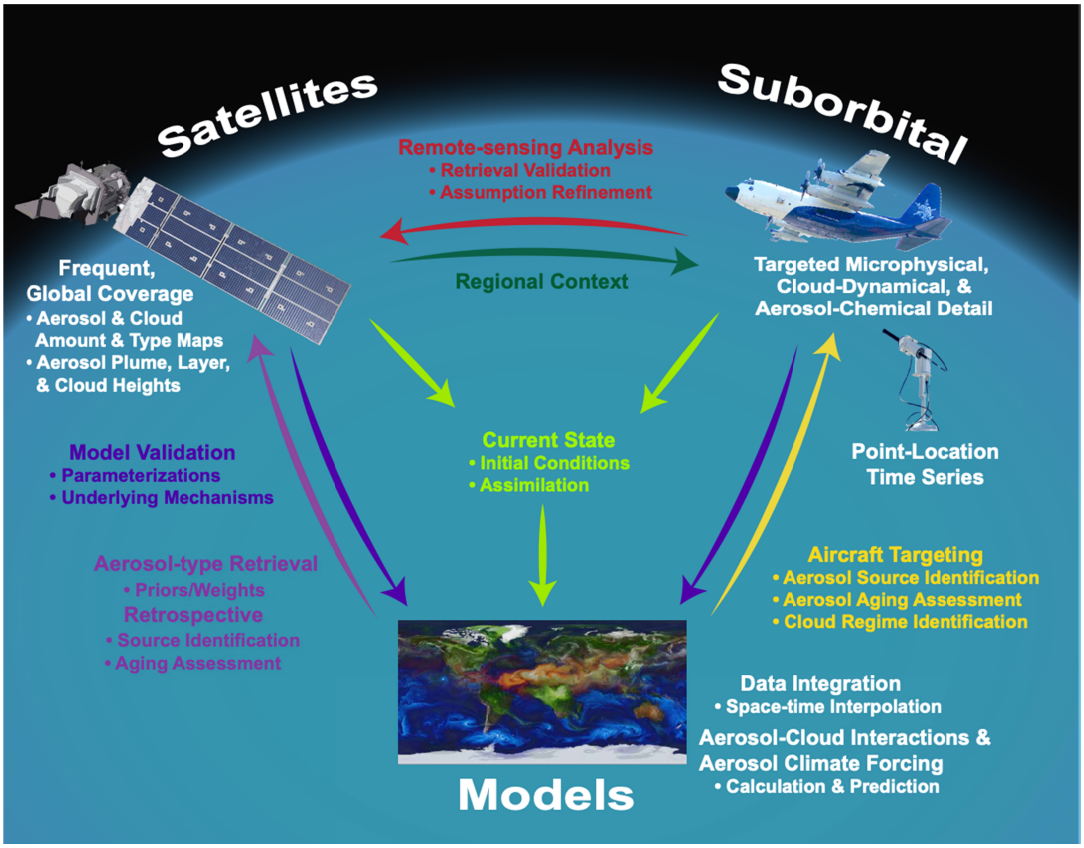

Figure 1. A framework for considering the research activities needed to reduce the persistent aerosol forcing uncertainty. It illustrates the relationships among the measurements, the modeling contributions, and the interactions among these elements that we identify as central to this effort (adapted from Kahn, 2012). The arrows and associated, color-coded annotations indicate the contributions each element can make to the other two – a three-way street.

2. Satellite Contributions

Earth-orbiting satellites offer the opportunity to collect frequent, global data, and are well suited to remotely measure quantities that vary on kilometer or even smaller spatial scales, at daily or even shorter timescales.

2.1. Aerosol Amount – Total-Column Optical Depth and 3-D Distribution

For more than 20 years, satellites have demonstrated the ability to provide extensive spatial and frequent temporal coverage of aerosol column amount globally, primarily under clear-sky conditions, and generally reported as the mid-visible aerosol optical depth (AOD). This quantity is widely used as a constraint on climate models, or for model evaluation (e.g., Kinne et al., 2006; Buchard et al., 2017; Gelaro et al., 2017; Randles et al., 2017; Rubin et al., 2017; Schutgens, et al., 2020; Gliß et al., 2021), though non-negligible differences among AOD retrieval products must yet be addressed (Sogacheva et al., 2020; Li et al., 2020). Tables summarizing currently available satellite AOD products are given in Sogacheva et al. (2020) and Kahn and Samset (2022). Space-based lidars provide the altitudes of extended aerosol layers with the precision of tens of meters (e.g., Winker et al., 2009; Yorks et al., 2016), and usually sample well downwind of aerosol sources, where the AOD tends to be low enough for the signal to penetrate the column under cloud-free conditions. Lidar measurements are complemented by near-source aerosol plume height from multi-angle imagery obtained in low-Earth or geostationary orbit (Kahn et al., 2008; Nelson et al., 2013; Carr et al., 2022). Such information can help characterize aerosol vertical extent or source injection height in models (e.g., van Donkelaar et al., 2010; Val Martin et al., 2018). Although coverage by these measurements is currently limited, their precision has demonstrated value in constraining and/or validating models aimed at simulating downwind wildfire smoke and volcanic ash dispersion (e.g., Zhu et al., 2018a; Vernon et al., 2018). A number of satellite-based, passive-imager spectral techniques, at wavelengths ranging from the ultraviolet (UV) to the infrared, offer greater aerosol layer-height coverage, at the cost of additional assumptions that increase uncertainty (e.g., Jeong & Hsu, 2008, Griffin et al., 2020; Go et al., 2020; Kylling et al., 2018; Lu et al., 2021; Lyapustin et al., 2020). Passive-imager techniques, especially in UV channels, have also been effective in constraining aerosol occurrence, and even amount, over cloudy scenes (e.g., Torres et al., 2012; Meyer et al., 2015; Sayer et al., 2019).

Models require the source strength in addition to the injection height to represent aerosol sources. Efforts at using satellite AOD measurements to constrain source strength have applied either inverse modeling to infer source location along with strength from broad-swath-imager AOD maps (e.g., Dubovik et al., 2008), or forward modeling, where a measured smoke-plume AOD snapshot is compared to a model, run in the forward direction and initialized with varying source strengths (Petrenko et al., 2017). These methods work best for high-AOD aerosol plumes

with little background or aged aerosol in the scene to complicate interpretation. Other approaches, specifically for wildfires, use the magnitude of the 4-micron brightness temperature anomaly, which can be observed from remote sensing, combined with empirically derived, ecosystem-specific emission factors, to estimate smoke source strength (e.g., Ichoku & Ellison, 2014; Wiggins et al., 2020; see review by Andreae, 2019). Next-generation spacecraft deployments promise to greatly increase coverage frequency and diurnal sampling, with instrumentation such as multi-angle, multi-spectral polarimeter imagers and High Spectral Resolution Lidars offering advanced measurement capabilities (e.g., https://aos.gsfc.nasa.gov, last accessed, June 2022). Further, recent advances in estimating satellite-retrieved AOD uncertainty, even at the individual pixel level, have increased the utility of the satellite AOD measurements as constraints on aerosol forcing (Sayer et al., 2020; Witek et al., 2018).

The largest remaining uncertainties in the measured global distribution of aerosol amount for climate forcing applications are due to (a) difficulties in retrieving AOD over snow, ice, bright or topographically complex land, and cloud-covered surfaces, (b) accounting in satellite retrieval algorithms for the detailed aerosol light-scattering and light-absorption properties, as well as the hydration state of different aerosol types under ambient conditions that affect retrieved aerosol amount, (c) obtaining adequate spatial coverage for aerosol layer-height observations, and (d) specifying the relative-humidity-dependent Mass Extinction Efficiency (MEE) that is required to translate between AOD derived optically from remote sensing and the dry aerosol mass that is bookkept in climate and air quality models. Nighttime AOD is also poorly sampled by passive sensors including broad-swath imagers at present, and it can be important in accounting for the infrared part of the energy balance, especially in areas where airborne mineral dust is abundant. This is probably a secondary issue in representing global aerosol climate forcing, given the uncertainties under daylight conditions (e.g., Adebiyi & Kok, 2020); however, some space-based thermal IR (Vandenbussche et al., 2013; Capelle et al., 2018) and even visible (Wang et al., 2016) remote-sensing techniques hold promise to provide extensive nighttime AOD coverage beyond what is possible with active sensors. As discussed in subsequent sections, appropriate combinations of modeling and suborbital measurements with the satellite observations (Figure 1) appear to be key to addressing the limitations of the space-based AOD data record.

2.2. Satellite Constraints on Aerosol Particle Properties

Satellite observations have up to now been less successful at providing the quantitative particle optical, microphysical, and chemical properties needed to model aerosol forcing. For example, uncertainty in aerosol light-absorption, usually represented by the spectral single-scattering albedo (SSA), is comparable or possibly of greater importance compared to AOD in setting the overall uncertainty in aerosol direct forcing of Earth's energy budget (McComiskey et al., 2008; Loeb & Su, 2010; Thorsen et al., 2021; Li et al., 2022). Along with aerosol vertical distribution, SSA is also critical for modeling the semi-direct aerosol effect, i.e., the aerosol impact on solar-energy absorption and atmospheric heating, along with the attendant effects on clouds.

The ability to obtain quantitative or even qualitative constraints on properties such as particle size, SSA, and sphericity depends on retrieval conditions as well as instrument characteristics. These variables can be derived from multi-angle, multi-spectral observations when the surface is not too complex and the AOD is sufficiently high – typically exceeding 0.2 to 0.5 at mid-visible wavelengths. This is often challenging, as the global average mid-visible AOD is ~0.14 (e.g., Andrews et al., 2017). For example, much of wildfire smoke instantaneous radiative forcing occurs over extended areas downwind, where smoke AOD < 0.1 (Schill et al., 2020). In addition, the sun-spacecraft observing geometry must provide an adequate range of scattering angles for robust results using this technique (e.g., Kalashnikova et al., 2013; Kahn & Gaitley, 2015; Fougnie et al., 2020).

Temporal compositing of single-view sensor measurements acquired at different times with different viewing angles (Lyapustin et al., 2021) and/or different AOD (e.g. Seidel and Popp, 2011; Wells et al., 2012) can also be used to enhance their information content and obtain tighter AOD or SSA constraints. Polarized, multi-spectral multi-angular measurements improve retrieval sensitivity to the real part of the particle refractive index and to the width of the particle size distribution, as well as broadening the range of conditions under which meaningful particle-type constraints can be obtained (e.g., Mishchenko & Travis, 1997; Dubovik et al., 2011; Hasekamp et al., 2011). Despite this, systematic differences in aerosol absorption derived from different algorithms applied to the same polarimetric sensor still exceed the requirements for adequate constraints on aerosol forcing (Schutgens et al., 2021). Although the deployment of new polarimeters with increased polarimetric accuracy (e.g., Werdell et al., 2019) should reduce these discrepancies to some degree, algorithmic assumptions will still be required (e.g., when

layers containing different aerosol types occur in the atmospheric column); as with all new measurements, the quality of the results remains to be demonstrated.

Space-based, two-channel backscatter lidar with polarization sensitivity currently provides a height-resolved classification of six empirically defined aerosol types operationally, using the retrieved depolarization ratio, attenuated backscatter, layer top and base altitudes, and surface type as imput (Kim et al., 2018). Future spacecraft deployment of High-Spectral-Resolution Lidar (HSRL) instruments promises to refine lidar aerosol-type retrieval by constraining, in addition, the light extinction-to-backscatter ratio (e.g., Burton et al., 2012; Hélière et al., 2012; Russell et al., 2014; Dawson et al., 2017), a key aerosol-type-dependent microphysical property that must be assumed when interpreting backscatter-only observations in terms of aerosol amount. The inclusion of an ultraviolet channel in a future space-based lidar instrument for greater particle-type discrimination, especially with depolarization sensitivity, is also possible (Burton et al., 2015; Nicolae et al., 2018; Papagiannopoulos et al., 2018).

Overall, within the AOD and other retrieval caveats discussed above, satellite remote sensing can contribute to constraining direct aerosol forcing by mapping aerosol types qualitatively and identifying differences in particle size and light-absorption. However, to date, constraints from satellites alone on SSA and particle size distribution lack the coverage, the accuracy, as well as the error characterization, required for closing the aerosol forcing uncertainty gap in most environments.

When retrieval conditions are favorable, some general deductions about the mechanisms driving aerosol property evolution have also been gleaned from satellite data, such as near-source in wildfire smoke and volcanic plumes. For example, changes in AOD, and in retrieved effective particle size and light-absorption downwind of major sources, have been used to infer size-selective or size-independent particle deposition, particle hydration or oxidation, secondary aerosol formation, the condensation of volatiles on existing particles, and particle activation processes (Junghenn Noyes et al., 2020; Flower & Kahn, 2020a; b). In addition, having plume-level wind vectors, e.g., derived from multi-angle stereo imaging, makes it possible to estimate timescales for the observed transitions, at least near major aerosol sources. Coincident field data, primarily from aircraft making detailed *in situ* measurements and in some cases from ground

observers, have been essential for building confidence in these inferences made from remote-sensing observations (e.g., Junghenn Noyes et al., 2020). At the same time, extensive, qualitative aerosol property mapping from satellites can place the detailed aircraft measurements, acquired, e.g., along disjoint transects of a smoke plume, into the larger context of plume-particle evolution. This complementary exchange of information is illustrated in Figure 1 (red and dark green arrows). Further, despite the qualitative nature of satellite-based particle-evolution-process deductions, the satellite data also offer broad spatial coverage that allows for multi-year assessment of the dominant particle evolution processes and associated timescales where data quality is adequate (e.g., Junghenn Noyes et al., 2022).

Several approaches have been taken to using satellite measurements and the associated inferences as validation or constraints on models (Figure 1, blue and light green arrows). In addition to statistical comparisons between aggregated satellite observations and models of particle evolution processes and timescales, by configuring models to simulate specific instrument measurements, more quantitative, direct comparisons between models and observations can be made in some cases. The results can be especially useful as a means of diagnosing model behavior, leading to better constraints on model parameterizations, at least statistically. Such work has been performed mainly on daily snapshots from polar-orbiting instruments. However, as the capabilities of instruments on geostationary (Gupta et al., 2019; Lim et al., 2018; Zhang et al., 2020a) and even more remote platforms (Marshak et al., 2018) advance, data from these sources promise to increase constraints on aerosol processes by resolving the time-evolution of particle properties over large areas.

2.3. Strengths and Limitations of Space-based Aerosol-Cloud Interactions (ACI) Observations

From the satellite perspective, reducing the forcing uncertainties due to the indirect effects of aerosol particles on clouds is yet more challenging, given the subtlety and complexity of the aerosol as well as the cloud processes involved, and the range of spatial and temporal scales on which they operate (e.g., reviews by Rosenfeld et al., 2014 Seinfeld et al., 2016; Mülmenstädt & Feingold 2018; McCoy et al., 2016; Quaas et al., 2020; Bellouin et al., 2020). Cloud-formation processes require aerosol particles to serve as cloud condensation nuclei (CCN), except under

very cold, homogeneous-nucleation conditions. More generally, the CCN size spectrum and concentration, particle hygroscopicity, and SSA are all required to model cloud-droplet formation and ACI. However, the mutual interactions between aerosol particles and clouds mean that the challenge is much greater than just measuring CCN, because a host of other cloud microphysical and dynamical processes change ('adjust'), depending in part on CCN properties and concentrations.

A fundamental limit to the contribution satellites can make regarding ACI is that particles smaller than about 0.1 μm in diameter are difficult to characterize with remote sensing; they are very inefficient at scattering light and are essentially indistinguishable from the background atmospheric gas. Yet, such small particles can represent a significant fraction of the activated CCN, particularly under high updraft velocities (e.g., Twomey, 1974; Stier, 2016). Broad-swath, single-view imagers can provide AOD and can constrain its spectral dependence under good retrieval conditions. The extinction Ångström Exponent (ANG), defined as the negative slope of AOD vs. wavelength (both in log space) is inversely proportional to particle size, provided a single aerosol mode dominates the atmospheric column. The product of AOD and ANG (derived from AOD retrieved at several wavelengths), designated the aerosol index (AI), yields a metric that weights AOD toward smaller (CCN-like) sizes (Nakajima et al., 2001). As an estimate of CCN, AI is a qualitative indicator at best, in part because particle hygroscopicity is generally unconstrained by remote-sensing measurements but can dominate the relationship between retrieved AOD and particle number, especially for small particles (Kapustin et al., 2006; Cao et al., 2023), and in part because extrapolating the observed part of the aerosol spectrum to smaller sizes engenders additional assumptions. However, AI is often treated as quantitative for lack of other observational constraints.

At the other end of the CCN size distribution, giant CCN, on the order of several microns in diameter, can have an inordinately large effect on precipitation formation in warm clouds, even at concentrations on the order 1/liter (Feingold et al., 1999), far too low to be retrieved from space-based remote sensing. High concentrations of coarse-mode and giant CCN (~ 1-2 microns in diameter and larger) can also have a disproportionately large impact on cloud supersaturation and susceptibility to aerosol perturbations (e.g., Ghan et al., 1998; Morales Betancourt & Nenes, 2014). As such, giant CCN characterization must rely primarily on suborbital measurement.

Some recent advances in aircraft instrumentation and analysis are improving our ability to characterize giant CCN *in situ* (e.g., Jung et al., 2015; Dadashazar et al., 2017; Gonzalez et al., 2022). Yet, giant nuclei measurement remains challenging, due to issues such as losses in conventional inlets (e.g., Wilson et al., 2004), sizing uncertainties in wing-mounted probes (e.g., Gonzalez et al., 2022), generally low number concentrations, and the need to know the time-history of relative humidity exposure to relate dry and ambient particle properties.

Particle light-absorption is also challenging to constrain quantitatively with remote sensing (Section 2.2 above). Yet aerosol composition, particularly smoke or dust aerosol having low SSA, can play an important role in driving cloud-mediated radiative forcing. Such particles can heat the atmosphere, reducing the ambient relative humidity and causing cloud-droplet evaporation (the so-called semi-direct effect). Further, by affecting the atmospheric vertical stratification, they can either suppress (e.g., Koren et al., 2004) or possibly enhance convection, depending on the location of the aerosol (Koren et al., 2005; Jiang & Feingold, 2006). Well-defined smoke plumes can apparently also alter mesoscale (Lee et al., 2014) and large-scale (Williams et al., 2022) circulation patterns.

Satellite-based mapping of aerosol amount over extended regions is obtained from passive measurements of column-integrated AOD. Attempts at correlating AOD directly with CCN concentration measured *in situ* have yielded only qualitative relationships (e.g., Andreae, 2009; Shinozuka et al., 2015; Shen et al., 2019), and vertical decoupling introduces significant additional uncertainty (Stier, 2016). Of particular relevance for this application, the CCN that actually participate in the cloud-droplet-formation process are typically just below cloud base, a region inaccessible to passive measurements from space. These CCN can be measured directly only with *in situ* aircraft instruments or inferred from surface remote sensing, by inverting cloud microphysical measurements (Feingold et al., 1998), athough indirect methods offer promise for some situations (e.g., Rosenfeld et al., 2016).

Further complicating the satellite contribution to measuring ACI is the complexity of interpreting remote-sensing observations in the vicinity of clouds (e.g., Marshak et al., 2021). This region, sometimes termed the “twilight zone,” contains a continuum of hydrated aerosol, evaporating cloud droplets, and cloud fragments (Koren et al., 2007). To interpret remote-sensing observations, both “cloud contamination” of the aerosol signal and light-scattering by the cloud

411 must be taken into account. Standard retrieval algorithms assume 1-D radiative transfer whereas
412 these situations are inherently three-dimensional; the 3-D radiative transfer that applies here can
413 produce significantly different AOD retrieval results and aerosol forcing estimates (e.g., Wen et
414 al., 2007; Yang et al. 2022). Furthermore, considering the hygroscopic growth of particles in the
415 vicinity of clouds as a continuum means that the aerosol forcing is driven by the 3-D spatial
416 distribution of humidity in the cloud field (which varies on much smaller scales than the aerosol
417 properties (e.g., Anderson et al., 2003)).

418 Atmospheric dynamics plays an important role in determining the effects of aerosol on clouds,
419 and this too is difficult to constrain from space at the relevant spatial and temporal scales, and
420 with the required accuracy. Water vapor supersaturation, responsible for cloud droplet and ice
421 crystal formation, is generated by expansion cooling that is directly proportional to vertical
422 velocity. The fraction of particles that activate to form cloud droplets is often strongly dependent
423 on updraft velocity, although aerosol number concentration, size distribution, and to a lesser
424 extent composition also affect the result (Feingold, 2003; Ervens et al., 2005; McFiggans et al.,
425 2006). Vertical velocity is more important when cloud supersaturation is low; such "velocity-
426 limited conditions" occur frequently around the globe (Reutter et al., 2009; Morales Betancourt
427 & Nenes, 2014; Sullivan et al., 2016). When low supersaturation occurs under high aerosol
428 (polluted) conditions, drop concentration depends on both CCN concentration and vertical
429 velocity (e.g., Kacarab et al., 2020; Bougiatioti et al., 2020; Georgakaki et al., 2021**;** Foskinis et
430 al., 2022); however, factors such as aerosol size distribution and chemical composition can also
431 become more important under these conditions (e.g., Rissman et al., 2004; McFiggans et al.,
432 2006). In aerosol-poor environments such as the Arctic, the factor limiting droplet formation is
433 often the number concentration of CCN, and vertical velocity uncertainty becomes less important
434 (e.g., Feingold et al., 2003; McFiggans et al., 2006). Yet, vertical velocity is rarely constrained
435 by measurements contemporaneous with aerosol amount and properties such as particle size
436 distribution and hygroscopicity (e.g., Kacarab et al. 2020; Bougiatioti et al. 2021). Further,
437 despite its high spatial variability globally (order 10s of meters), vertical velocity only can be
438 measured at the required accuracy and spatial resolution with limited sampling, using active
439 Doppler lidar or Doppler radar from suborbital platforms (e.g., Guimond et al., 2014; Schroeder
440 et al., 2020). At least until next-generation satellite instruments are developed, updraft velocity

441 measurements relevant to drop and ice formation are unlikely to be possible from spacecraft
442 alone.

443 The most widely observed ACI process is cloud brightening, which is caused by an increase in
444 CCN concentration generating an increase in cloud droplet number, and all else being equal, a
445 decrease in droplet size (Twomey, 1974). However, this conceptually simple mechanism is also
446 strongly affected by cloud adjustments (e.g., Twomey, 1977; Quaas et al., 2020). Much work has
447 focused on aerosol-related cloud brightening with all other factors, especially cloud liquid water
448 path (LWP), held constant. However, both LWP and cloud fraction have been shown to exhibit
449 non-monotonic responses to aerosol perturbations. Under clean conditions, increases in aerosol
450 tend to increase LWP, whereas under polluted conditions LWP might be reduced (Wang et al.
451 2003; Ackerman et al. 2004, Bretherton et al. 2007; Gryspeerdt et al. 2019). Similar results
452 appear to be valid for cloud fraction (e.g., Xue et al. 2008; Gryspeerdt et al. 2021). Although
453 some in-cloud processes can be inferred from satellite observations (e.g., Rosenfeld et al., 2016),
454 at least with assumptions that must be carefully assessed (Grosvenor et al., 2018; Zhu et al.,
455 2018b), the spatial and temporal resolution required to observe most ACI processes is beyond
456 present-day satellite-measurement capabilities. Further, the lack of simultaneous, detailed, space-
457 based observations of the multiple factors of import in these processes precludes resolving the
458 key subtleties in these aspects of ACI with satellite data alone.

459 The processes associated with cloud brightening are relatively fast, e.g., on order 10 minutes for
460 aerosol activation in shallow clouds to approximately 20 h for LWP and cloud fraction
461 adjustments (Glassmeier et al., 2021). However, aerosol particles can also alter regional-scale
462 circulation patterns on much longer timescales, which in turn change cloud amount (e.g., Menon
463 et al., 2002). From climate-forcing and even extreme-weather perspectives, these circulation
464 responses are of great importance (Soden & Chung, 2017; Persad et al., 2022; Williams et al.,
465 2022). This is one aspect of ACI where satellite observations play a dominant role, providing
466 frequent, regional-to-global-scale measurements of cloud amount and bulk cloud properties
467 (Figure 1).

468 In summary, many of the factors that dominate aerosol-cloud-radiation interactions, such as CCN
469 concentration and composition, are difficult to measure with space-based remote sensing; this is

especially true for efforts to obtain coincident aerosol and related cloud properties. As such, knowledge of these quantities remains limited on a global scale (e.g., Seinfeld et al., 2016), leaving model-simulated aerosol indirect effects inadequately constrained by observations. Except in special circumstances and with stringent assumptions (e.g., Pahlow et al., 2006; Rosenfeld et al., 2016; Dawson et al., 2020), quantitative constraints on nearly all the key variables needed to characterize detailed ACI are beyond the capabilities of existing and currently planned space-based remote-sensing instruments. Nevertheless, satellite observations of well-defined aerosol sources such as ship tracks, industrial point sources of pollution, wildfire smoke and volcanic plumes, represent "natural laboratories" that have proven particularly useful in studying ACI, because the unaffected surroundings provide a control for the confounding factors associated with meteorology, at least to first order (Christensen et al., 2022). Also, the large datasets obtained from space can support fine-scale stratification of meteorological conditions, allowing the aerosol effects on clouds to be isolated statistically in some cases (e.g., Zamora & Kahn, 2020).

Characterizing ACI globally requires knowledge of co-varying aerosol properties and vertical velocity from suborbital platforms, as a function of meteorological variables that can also be monitored over broad regions from space or derived from reanalysis. The suborbital measurements would be targeted at characterizing statistically the key variables at cloud scale for a range of specific cloud types and meteorological conditions, some of which could be retrieved from space-based instruments with enough specificity to map out the appropriate ACI regimes. Steps in this direction have been taken by several field campaigns, including VOCALS (Bretherton et al., 2010; Mechoso et al., 2014), ORACLES (Redemann et al., 2021), and recently by ACTIVATE (Sorooshian et al., 2019; 2021). The complexity of ACI and resulting ramification calls for a further, comprehensive program of targeted efforts of this kind, especially with simultaneous radiative flux measurements included, so the indirect effect can be quantified and radiative closure assessed. Specifying such a program is beyond the scope of the current review.

3. Regional-to-Global-Scale Modeling Contributions

Climate modeling is of course essential for calculating direct and indirect aerosol forcing globally, for filling gaps in the observational record, and for making climate forcing and response predictions based on assumed future emissions scenarios. Such models can also play a key role in targeting aircraft *in situ* measurements to sample specific aerosol and cloud situations, and subsequently, in identifying the aerosol sources and aging histories for aerosol airmasses captured by satellite and suborbital observations (Figure 1, purple and yellow arrows, respectively). However, from emissions to aerosol climate forcing, models incorporate large numbers of assumptions and parameterizations of complex physical and chemical processes.

3.1. Model Representation of Aerosol Particle Properties

There are a few fundamental steps models take in simulating the global distribution of aerosol particles, and then calculating aerosol direct radiative forcing (e.g., Kinne et al., 2003). First, aerosol paticles and their precursor emissions are usually prescribed from existing datasets, such as for combustion aerosol (e.g., van der Werf et al., 2017), or calculated interactively by the models, typically for sea salt (de Leeuw et al., 2011) and mineral dust (e.g., Zender et al., 2003; Pu et al., 2020) based upon meteorological conditions such as near-surface wind shear and surface properties. The emission inventories specify the initial state of aerosol mass, and might provide some indication of particle composition, size distribution, and aerosol precursor gases (e.g., Randerson et al., 2012; Hoesly et al., 2018). These data, as well as the source locations and vertical distribution of emissions, must be supplied to the model, based on measurements where available (see Section 2), and inferred or assumed elsewhere. Then the atmospheric aerosol concentrations are calculated from simulated atmospheric processes, including chemical and physical transformation, transport, dry deposition, and wet removal. These processes are typically represented as parameterizations of physical mechanisms or as empirical relationships that depend on aerosol physical and chemical properties; they can be coupled interactively to meteorological conditions from the host general circulation model. Next, the aerosol optical properties, including the wavelength-dependent extinction (scattering + absorption), SSA, and particle single-scattering phase function (or asymmetry factor) are calculated as a function of aerosol mass composition, particle size distribution, particle-type-specific hygroscopic growth,

refractive indices, and sometimes particle shape, limited by the extent to which these properties have been measured for the aerosol types involved (see Section 4 below). In bulk aerosol treatments, the microphysical properties are prescribed, although a growing number of modal and sectional aerosol models calculate them explicitly from microphysical schemes (e.g., Stier et al., 2005; Liu et al., 2007; Bauer et al., 2008; Mann et al., 2010; Kokkola et al., 2018). The resulting optical and microphysical properties vary greatly depending on model parameterizations, choice of parameter values, and assumed meteorological conditions. Finally, the direct aerosol radiative effects are derived from the calculated or assumed aerosol optical properties, simulated mass loading, and meteorological conditions, typically using fast, simplified radiative transfer models, though often traceable to more detailed models (e.g., Iacono et al., 2008).

The best-constrained aerosol-related quantity in this process is probably the mid-visible AOD, thanks to the modern-era satellite observations from multiple space-borne platforms (see Section 2 above). This provides a basis for models to adjust their parameterizations and assumptions to better match at least the AOD. Soon after monthly global satellite AOD data products became available, they were used to validate model results (e.g., Kinne et al., 2006; Gliß et al., 2021). More recently, satellite as well as surface-network-based AOD has been used to constrain some models directly. Assimilation of aerosol data has become routine in many operational and research forecasting organizations (e.g., Benedetti et al., 2009; Randles et al., 2017). The International Cooperative for Aerosol Prediction (ICAP) offers a multi-model ensemble that includes major aerosol forecasting activities from around the world, often assimilating satellite-derived AOD as a constraint (Xian et al., 2019).

However, reproducing the global, mid-visible AOD to within the satellite retrieval uncertainty, typically a few tenths under good retrieval conditions, does not resolve the fundamental issues created by the intermediate steps leading up to the AOD calculation. This is clearly demonstrated in AeroCom studies showing that the diversity of model-simulated total AOD is much smaller than the diversities of individual aerosol species and other key quantities such as mass loading, mass extinction efficiency, and lifetime, for which there are far fewer observational constraints (Kinne et al., 2006; CCSP, 2009; Gliß et al., 2021). Apparently, different models match the observed AOD for different reasons. Further, within models, the diversity of all aerosol-forcing-

related quantities increases on regional spatial and seasonal/sub-seasonal temporal scales in most cases.

Although mid-visible AOD is by far the most common observable assimilated in these models, assimilation of lidar extinction and/or backscatter profiles as well as multi-wavelength reflectances from passive sensors is being explored (e.g., Benedetti et al., 2009; Sekiyama et al., 2010; Zhang et al., 2011; 2020b). Generally, in aerosol data assimilation, aerosol optical properties are specified by the host model; measurements are used to adjust only the aerosol constituent mass concentrations. As such, the process is limited by uncertainties in both the measurements and the model parameterizations. Further, the simultaneous adjustment of optical properties, particle size, and mass concentration has not yet received adequate attention, especially given the increasing availability of hyperspectral data (e.g., Lee et al., 2015).

3.2. Application of Model Aerosol Type to Complement Measurements

Aerosol composition based on source characteristics, transport, removal processes, as well as microphysical and chemical evolution, can be extracted from model simulations and used to complement satellite measurements. Specifically, models can in principle provide priors to aerosol remote-sensing retrieval algorithms, or retrospective refinement to the aerosol-type mixing state as part of the satellite aerosol-retrieval process when the aerosol-type information content of the satellite-observed signals is limited (Figure 1, purple arrow). For example, the model-simulated aerosol species, advected downwind, can be used to weight a loosely constrained range of particle properties derived from the satellite observations; the model-simulated aerosol type depends only on the source-receptor relationship, whereas the specificity of satellite-retrieved aerosol-type constraints varies with retrieval conditions, and is greatly diminished when the AOD is low or when a dominant type is lacking (Section 2 above). The challenge is associating the aerosol properties assumed by the model with the aerosol types derived by the satellite retrieval algorithm. An early effort keyed on ANG and absorbing AOD to connect aerosol properties simulated by an aerosol transport model with the satellite-retrieved aerosol type (Li et al., 2015). To make the comparisons, AOD fractions of the modeled aerosol components were ranked, reducing dependence of the result on absolute AOD values. This

application is in its infancy, and the process of associating satellite-constrained aerosol types with aerosol as represented in models requires substantial further investigation.

However, there is also considerable uncertainty in model-simulated particle property detail, arising in part from the lack of such detail in emission inventories used to initialize climate models, and limitations in the representation of particle microphysical and chemical aging processes. Model representation of aerosol properties often fails to capture the complexity found in nature, particularly in bulk models lacking microphysics, and generally, differences among commonly used aerosol representations in models can produce very different aerosol forcing results in model simulations (Nazarenkpo et al., 2017). Further, it has been shown many times in the literature that large diversity exists in model-simulated aerosol properties and both direct and indirect radiative forcing (e.g., Shindell et al., 2013; Schutgens et al., 2013; Stier et al., 2013; Fiedler et al., 2019). The first comprehensive inter-model comparisons of the main parameters determining the aerosol radiative forcing were documented by the AeroCom community, with simulation results from more than a dozen global models (Textor et al., 2006, 2007; Kinne et al., 2006; Schulz et al., 2006). Since then, the diversity among model simulations of aerosol forcing and climate response has persisted, especially at regional scales (e.g., Koch et al., 2009; Huneeus et al., 2011; Samset et al., 2014; Tsigaridis et al., 2014; Pan et al., 2015; Kim et al., 2014, 2019; Bian et al., 2017; Sand et al., 2017; Mortier et al., 2020; Gliß et al., 2021; Lee et al., 2021) despite the advances in satellite observation and *in situ* measurement (summarized in Sections 2 and 4, respectively).

For climate modeling applications, aerosol amount is quite well measured by the combination of satellite and ground-based instruments (Section 2.1 above), which also places constraints on aerosol source strength (e.g., Dubovik et al., 2008; Petrenko et al., 2017), transport, and to some extent, removal (e.g., Das et al., 2021; Kim et al., 2014; 2019; Reid et al., 2009). However, aerosol-type diversity and the associated uncertainty in aerosol-species-specific physical and optical properties, especially SSA (e.g., McComiskey et al., 2008; Loeb & Su, 2010; Thorsen et al., 2021), continue to play a leading role in simulated aerosol direct forcing uncertainty. The availability of generally good, global AOD datasets from satellite remote sensing (Section 2 above) helps constrain transport and removal processes in models, although uncertaintities in other, aerosol-type-specific properties such as SSA and mass extinction efficiency lead to much

poorer constraints on mass loading and direct aerosol forcing. For indirect forcing, uncertainty due to limited observational constraints on the processes by which aerosol particles affect different cloud types (Section 2.3 above) combines with particle and cloud property uncertainties (Carslaw et al., 2013). In addition, vertical velocity at cloud-scale cannot be resolved in current climate models, yet, uncertainties in this quantity can make models overly sensitive to aerosol or meteorological conditions, obscuring the underlying sources of modeled indirect forcing uncertainty (Sullivan et al., 2016). In addition, only a few models treat the aerosol effects on ice nucleation for cold clouds (e.g. Gettelman et al 2012), further contributing to spread due to model diversity. These aerosol-climate-forcing issues in turn make major contributions to the diversity in model climate prediction, and *model diversity represents only a lower bound on model uncertainty, especially when model diversity is reduced as a consequence of model inter-comparison studies rather than based on comparisons with measurements*. *To obtain estimates of actual model uncertainty requires having appropriate measurements againt which to compare*.

3.3. Strengths and Limitations of the Aerosol-Forcing Modeling Process

Within models, the terms associated with aerosol climate forcing can be isolated. Aerosol radiative forcing is usually defined as the change of net irradiance (in W m^{-2}) at either the tropopause after stratospheric temperature adjustment (e.g., Ramaswamy et al., 2001), or at the top-of-atmosphere, provided stratospheric adjustment has little effect on the aerosol radiative forcing (e.g., Schulz et al., 2006). As such, aerosol radiative forcing (ARF) is the combined result of aerosol-radiation interactions (ARI) and ACI (Boucher et al., 2013). ARI represents the change aerosol particles make to the incoming solar radiation reaching Earth's surface through both direct and semi-direct effects. As discussed in Section 2.3, ACI accounts for the impact aerosol particles have on cloud albedo and lifetime when they serve as CCN (or Ice Nuclei, IN), in turn altering radiative fluxes. ARF can produce positive or negative net climate forcing, depending on the aerosol type, meteorological conditions, surface properties, and the spatial distribution of aerosol particles relative to clouds.

To calculate ARI, models must represent the vertically and spectrally resolved aerosol extinction, SSA, and phase function (or asymmetry factor) of airborne particles. As discussed in Sections

3.1 and 3.2 above, these quantities depend on the emissions inventories and meteorology used to initialize the model, the atmospheric transport, removal, chemical processes used to simulate aerosol mass concentrations and composition, and the underlying aerosol physical and optical properties including particle mixing state, size distribution and shape, hydroscopic growth, and spectral-dependent complex refractive indices. The parameterizations associated with each step in model simulation entail considerable uncertainties. Although most of the required quantities are either inferred from measurements or derived from theory, measurements to constrain the models are grossly limited or unavailable. Extrapolating sparse optical property measurements over much larger spatial and temporal domains is risky, resulting in large uncertainties propagating throughout the multiple stages of radiative forcing calculations (e.g., Schutgens et al., 2013).

Modeling the ACI component of aerosol radiative forcing is even more uncertain than that for ARI, as it involves the liquid and ice cloud formation processes, lifetime, and properties along with aerosol particle microphysical characteristics (Section 2.3 above; comprehensively reviewed by Bellouin et al., 2020). For example, changes in CCN can affect cloud-droplet number; clouds can respond with 'adjusted' mass and extent. Similarly, aerosol particles can affect precipitation and latent heat release, although changes in precipitation on regional scales are mediated primarily by energy and water budget considerations (e.g., Held & Soden, 2006; O'Gorman et al., 2012; Gettelman & Sherwood, 2016; Dagan and Stier, 2021). Studies of aerosol impacts on deep convective clouds find that with increased aerosol loading, precipitation and convective updrafts might increase, decrease, or remain relatively unchanged, apparently depending upon other, unidentified factors (Tao et al., 2012; Boucher et al., 2013), results that entail significant model uncertainties (Marinescu et al., 2021). As discussed earlier, quantifying aerosol indirect effects also involves modeling cloud formation, lifetime, and properties, and is highly sensitive to cloud-scale vertical velocities and the above-mentioned aerosol characteristics. Many of the relevant processes operate on spatial scales far smaller than the resolution of global models and are therefore either highly parameterized or simply not included, such as some aerosol effects on convective clouds. Large Eddy Simulations (LES) are valuable research tools, complementary to climate models in that they can resolve many of the smaller-scale processes; however, they still entail assumptions that are often unconstrained by

675 measurements, are impractical to run over large areas or long time periods, and cannot provide
676 estimates of forcing (e.g., Bellouin et al., 2020).

677 Further, the review of aerosol forcing modeling above addresses only present-day conditions. In
678 most modeling-oriented climate assessments such as the IPCC reports, aerosol radiative forcing
679 requires accounting for the difference between present-day and "pre-industrial" conditions, i.e.,
680 conditions at a time when it is assumed anthropogenic influences were minimal, commonly taken
681 as 1750 or 1850. This of course engenders furthur uncertainties, as discussed in Section 1.

682

683 **4. The Role of Aircraft and Ground-based *In situ* and Remote-sensing Measurements**

684 Existing aircraft and ground-based *in situ* and remote-sensing aerosol measurements are
685 sometimes described collectively as the suborbital aerosol program-of-record. These data
686 provide the key information that has been applied by both the satellite and modeling
687 communities as constraints on aerosol microphysical properties (e.g., Reddington et al., 2017;
688 Gliß et al., 2021) and vertical structure (Watson-Parris et al, 2019). The particle property
689 measurements are used to assign priors and to make other assumptions in many satellite remote-
690 sensing aerosol retrieval algorithms. They also form the basis for the microphysical property
691 assumptions of aerosol source characteristics in most models, and they can contribute to model
692 validation (Figure 1). For climate applications, these suborbital measurements are usually used in
693 a statistical sense due to their significant spatial and/or temporal sampling limitations.

694 **4.1. Surface-based Observation Network and Aircraft Field Campaign Contributions**

695 Networks of surface-based remote-sensing instruments, such as sun photometers (e.g., the
696 Aerosol Robotic Network, AERONET, Holben et al., 1998; the Marine Aerosol Network,
697 MAN, Smirnov et al., 2011) and aerosol lidars (e.g., the Micro-Pulse Lidar Network, MPLNET,
698 Welton et al., 2001; the European Aerosol Research Lidar Network, EARLINET, Pappalardo et
699 al., 2014), generate a substantial database of aerosol amount, vertical distribution, and some
700 particle property information. In addition, heavily instrumented surface sites, such as the US
701 Department of Energy's Atmospheric Radiation Measurement (ARM) program at their Southern
702 Great Plains site and Mobile Facilities (e.g., Vogelman et al. 2012; Mather & Voyles, 2013)

contribute detailed, local aerosol, cloud, and radiation measurements. (A table of leading surface-based aerosol measurement networks is given in Kahn et al. (2004).) Direct *in situ* sampling at ground stations can add physical, optical, and chemical detail to the remotely sensed particle optical properties and can document long-term variability, but such measurements are acquired only near or at the surface and at only a few sampling sites, mostly concentrated in a few regions of the globe (e.g., The US Interagency Monitoring of Protected Visual Environments network, IMPROVE, Malm & Hand, 2007; the Surface Particulate Matter Network, SPARTAN, Snider et al., 2015; the Global Atmospheric Watch (GAW) network, Andrews et al., 2019; Rose et al., 2021; Laj et al., 2020; the European Commission's Aerosol, Clouds, and Trace Gases Research Infrastructure (ACTRIS) (https://www.actris.eu; last accessed: March 2023); plus studies of collected data by Fanourgakis et al., 2019; Burgos et al., 2020).

Intensive aircraft field campaigns complement the surface-based measurements, offering much greater flexibility in sampling the atmosphere near and downwind of aerosol sources; this allows for *in situ* as well as remote-sensing measurement of transported aerosol layers aloft as they evolve, and further, can provide insight into the processes involved in aerosol evolution and aerosol-cloud interaction (e.g., the Atmospheric Tomography Mission, ATom, Brock et al., 2021; the Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys campaign, SEAC^{4}RS, Toon et al., 2016; The Green Ocean Amazon Experiemnt, GOAMAZON, Martin et al., 2017; the Observations of Aerosols above Clouds and their Interactions campaign, ORACLES, Redemann et al., 2021; the Aerosol Cloud Meteorology Interactions over the Western Atlantic Experiment, ACTIVATE, Sorooshian et al., 2021). However, such campaigns can target only selected regions and are usually limited in time, typically lasting only a few weeks or months. Finally, laboratory analysis of collected samples is sometimes required to obtain compositional and some key optical results, but are currently also severely limited in scope (e.g., Di Biagio et al., 2017; McNeill et al., 2020).

4.2. Strengths and Limitations of Aerosol Property Data Available from Suborbital Sources

Aerosol property databases have been developed in the past. A leading example is the Optical Properties of Aerosols and Clouds (OPAC) database (Hess et al., 1998). It provides tabulated, digital, microphysical and optical properties, covering UV, visible, and infrared spectral ranges,

for aerosol components as well as mixtures representing various aerosol types, and includes data calculated at a number of relative humidity values. The need for such databases is demonstrated by the widespread use of OPAC optical properties in models, satellite remote sensing retrieval algorithms, and as precomputed packages for use in radiative transfer codes (e.g., Chin et al., 2002; Thomas et al., 2009; Dee et al., 2011; Gasteiger & Wiegner, 2018; Vicent et al., 2020). Indeed, OPAC has over 3,000 citations at the time of writing, according to Google Scholar. However, OPAC has known, significant limitations, due to the lack of representative (or in some cases, any) measurements of the required aerosol properties. These limitations have been documented in the literature (e.g., Colarco et al., 2013; Zieger et al., 2013; Alvarado et al., 2016) and, despite the availability of much better instrumentation than that used decades ago to help construct OPAC, this situation persists today.

As consistently and systematically measured particle chemical, microphysical, and optical properties are currently lacking, it is difficult to critically assess and improve model assumptions. Recent efforts to collect retrospective, *in situ* aerosol data into a better organized "program-of-record" promise to help refine our sense for the range of particle property values in different places (e.g., Reddington et al., 2017; Balkanski et al., 2021; Schuster & Trepte, 2021). However, past aerosol property measurements were not designed to provide the comprehensive inputs needed for global climate modeling. To be most useful as constraints on modeling aerosol forcing, suborbital measurements of particle microphysical and chemical properties must meet the following four criteria:

• A ***statistical sampling*** of key variables for major aerosol airmass types is needed. (An aerosol airmass describes the spatial and temporal distribution of aerosol particles having similar properties, based on remote-sensing retrieval of aerosol type or on model simulation from a particular source and evolution.) Aircraft *in situ* aerosol measurements are not often made with the aim of obtaining the distribution (e.g., the probability distribution function or PDF) of particle property values associated with specific aerosol types. Except at sparse surface-based sampling stations, a limited number of individual measurements of a given aerosol airmass type is the best we currently have in most cases, whereas to establish the PDF of a quantity, a sufficient number of measurements is required so that subsequent measurements reproduce the distribution of values already obtained.

762 • A ***suite of specific physical, chemical, and optical quantities*** is needed to adequately
763 characterize an aerosol type for climate applications, and would also be useful for air quality
764 modeling (Table 1). All the required types of measurements have been made in the past, but
765 rarely have they all been made together for the same aerosol airmass and under appropriate
766 ranges of meteorological conditions.

767 • ***Particle property evolution during transport needs to be characterized***. The properties of
768 many aerosol types change significantly as they age. As such, the particle properties of major
769 aerosol types need to be characterized systematically *downwind from the source* (or upwind
770 toward the source) to adequately capture the aging process. Further, the *source, the age, and the*
771 *associated environmental conditions* need to be recorded for each sample, best done in most
772 cases by aerosol transport modeling (Figure 1, yellow arrow). (The timescales for particle
773 evolution vary. Changes in wildfire smoke particles and secondary aerosol formation typically
774 occur over minutes to hours (e.g., Yokelson et al., 2009; Kleinman et al., 2020), whereas
775 alteration of mineral dust properties can take days (e.g., Denjean et al., 2016) and is unlikely to
776 be fully captured during a single aircraft flight.) Such aircraft measurements can also acquire
777 information characterizing *aerosol variability at spatial scales of a few km or less*, which can be
778 important for representing sub-grid-scale aerosol properties and ACI processes in climate models
779 (Haywood et al., 1997; Fast et al., 2022). With the help of aerosol transport modeling and/or
780 synoptic-scale satellite observations, aerosol sources and transport history can be determined,
781 even when aerosol layers are superposed at different elevations and from different sources, but
782 again, this has not been done for much of the program-of-record.

783 • ***Uncertainty characterization is critical*** to assess the resulting uncertainty in derived aerosol
784 forcing. However, estimates of measurement uncertainty are difficult to obtain for many aircraft
785 *in situ* instruments. This situation can be improved considerably with techniques that were
786 unavailable for most field campaigns until recently. For example, open-path instruments, some
787 existing and some in development, can measure aerosol properties under ambient conditions.
788 Such properties include aerosol scattering and extinction, as well as particle size distribution
789 (Martins, 2016) and light-absorption estimates (Gordon et al., 2015). These measurements
790 facilitate uncertainty estimate calculations for the particle hygroscopicity derived from other, in-
791 aircraft measurements. In addition, the open-path instruments can produce uncertainty estimates

792 (or even corrections) for directly measured inlet efficiency, which is especially important for
793 super-micron particles.

794

795 **Table 1*.** The suite of aerosol properties that need to be measured systematically, primarily from
796 aircraft, and the purpose of each, for the climate-forcing application[†].

797

Aerosol Properties Obtained from *In Situ* Measurements[§] and Integrated Analysis	
Spectral extinction coefficient	- To constrain and to interpret spectral Aerosol Optical Depth (AOD)
Spectral absorption or single-scattering albedo	- To determine atmospheric spectral light-absorption and heating - To constrain spectral AOD retrievals
Particle hygroscopic growth factor	- To connect particle properties with ambient RH conditions - To model particle activation and aerosol-cloud interactions
Particle size distribution	- To model particle optical properties and loss mechanisms - To model particle activation and aerosol-cloud interactions - As a complement to chemical composition discrimination - Required for deriving aerosol Mass Extinction Efficiency (MEE)
Particle composition	- For source identification - To classify measurements in terms of aerosol types as specified in most models, e.g., sea salt, sulfate, mineral dust, black carbon (BC), brown carbon (BrC), especially important for aerosol-cloud-interaction modeling - To support deriving properties of the anthropogenic fraction, as needed to calculate anthropogenic aerosol "climate" forcing (also supports air quality applications)
Spectral single-scattering phase matrix [all possible angles]	- To calculate radiation fields - To constrain multi-angle radiance AOD retrievals - *Polarized* – to help determine aerosol type, and to constrain remote-sensing observations where polarized data are included
Mass extinction efficiency (MEE)	- To translate between optical remote-sensing measurements and model aerosol mass [Can be derived from integrated analysis of particle size distribution and extinction coefficient, with density deduced from particle compositional constraints or measured directly from samples]
Real Refractive Index	- To model aerosol optical properties

	- To constrain AOD retrievals to the level-of-detail required for aerosol forcing
Variables Providing Meteorological Context	
Carbon Monoxide (CO; also possibly CO_2, NO_2, O_3)	- As a tracer for smoke, to help distinguish smoke from urban pollution in some cases - For modeling heterogeneous reactions, secondary aerosol formation
Ambient temperature, Pressure, and Relative humidity (RH)	- To help interpret ambient measurements - To translate between instrument and ambient conditions
Aircraft 3-D location	- To relate aircraft measurements to model simulations and to any available satellite observations, for source identification and aging history
Variables Providing Ambient, Remote-Sensing Context	
Ambient Spectral single-scattering phase matrix [all possible angles]	- To assess in-aircraft measurements by comparing with ambient conditions - To help calculate radiation fields and constrain remote-sensing AOD retrievals - *Polarized* – to help determine aerosol type, and to constrain remote-sensing retrievals where polarized data are included
Ambient Spectral extinction coefficient	- To constrain remote-sensing spectral AOD retrievals and assess in-aircraft measurements by comparing with ambient conditions
Large particle / cloud probe	- To provide information about dust and other particles larger than the inlet size cut of other instruments - As an independent measure of possible cloud impact on the reliability of other data
Aerosol layer heights	- To determine flight levels for subsequent direct sampling - To correlate with meteorological conditions - As a constraint on trajectory modeling to identify aerosol sources and evolution

*Adapted from Kahn et al., 2017; © American Meteorological Society. Used with permission. This table was originally developed for aircraft aerosol measurements, as these can sample aerosol aloft and can characterize particle property evolution systematically. However, a subset of these measurements acquired from surface-based instruments can contribute as well, especially for the near-surface components that are difficult to sample from airborne platforms.

†The variables in this table are aimed at characterizing aerosol properties for the major aerosol air mass types in general. CCN and Ice Nucleii (IN) would be included, especially in the hygroscopic growth factor and size distribution measurements. However, systematic and comprehensive measurements of additional cloud and meteorological variables associated with

aerosol-cloud interactions, for the major cloud types and meteorological regimes, globally, as introduced in Section 2.3, would require a different aircraft program, beyond the scope of this review.

§These quantities are typically measured inside the aircraft, at low or at least altered RH.

Although many current models are not equipped to apply detailed *in situ* aerosol property measurements directly due to the simplicity of their parameterizations, such information remains critical for characterizing model error. By systematically collocating measured and modeled aerosol optical properties in space and time, the probability distribution of errors in these parameters can be estimated. Such diagnostics can help refine model parameterizations and better quantify the uncertainties in the derived aerosol radiative forcing, especially those associated with simplified assumptions and parameterizations. And as computer capabilities allow climate models to advance, the applicability of information provided by such measurement detail will increase. For example, more sophisticated parameterizations of aerosol-cloud interactions and more subtle particle physical and chemical changes during particle aging could be captured in the simulations.

5. Discussion – Addressing Suborbital Data Limitations

Of the three elements shown in Figure 1, the suborbital measurement component is the least well addressed for the aerosol climate forcing application by existing and planned efforts. In this section, we review the more comprehensively defined possibilities for aerosol property measurements first, and touch on those for ACI subsequently.

The current aerosol measurement program-of-record does not meet the criteria summarized in Section 4. This is reflected in the *diversity of assumed aerosol properties* in remote-sensing retrieval algorithms, and the even greater particle property diversity among model assumptions (e.g., CCSP, 2009; Myhre et al., 2013; Gliß et al., 2021). The lack of consensus about aerosol intensive properties provides some indication of what is yet to be achieved with systematic *in situ* aerosol measurements. *All measurements have uncertainties. However, in situ measurements*

836 *can provide the most direct way to quantify particle SSA and size distribution, offer by far the*
837 *best constraints on particle hygroscopicity and the CCN size spectrum, and produce the only*
838 *available constraints on MEE and actual particle composition.* This is illustrated, for example,
839 by the work of Brock et al. (2021).

840 As such, a comprehensive database of key *in situ* measurements targeting the major aerosol
841 airmasses would fill this persistent gap in our ability to model the effects of aerosol particles on
842 climate. It would also add value to several decades of existing as well as all future global satellite
843 remote-sensing measurements by providing more robust, complete, and detailed particle
844 microphysical property information than is currently available to use as priors or constraints on
845 remote-sensing retrieval algorithms. For data assimilation, such measurements would provide a
846 better basis for quantifying errors arising from faulty or uncertain assumptions about aerosol
847 optical properties in the retrieval process and would open the door for the assimilation of aerosol
848 properties beyond AOD. The data would be of value for air quality applications as well, by
849 contributing to the accuracy of aerosol species as represented in air quality models.

850 One simplifying factor for developing a database addressing current needs is that, *for a given*
851 *source in a given season, the particle intensive microphysical and optical properties tend to be*
852 *repeatable*, even as the aerosol amount varies on many spatial and temporal scales. For example,
853 although the amount of dust raised from a particular desert source region can vary diurnally,
854 seasonally, and interannually, the emitted dust microphysical properties generally remain
855 unchanged (e.g., Reid et al., 2008). Similarly, wildfires consuming the same vegetation types in
856 the same ecosystem and season tend to produce smoke particles having similar properties (e.g.,
857 Reid et al., 2005; Jungehnn Noyes et al., 2022). This means a program aimed at making the suite
858 of key *in situ* particle intensive property measurements systematically is at least feasible.

859 Complementary satellite measurements are also needed (Figure 1), frequently, over large spatial
860 scales for regional context, (a) to map the varying extensive properties, i.e., aerosol amounts and
861 3-D spatial distributions of different aerosol types, and (b) to limit as much as possible sampling
862 bias (e.g., Schutgens et al., 2017). Aerosol type, the classification possible based on particle
863 size, shape, and SSA constraints that can be derived from satellite remote-sensing measurements,
864 is the key to linking the extensive aerosol air masses, mapped out frequently and globally by

satellites, with the detailed intensive properties that can only be derived from *in situ* measurements. Hence, systematic *in situ* aircraft measurements will have made a key contribution to reducing aerosol forcing uncertainty once they have adequately characterized the detailed particle microphysical properties for major aerosol sources and their downwind evolution. Further, the *in situ* measurements can be acquired under non-precipitating cloudy conditions, in some cases even if the aerosol particles are concentrated within a cloud layer, and the observations need not be obtained in synchrony with satellite observations (although doing so at times would be important, e.g., for remote-sensing retrieval validation and assessment of consistency among different measurement approaches).

Aircraft *in situ* programs aimed at measuring aerosol microphysical or optical properties systematically have been deployed in the past, though with different objectives from the one discussed here (e.g., the Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container, CARIBIC, Nguyen et al., 2006; the Vertical Profiles of Aerosol Optical Properties over rural Oklahoma program, Andrews et al., 2011; the Vertical Profiles of Aerosol Optical Properties over Central Illinois program, Sheridan et al., 2012). Some recent airborne science projects have also included components aimed at characterizing aerosol properties or processes systematically (Brock et al., 2021; Redemann et al., 2021, Sorooshian et al., 2019; 2021). These efforts offer some guidance as to how such programs might operate. However, the level of effort needed to characterize statistically the required suite of aerosol properties (Table 1), for many aerosol types, has been beyond the scope of past campaigns, which have either focused on one specific region or included only commercial airline pathways, and some quantities such as MEE have not been constrained in most of these experiments.

A possible future aircraft program addressing the needed aerosol microphysical properties for reducing aerosol forcing uncertainty has been outlined previously, showing that the key variables could be measured with a relatively small but dedicated aircraft, and with technologies that existed even in 2014, though there have been some significant improvements in instrumentation since (Kahn et al., 2017). The aircraft would fly two-to-three times per week from an initial base of operations, with fixed flight plans aimed at sampling all the major, climatologically likely aerosol airmasses accessible from the base, as identified in advance from model simulations and satellite aerosol-type mapping. The flight plan would begin with a high-altitude traverse along

one of the pre-determined paths, and a simple lidar on board would determine the elevations of the aerosol layers below. The layers would then be sampled, as far up- or downwind as feasible, and the aircraft would return to base.

Once the PDFs of the key variables were acquired for the accessible aerosol airmasses, the aircraft would move to a new base of operations, again informed by an analysis of aerosol transport model simulations and aircraft siting considerations. Unlike typical aircraft field campaigns, the data would be processed routinely, probably at a central location, as is common for NASA satellite missions. In addition to helping deployment planning in advance, by identifying important source locations and likely dispersion pathways as sampling targets, aerosol transport models would also be run routinely during deployment, to characterize the sources and aging histories of the aerosol particles sampled during each flight (Figure 1, yellow arrow). As opportunities arose, the aircraft would overfly ground stations for cross-validation, and could also participate in intensive field campaigns when appropriate. Taking account of typical aircraft maintenance considerations, the availability of basing facilities, and the seasonality of some aerosol sources, we estimate that a deployment of roughly three-to-four years would be required to sample the major aerosol airmasses of North America. There is additional uncertainty in this estimate, however, due to limited knowledge about the number of samples required to obtain PDFs for different key variables and for different aerosol types, depending also on the capabilities of the designated aircraft and the robustness of the instrumentation. The successful demonstration of such a program could engender similar programs hosted in other regions around the world, leading to characterization of the major aerosol airmass types, globally.

The requirements for suborbital measurements to comprehensively reduce uncertainties in modeling ACI processes and effects are not as well established as those described above for addressing aerosol properties. A suborbital ACI measurement program could be considerably more complex. Reviews of past modeling and measurement work (e.g., Rosenfeld et al., 2014; Mülmenstädt & Feingold, 2018; Bellouin et al., 2020) and experience from recent aircraft campaigns aimed at characterizing aerosol particles, clouds, and their interactions in specific regions (Behrenfeld et al., 2019; Sorooshian et al., 2019; 2021; Crosbie et al., 2022) provide some indication of what would be involved. Both aerosol and cloud properties would need to be

925 measured for this application, on spatial scales ranging from 10^{-7} to 10^{6} m and temporal scales of
926 minutes to hours or more. Sampling would have to emcompass major cloud types, and several
927 coordinated aircraft would probably be required to provide an combination of remote-sensing
928 and *in situ* observations capable of resolving variations both vertically as well as horizontally
929 within and surrounding cloud systems. Variables identifying distinct cloud and meteorological
930 regimes, that could subsequently be mapped with space-based remote sensing, would be required
931 to allow the field results of be extrapolated to global scale and extended time periods. A future
932 study identifying the required variables, likely instruments to make the key measurements,
933 targeting requirements, and observation strategies aimed at reducing ACI modeling uncertainties
934 in general is indicated.

935

936 **6. Conclusions – A Three-Way Street**

937 The aerosol-related climate forcing uncertainties have not diminished appreciably in more than
938 20 years, despite substantial progress in other aerosol- and cloud-related measurement and
939 modeling areas. Forcing uncertainty translates directly into climate change attribution and
940 prediction uncertainty.

941 Unlike most previous reviews of aerosol climate forcing constraints and uncertainties, we take a
942 measurement-oriented perspective. As illustrated in Figure 1, the satellite and suborbital
943 measurement as well as the modeling research communities each have unique and essential
944 contributions to make to the other two to achieve substantial progress in reducing the uncertainty
945 in aerosol climate forcing overall. The application of loose satellite constraints on aerosol type
946 and precursor gas distributions to improve model simulations needs to be explored in broader
947 terms than previously, in part by taking the limited retrieved information content on particle size,
948 shape, and light-absorption where available and linking it to more specific aerosol species, based
949 on source properties projected to the observed locations by aerosol transport modeling. This is an
950 example of a two-way street – the models help refine the satellite-retrieved aerosol type results,
951 especially where the AOD is too low for reliable satellite aerosol-type retrieval or where cloud,
952 complex surface topography or snow cover entirely precludes remote-sensing aerosol
953 measurement; the aggregate of satellite instruments provides the 3-D spatial distribution of

954 aerosol amount and type to constrain and/or validate the models, improving aerosol source
955 initialization, transport, transformation, and deposition, and reducing model bias. Aerosol data
956 assimilation offers another example: for each analysis cycle, the model projects past observations
957 into the future, which in turn provides additional constraints for the assimilation of new data.

958 As outlined in Section 4 above, for the suborbital component to adequately contribute to
959 modeling and to the interpretation of satellite observations (the third way), a database of
960 systematic, statistically representative *in situ* measurements needs to be developed, containing
961 the key particle microphysical, optical, and chemical properties and associated uncertainty
962 estimates that cannot be derived from remote-sensing measurements alone. Such *in situ*
963 measurements are needed to improve assumptions and parameterizations in models as well as
964 observation operators for data assimilation, putting the model-assumed aerosol particle
965 properties on more solid and consistent footing. The detailed aerosol properties will also serve to
966 improve the priors or constraints on satellite retrieval algorithms, adding value to more than two
967 decades of retrospective as well as planned future satellite aerosol observations. Further, such *in*
968 *situ* aerosol data, with attention to the co-variability among aerosol and meteorological variables,
969 would make more precise and *quantitative* connections between the satellite optical constraints
970 on aerosol amount and type and the modeled, species-specific, aerosol mass (e.g., the MEEs), a
971 key requirement for data assimilation and model validation. A separate suborbital measurement
972 program is needed to characterize the processes by which the aerosol particles and clouds
973 interact, reporting detailed cloud properties in the presence of different aerosol amounts and
974 types, under a range of meteorological conditions.

975 Completing the pattern of relationships, models, constrained by satellite measurements, can help
976 in targeting the systematic *in situ* measurements to regions where they will matter most.
977 Subsequently, models can help interpret the aerosol source and aging history, as well as cloud
978 regime-type and evolution acquired through satellite and suborbital observations. In addition to
979 its application to climate-related study, this approach would help improve model representation
980 of surface-level particulate matter (PM) concentrations and composition, which is at the heart of
981 air quality monitoring on regional and global scales (e.g., Appel et al., 2008; Hammer et al.,
982 2019; van Donkelaar et al., 2021). Specifically, two leading sources of uncertainty for air quality
983 applications relate to the aerosol vertical distribution and aerosol composition, which are also

among the leading uncertainties in aerosol contributions to climate forcing, though the observational requirements for air quality applications are somewhat different, e.g., requiring near-surface aerosol chemical toxicity and possibly tighter constraints on particle size. For air quality applications, recent advances in data analysis techniques are likely to make important contributions (e.g., Bellinger et al., 2017)

Clearly, programs supporting advanced, global-scale satellite and surface-based aerosol, precursor gas, and cloud observations, climate modeling, surface networks, and intensive field campaigns aimed at characterizing the underlying physical and chemical processes involved, as currently planned, are all essential. The main parts of the overall picture missing from current programs or those planned for the near future are: (1) a program of long-term, systematic aircraft measurements aimed at creating a climatology of the key aerosol microphysical properties driving aerosol-climate interactions for the major aerosol airmass types, along with their statistical distributions and uncertainties, (2) a suborbital program aimed at filling gaps in aerosol-cloud-interaction-related cloud-scale microphysics, cloud optical properties, and associated dynamics, and (3) the mandate, the support, and the computational resources for the respective communities to work together in detail on the *synthesis* effort – the three-way street (Figure 1). Such efforts would go a long way toward 'model-data fusion' (e.g., Gettelman et al., 2022); that is, developing consistent representations of aerosol properties as retrieved from remote sensing, measured *in situ*, and adopted by models (sometimes referred to in the AeroCom, AeroSat, and ICAP communities as "harmonizing" aerosol types among different platforms and communities). It would help members of the involved research communities agree upon those properties that most affect the aerosol analysis or application in a given place and time, and would greatly improve the feasibility of exchanging aerosol information among and between measurements and models. The cloud-related requirements for constraining aerosol-cloud interactions, such as better characterization of ice-cloud processes, documentation of aerosol effects on cloud condensate and cloud fraction, as well as vertical velocity constraints, especially in convective systems, are also needed. Yet, fully effecting the three-way street seems to be essential for substantially reducing the aerosol direct and indirect forcing uncertainty that persistently dominates climate and also affects air quality prediction uncertainty. The technical capabilities to achieve this are all currently available.

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Open Research

This paper represents a review and analysis of previously published work. As such, the underlying data are made available from the original sources through the cited literature.

References

Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., & Toon, O. B. (2004). The impact of humidity above stratiform clouds on indirect aerosol climate forcing. *Nature 432*, doi:10.1038/nature03174.

Adebiyi, A.A., & Kok, J.F. (2020). Climate models miss most of the coarse dust in the atmosphere. Sci. Adv. 6, eaaz9507, doi:10.1126/sciadv.aaz9507.

Alvarado, M.J., Lonsdale, C.R., Macintyre, H.L., Bian, H., Chin, M., Ridley, D.A., et al. (2016). Evaluating model parameterizations of submicron aerosol scattering and absorption with *in situ* data from ARCTAS 2008. *Atmos. Chem. Phys., 16, 9435–9455*, doi:10.5194/acp-16-9435-2016.

Anderson, T.L., Charlson, R.J., Winker, D.M., Ogren, J.A., & Holmen, K. (2003). Mesoscale variations of tropospheric aerosols., J. Atmosph. Sci. 60, 119-135, doi:10.1175/1520-0469(2003)060.

Andreae, M.O. (2009). Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions. Atmos. Chem. Phys., 9, 543–556, doi:atmos-chem-phys.net/9/543/2009.

Andreae, M.O. (2019). Emission of trace gases and aerosols from biomass burning – an updated assessment. *Atmos. Chem. Phys., 19, 8523–8546*, doi:10.5194/acp-19-8523-2019.

Andrews, E., Sheridan, P.J., & Ogren, J.A. (2011). Seasonal differences in the vertical profiles of aerosol optical properties over rural Oklahoma. *Atmos. Chem. Phys. 11, 10661–10676*, doi:10.5194/acp-11-10661-2011.

Andrews, E., Ogren, J.A., Kinne, S., & Samset, B. (2017). Comparison of AOD, AAOD and column single scattering albedo from AERONET retrievals and in situ profiling measurements. *Atmos. Chem. Phys., 17, 6041–6072*, doi:10.5194/acp-17-6041-2017.

Andrews, E., Sheridan, P.J., Ogren, J.A., Hageman, D., Jefferson, A., et al. (2019). Overview of the NOAA/ESRL Federated Aerosol Network. *Bull. Am. Meteorol. Soc. 123-135*, doi:10.1175/BAMS-D-17-0175.1.

Appel, K.W., Bhave, P.V., Gilliland, A.B., Sarwar, G., & Roselle, S.J. (2008). Evaluation of the community multiscale air quality (CMAQ) model version 4.5: Sensitivities impacting model performance; Part II—particulate matter. *Atmos. Env. 42, 6057-6066,* doi:10.1016/j.atmosenv.2008.03.036.

Balkanski, Y., Mona, L., Andrews, E., Bellouin, N., Carslaw, K., Chin, M., et al. (2021). AeroCom-AeroSat Commission on Constraining Aerosol Properties. AeroCom-AeroSat virtual meeting, 14 October 2021.

Bauer, S. E., Wright, D. L., Koch, D., Lewis, E. R., McGraw, R., Chang, L.-S., et al., (2008). MATRIX 534 (Multiconfiguration Aerosol TRacker of mIXing state): an aerosol microphysical module for 535 global atmospheric models. *Atmos. Chem. Phys. 8*(20*), 6003–6035*, doi:10.5194/acp-8-6003-2008.

Behrenfeld, M.J., Moore, R.H., Hostetler, C.A., Graff, J., Gaube, P, et al. (2019). The North Atlantic Aerosol and Marine Ecosystem Study (NAAMES): Science Motive and Mission Overview. *Front. Mar. Sci. 6, 122*, doi:10.3389/fmars.2019.00122.

Bellinger, C., Jabbar, M.S.M, Zaiane, O., & Osornio-Vargas, A. (2017). A systematic review of data mining and machine learning for air pollution epidemiology. *BMC Public Health 17 (907)*, doi:10.1186/s12889-017-4914-3.

Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., et al. (2020). Bounding global aerosol radiative forcing of climate change. *Rev. Geophys. 58(1), e2019RG000660*, doi:10.1029/2019RG000660.

Benedetti, A., Morcrette, J.-J., Boucher, O., Dethof, A., Engelen, R. J., Fisher, M., et al. (2009). Aerosol analysis and forecast in the European Centre for Medium-Range Weather Forecasts Integrated Forecast System: 2. Data assimilation. *J. Geophys. Res., 114, D13205*, doi:10.1029/2008JD011115.

Bian, H., Chin, M., Hauglustaine, D. A., Schulz, M., Myhre, G., Bauer, S. E., et al. (2017). Investigation of global particulate nitrate from the AeroCom phase III experiment. *Atmos. Chem. Phys. 17, 12911–12940*. doi:10.5194/acp-17-12911-2017.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., et al. (2013). Clouds and Aerosols. Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., et al. (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Bougiatioti, A., Nenes, A., Lin, J.J., Brock, C.A., de Gouw, J., Liao, J., et al. (2020). Drivers of cloud droplet number variability in the summertime with focus on the Southeast United States. *Atmos.Chem.Phys 20, 12163–12176*, doi:10.5194/acp-20-12163-2020.

Bretherton, C.S., Wood, R., George, R.C., Leon, D., Allen, G., & Zhent, X. (2010). Southeast Pacific stratocumulus clouds, precipitation and boundary layer structure sampled along 20 S during VOCALS-Rex. *Atmos. Chem. Phys., 10, 10639–10654*, doi: 10.5194/acp-10-10639-2010.

Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, *Geophys. Res. Lett., 34, L03813*, doi:10.1029/2006GL027648.

Brock, C.A., Froyd, K.S., Dollner, M., Williamson, C.J., Schill, G., Murphy, D.M., et al. (2021). Ambient aerosol properties in the remote atmosphere from global-scale in situ measurements, *Atmos. Chem. Phys., 21, 15023–15063,* doi:10.5194/acp-21-15023-2021.

Buchard, V., Randles, C.A., Silva, A.M. da, Darmenov, A., Colarco, P.R., Govindaraju, et al. (2017). The MERRA-2 aerosol aeanalysis, 1980 onward. Part II: Evaluation and case studies. *J. Clim. 30, 6851–6872*, doi:10.1175/JCLI-D-16-0613.1.

Burgos, M.A., Andrews, E., Titos, G., Benedetti, A., Bian, H., Buchard, V., et al. (2020). A global model–measurement evaluation of particle light scattering coefficients at elevated relative humidity. *Atmos. Chem. Phys., 20, 10231–10258*, doi:10.5194/acp-20-10231-2020.

Burton, S.P., Ferrare, R.A., Hostetler, C.A., Hair, J.W., Rogers, R.R., Obland, M.D., et al. (2012). Aerosol classification using airborne High Spectral Resolution Lidar measurements – methodology and examples. *Atmos. Meas. Tech. 73–98*, doi:10.5194/amt-5-73-2012.

Burton, S.P., Hair, J.W., Kahnert, M., Ferrare, R.A., Hostetler, C.A., Cook, A.L., et al. (2015). Observations of the spectral dependence of linear particle depolarization ratio of aerosols using NASA Langley airborne High Spectral Resolution Lidar. Atmos. Chem. Phys., 15, 13453–13473, doi:10.5194/acp-15-13453-2015.

Cao, Y., Zhu, Y., Wang, M., Rosenfeld, D., Liang, Y., Liu, J., et al. (2023). Emission reductions significantly reduce the hemispheric contrast in cloud droplet number concentration in recent two decades. *J. Geophys. Res. Atmosph. 128, e2022JD037417*, doi:10.1029/2022JD037417.

Capelle, V., Chedin, A., Pondrom, M., Crevoisier, C., Armante, R., et al. (2018). Infrared dust aerosol optical depth retrieved daily from IASI and comparison with AERONET over the period 2007–2016. Remt. Sens. Env. 206, 15-32, doi:10.1016/j.rse.2017.12.008.

Carr, J.L., Á. Horváth, D.L. Wu, & M.D. Friberg (2022). Stereo plume height and motion retrievals for the record-setting Hunga Tonga-Hunga Ha'apai eruption of 15 January 2022. *Geophys. Res. Lett. 49, e2022GL098131*, doi:10.1029/2022GL098131

Carslaw, K.S., Lee, L.A., Reddington, C.L., Pringle, K.J., Rap, A., Forster, P.M., et al. (2013). Large contribution of natural aerosols to uncertainty in indirect forcing. *Nature 503, 67-74*, doi:10.1038/nature12674.

Carslaw, K.S., Gordon, H., Hamilton, D.S., Johnson, J.S., Regayre, L.A., et al. (2017). Aerosols in the pre-industrial atmosphere. *Curr. Clim Change Rep. 3, 1-15*, doi:10.1007/s40641-017-0061-2.

CCSP (U.S. Climate Change Science Program) Synthesis and Assessment Product 2.3 (2009). Atmospheric aerosol properties and climate impacts. Chin, M., R.A. Kahn, & S. Schwartz, Eds. Pp. 116.

Chin, M., Ginoux, P., Kinne, S.,Torres, O., Holben, Duncan, B.N., et al. (2002). Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sun photometer measurements. *J. Atmos. Sci. 59 (3)*, doi:10.1175/1520-0469(2002).

Christensen, M., Gettelman, A., Cermak, J., Dagan, G., Diamond, M., Douglas, A., et al. (2022). Opportunistic experiments to constrain aerosol radiative forcing. *Atmos. Chem. Phys. 22, 641–674*, doi: 10.5194/acp-22-641-2022.

Colarco, P.R., Nowottnick, E.P., Randles, C.A., Yi, B., Yang, P., Kim, K-M., et al. (2013). Impact of radiatively interactive dust aerosols in the NASA GEOS-5 climate model: Sensitivity to dust particle shape and refractive index. *J. Geophys. Res. Atmos.,119, 753–786*, doi:10.1002/2013JD020046.

1193 Crosbie, E., Ziemba, L.D., Shook, M.A., Robinson, C.E., Winstead, E.L., et al. (2022).
1194 Measurement report: Closure analysis of aerosol–cloud composition in tropical maritime warm
1195 convection. *Atmos. Chem. Phys., 22, 13269–13302*, doi:10.5194/acp-22-13269-2022.
1196
1197 Das, S., Harshvardhan, H., Bian, H., Chin, M., Curci, G., et al. (2017), Biomass burning aerosol
1198 transport and vertical distribution over the South African-Atlantic region. *J. Geophys. Res.*
1199 *Atmos., 122, 6391–6415*, doi:10.1002/2016JD026421.
1200
1201 Dawson, K.W., Meskhidze, N., Burton, S.P., Johnson, M.S., Kacenelenbogen, M.S., Hostetler,
1202 C.A., & Hu, Y. (2017). Creating Aerosol Types from Chemistry (CATCH): A new algorithm to
1203 extend the link between remote sensing and models. *J. Geophys. Res., Atmos. 122, 12,366–*
1204 *12,392*, doi:10.1002/2017JD026913.
1205
1206 Dagan, G. & P. Stier (2020). Constraint on precipitation response to climate change by
1207 combination of atmospheric energy and water budgets. *npj Clim. Atmos. Sci. 3 (34)*.
1208 doi:10.1038/s41612-020-00137-8.
1209
1210 Dadashazar, H., Wnag, Z., Crosbie, E., Brunke, M., Zeng, X., et al. (2017). Relationships
1211 between giant sea salt particles and clouds inferred from aircraft physicochemical data. *J.*
1212 *Geophys. Res. Atmos., 122, 3421–3434*, doi:10.1002/2016JD026019.
1213
1214 Dawson, K.W., Ferrare, R.A., Moore, R.H., Clayton, M.D., Thorsen, T.J., & Eloranta, E.W.
1215 (2020). Ambient aerosol hygroscopic growth from combined Raman lidar and HSRL. *J.*
1216 *Geophys. Res. Atmos. 125, e2019JD031708*, doi:10.1029/2019JD031708.
1217
1218 Dee, D.P., S.M. Uppala, A.J. Simmons, P. Berrisford, P. Poli, S. Kobayashi, U., et al. (2011).
1219 The ERA-Interim reanalysis: configuration and performance of the data assimilation system. *Q.*
1220 *J. Royal Met. Soc. 137 (656), 553-597*, doi:10.1002/qj.828.
1221

de Leeuw, G., Andreas, E.L., Anguelova, M.D., Fairall, C.W., Lewis, E.R., O'Dowd, C., Schulz, M., &. Schwartz S.E. (2011). Production flux of sea spray aerosol. *Rev. Geophys. 49, RG2001*, doi:10.1029/2010RG000349.

Denjean, C., Formenti, P., Desboeufs, K., Desboeufs, K., Chevaillier, S., Triquet, S., et al. (2016). Size distribution and optical properties of African mineral dust after intercontinental transport. *J. Geophys. Res. Atmos., 121, 7117–7138*, doi:10.1002/2016JD024783.

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., et al. (2017). Global scale variability of the mineral dust long-wave refractive index: a new dataset of in situ measurements for climate modeling and remote sensing. *Atmos. Chem. Phys., 17, 1901–1929*, doi:10.5194/acp-17-1901-2017.

Dubovik, O., Lapyonok, T., Kaufman, Y.J., Chin, M., Ginoux, P., Kahn, R.A., & Sinyuk, A., (2008). Retrieving global sources of aerosols from satellites using inverse modeling. *Atmos. Chem. Phys. 8*, 209-250, doi:10.5194/ACP-8-209-2008.

Dubovik, O., Herman, M., Holdak, A., Lapyonok, T., Tanre, D., Deuze, J.L., et al. (2011). Statistically optimized inversion algorithm for enhanced retrieval of aerosol properties from spectral multi-angle polarimetric satellite observations. *Atmos. Meas. Tech. 4, 975–1018,* doi: 10.5194/amt-4-975-2011.

Ervens, B., Feingold, G., & Kreidenweis, S. M. (2005). The influence of water-soluble organic carbon on cloud drop number concentration. *J. Geophys. Res., 110, D18211*, doi:10.1029/2004JD005634.

Fanourgakis, G. S., Kanakidou, M., Nenes, A., Bauer, S.E., Bergman, T., Carslaw, K.S., et al. (2019). Evaluation of global simulations of aerosol particle and cloud condensation nuclei number, with implications for cloud droplet formation *Atmos. Chem. Phys., 19, 8591–8617*, doi: 10.5194/acp-19-8591-2019.

1253 Fast, J.D., Bell, D.M., Liu, J., Mei, F., Saliba, G., Shilling, J.E., et al. (2022). Using aircraft
1254 measurements to characterize subgrid-scale variability of aerosol properties near the ARM
1255 Southern Great Plains site. *Atmos. Chem. Phys., 22, 11217–11238,* doi:10.5194/acp-22-11217-
1256 2022.
1257
1258 Feingold, G., Yang, S., Hardesty, R.M., & Cotton, W.R. (1998). Feasibility of retrieving cloud
1259 condensation nucleus properties from doppler cloud radar, microwave radiometer, and lidar. *J.*
1260 *Atmos. Ocean Tech. 15, 1188-1195*. doi:10.1175/1520-0426.
1261
1262 Feingold, G., Cotton, W.R., Kreidenweis, S.M., & Davis, J.T. (1999). Impact of giant cloud
1263 condensation nuclei on drizzle formation in marine stratocumulus: Implications for cloud
1264 radiative properties. *J. Atmos. Sci., 56, 4100-4117*, doi:10.1175/1520-0469.
1265
1266 Feingold, G., Eberhard, W.L., Veron, D.E. & Previdi, M. (2003). First measurements of the
1267 Twomey aerosol indirect effect using ground-based remote sensors. *Geophys. Res. Lett., 30 (6),*
1268 *1287*, doi:10.1029/2002GL016633.
1269
1270 Feingold, G. (2003). Modeling of the first indirect effect: Analysis of measurement requirements.
1271 *Geophys. Res. Lett., 30 (19) 1997*, doi:10.1029/2003GL017967.
1272
1273 Fiedler, S., Kinne, S., Huang, W.T.K., Räisänen, P., O'Donnell, D., Bellouin, N., et al. (2019).
1274 Anthropogenic aerosol forcing – insights from multiple estimates from aerosol-climate models
1275 with reduced complexity. *Atmos. Chem. Phys., 19, 6821–6841*, doi: 10.5194/acp-19-6821-2019.
1276
1277 Flower, V.J.B., & Kahn, R.A. (2020a). Interpreting the volcanological processes of Kamchatka,
1278 based on multi-sensor satellite observations. *Remote Sens. Environ. 237, 111585*,
1279 doi:10.1016/j.rse.2019.111585.
1280
1281 Flower, V.J.B., & Kahn, R.A. (2020b). The evolution of Iceland volcano emissions, as observed
1282 from space. *J. Geophys. Res., 125, e2019JD031625*, doi:10.1029/2019JD031625.
1283

Forster, P., Storelvmo, T., Armour, K., Collins, W., Dufresne, J.-L., Frame, D., et al. (2021). The Earth's Energy Budget, Climate Feedbacks, and Climate Sensitivity. In Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change [Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S.L., Péan, C., Berger, S., et al. (eds.)]. *Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 923–1054*, doi:10.1017/9781009157896.009.

Foskinis, R., Nenes, A., Papayannis, A., Georgakaki, P., Kokkalis, P., Eleftheriadis, E., Komppula, M., Vratolis, S., Soupiona, O., Gini, M., Vakkari, V., Tombrou, M., & Bossioli, E. (2022). Towards reliable retrievals of cloud droplet number for non-precipitating planetary boundary layer clouds and their susceptibility to aerosol. *Front. Remote Sens., 3:958207*. doi:10.3389/frsen.2022.958207.

Fougnie, B., Chimot, J., Vazques-Navarro, M., Marbach, T., & Bojkov, B. (2020). Aerosol retrieval from space –how does geometry of acquisition impact our ability to characterize aerosol properties. *J. Quant. Spect. & Radiative Transf. 256, 107304*, doi: 10.1016/j.jqsrt.2020.107304.

Gasteiger, J. & Wiegner, M. (2018). MOPSMAP v1.0: a versatile tool for the modeling of aerosol optical properties. *Geosci. Model Dev., 11, 2739–2762*, doi:10.5194/gmd-11-2739-2018.

Gelaro, R., McCarty, W., Suárez, M.J., Todling, R., Molod, A., Takacs, L., et al. (2017). The Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2). *J. Clim. 30, 5419–5454*. doi:10.1175/JCLI-D-16-0758.1.

Georgakaki, P., Bougiatioti, A., Wieder, J., Mignani, C., Ramelli, F., Kanji, Z.A., et al. (2021). On the drivers of droplet variability in Alpine mixed-phase clouds. *Atmos. Chem. Phys., 21, 10993–11012*, doi:10.5194/acp-21-10993-2021.

Gettelman, A., Liu, X., Barahona, D., Lohmann, U., & Chen, C.C. (2012). Climate Impacts of Ice Nucleation. *J. Geophys. Ress 117, D20201*, doi:10.1029/2012JD017950.

1315 Gettelman, A., & Sherwood, S.C. (2016). Processes Responsible for Cloud Feedback. *Curr.*
1316 *Clim. Change Rep. 2, 179–189*, doi:10.1007/s40641-016-0052-8.
1317
1318 Gettelman, A., Geer, A.J., Forbes, R.M., Carmichael, G.R., Feingold, G., Posselt, D.J., et al.
1319 (2022). The Future of Earth System Prediction: Advances in Model-Data Fusion. *Science*
1320 *Advances 8 (14): eabn3488*, doi:10.1126/sciadv.abn3488.
1321
1322 Ghan, S., Guzman, G., and Abdul-Razzak, H. (1998). Competition between sea salt and sulfate
1323 particles as cloud condensation nuclei. *J. Atmos. Sci., 55, 3340–3347*, doi:10.1175/1520-0469.
1324
1325 Glassmeier, F., Hoffmann, F., Johnson, J.S., Yamaguchi, T., Carslaw, K.S., & Feingold, G.
1326 (2021). Ship-track-based assessments overestimate the cloud-mediated cooling effect of
1327 anthropogenic aerosol. *Science, 371, 485 – 489*, doi:10.1126/science.abd3980.
1328
1329 Gliß, J., Mortier, A., Schulz, M., Andrews, E., Balkanski, Y., Bauer, S.E., et al. (2021).
1330 AeroCom phase III multi-model evaluation of the aerosol life cycle and optical properties using
1331 ground- and space-based remote sensing as well as surface in situ observations. *Atmos. Chem.*
1332 *Phys. 21, 87–128*. doi:10.5194/acp-21-87-2021.
1333
1334 Go, S., Kima, J., Mok, J., Irie, H., Yoon,J., Torres, O., et al. (2020). Ground-based retrievals of
1335 aerosol column absorption in the UV spectral region and their implications for GEMS
1336 measurements. *Remt. Sens. Env. 245, 111759*, doi: 10.1016/j.rse.2020.111759.
1337
1338 Gonzales, M.E., Corral, A.F., Crosbie, E., Dadazhazar, H., Diskin, G.S., et al. (2022).
1339 Relationships between supermicrometer particle concentrations and cloud water sea salt and dust
1340 concentrations: analysis of MONARC and ACTIVATE data. *Environ. Sci. Atmosph. 2, 738*, doi:
1341 10.1039/d2ea00049k.
1342
1343 Gordon, T.D., Wagner, N.L., Richardson, M.S., Law, D.C., Wolfe, D., Eloranta, E.W., et al.
1344 (2015). Design of a novel open-path aerosol extinction cavity ringdown spectrometer. *Aerosol*
1345 *Sci. Tech., 49:9, 717-726*, doi:10.1080/02786826.2015.1066753.

Griffin, D., Sioris, C., Chen, J., Dickson, N., Kovachik, A., deGraaf, M., et al. (2020). The 2018 fire season in North America as seen by TROPOMI: aerosol layer height validation and evaluation of model-derived plume heights. *Atmos. Meas. Tech. 13, 1427–1445*, doi:10.5194/amt-13-1427-2020.

Gryspeerdt, E., Goren, T., Sourdeval, O., Quaas, J., Mülmenstädt, J., Dipu, S., Unglaub, C., Gettelman, A., and Christensen, M. (2019). Constraining the aerosol influence on cloud liquid water path. *Atmos*. *Chem. Phys., 19, 5331–5347*, doi:10.5194/acp-19-5331-2019.

Gryspeerdt, E., Goren, T., & Smith, T.W.P. (2021). Observing the timescales of aerosol–cloud interactions in snapshot satellite images. *Atmos. Chem. Phys., 21, 6093–6109*, doi:10.5194/acp-21-6093-202.1.

Guimond, S.R., Tian, L., Heymsfield, G.M., & Fraiser, S.J. 2014. Wind retrieval algorithms for the IWRAP and HIWRAP airborne doppler radars with applications to hurricanes. *J. Atmos. Ocean Tech. 31, 1189-1214*, doi:10.1175/JTECH-D-13-00140.1.

Gupta, P., Levy, R.C., Mattoo, S., Remer, L.A., Holz, R.E., & Heidinger, A.K. (2019). Applying the Dark Target aerosol algorithm with Advanced Himawari Imager observations during the KORUS-AQ field campaign. *Atmos. Meas. Tech., 12, 6557–6577*, doi:10.5194/amt-12-6557-2019.

Hamilton, D.S., Hantson, S., Scott, C.E., Kaplan, J.O., Pringle, K.J., et al. (2018). Reassessment of pre-industrial fire emissions strongly affects anthropogenic aerosol forcing. *Nat. Comm. 9, 3182*, doi:10.1038/s41467-018-05592-9.

Hammer, M.S., van Donkelaar, A., Martin, R.V., Li, C., Lyapustin, A., Sayer, A.M., Hsu, C.N., et al. (2019). Improved global estimates of fine particulate matter concentrations and trends derived from updated satellite retrievals, modeling advances, and additional ground-based monitors. *Environ. Sci. Tech. 54, 7879−7890*, doi:10.1021/acs.est.0c01764.

Hasekamp, O.P., Litvinov, P., & A. Butz, A. (2011). Aerosol properties over the ocean from PARASOL multiangle photopolarimetric measurements. *J. Geophys. Res. 116, D14204*, doi:10.1029/2010JD015469.

Haywood, J.M., Ramaswamy, V., & Donner, L.J. (1997). A limited-area-model case study of the effects of sub-grid scale Variations in relative humidity and cloud upon the direct radiative forcing of sulfate aerosol. *Geophys. Res. Lett. 24, 143-146*, doi: 10.1029/96GL03812.

Held, I.M., and Soden, B.J. (2006). Robust responses of the hydrological cycle to global warming. *J. Clim. 19, 5686-5698*, doi: 10.1175/JCLI3990.1.

Hélière, A., Gelsthorpe, R., Le Hors, L., & Toulemont, Y. (2012). ATLID, the atmospheric lidar on board the Earthcare Satellite. *Proc. SPIE 10584, 105842D*. doi:10.1117/12.2309095.

Hess, M., Koepke, P., & Schult, I. (1998). Optical Properties of Aerosols and Clouds: The software package OPAC. *Bull. Am. Met. Soc. 79, 831-844*, doi: 10.1175/1520-0477(1998)079.

Hoesly, R.M., Smith, S.J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Bolt, R.M., et al. (2018). Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS). *Geosci. Model Dev., 11, 369–408*, doi:10.5194/gmd-11-369-2018.

Holben, B. N., Eck, T. F., Slutsker, I., Tanré, D., Buis, J. P., Setzer, A., et al. (1998). AERONET — A federated instrument network and data archive for aerosol characterization *Remt. Sens. Environ., 66, 1–16*, doi:10.1016/S0034-4257(98)00031-5.

Holzer-Popp, T., de Leeuw, G., Griesfeller, J., Martynenko, D., Kluser, L., Bevan, S., et al. (2013). Aerosol retrieval experiments in the ESA Aerosol cci project. *Atmos. Meas. Tech., 6, 1919–1957*, doi: 10.5194/amt-6-1919-2013.

Huneeus, N., Schulz, M.,. Balkanski, Y., Griesfeller, J., Prospero, J.A., Kinne, S., et al. (2011). Global dust model intercomparison in AeroCom Phase I. *Atmos. Chem. Phys., 11, 7781– 7816*, doi:10.5194/acp-11-7781-2011.

Iacono, M.J., Delamere, J.S., Mlawer, E.J., Shephard, M.W., Clough, S.A., & Collins, W.D. (2008). Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer models. *J. Geophys. Res. 113, no. D13103*, doi:10.1029/2008JD009944.

Ichoku, C., & Ellison, L. (2014), Global top- down smoke- aerosol emissions estimation using satellite fire radiative power measurements. *Atmos. Chem. Phys., 14, 6, 643– 6,667*, doi: 10.5194/acp-14-6643-2014.

IPCC (2013). Summary for Policymakers. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., et al. (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Jenkins, S., Povey, A., Gettelman, A., Grainger, R., Stier, P., & Allen, M. (2022). Is anthropogenic global warming accelerating?. *J. Clim. 35, 4273-4290*, doi:10.1175/JCLI-D-22-0081.1.

Jeong, M-J., & Hsu, N.C. (2008). Retrievals of aerosol single-scattering albedo and effective aerosol layer height for biomass-burning smoke: Synergy derived from “A-Train” sensors. *Geophys. Res. Lett. 35, L24801*, doi:10.1029/2008GL036279.

Jiang, H., & Feingold, G. (2006). Effect of aerosol on warm convective clouds: Aerosol-cloudsurface flux feedbacks in a new coupled large eddy model. *J. Geophys. Res., 111, D01202*, doi:10.1029/2005JD006138.

Jung, E., Albrecht, B.A., Jonsson, H.H., Chen, Y.-C., Seinfeld, J.H., et al. (2015). Precipitation effects of giant cloud condensation nuclei artificially introduced into stratocumulus clouds. *Atmos. Chem. Phys., 15, 5645–5658*, doi:10.5194/acp-15-5645-2015.

Junghenn Noyes, K.T., Kahn, R.A., Sedlacek, A., Kleinman, L., Limbacher, J., Li, Z. (2020). Wildfire smoke particle properties and evolution, from space-based multi-angle imaging. *Remote Sens. 12, 769*, doi:10.3390/rs12050769.

Junghenn Noyes, K.T., Kahn, R.A., Limbacher, J.A., & Li, Z. (2022). Canadian and Alaskan wildfire smoke particle properties, their evolution, and controlling factors, using satellite observations. *Atmos. Chem. Phys. 22, 10267–10290,* doi:10.5194/acp-22-10267-2022.

Kacarab, M., Thornhill, K.L., Dobracki, A., Howell, S.G., O'Brien, J.R., Freitag, S., et al. (2020). Biomass burning aerosol as a modulator of droplet number in the southeast Atlantic region. *Atmos. Chem. Phys., 20, 3029–3040*, doi:10.5194/acp-20-3029-2020.

Kahn, R.A., Ogren, J.A., Ackerman, T.P., Bosenberg, J., Charlson, R.J., Diner, D.J., et al. (2004). Aerosol data sources and their roles within PARAGON, *Bull. Am. Met. Soc. 85, 1511-1522*, doi:10.1175/BAMS-85-10-1511.

Kahn, R.A., Chen, Y., Nelson, D.L., Leung, F-Y., Li, Q., Diner, D.J., & Logan, J.A. (2008). Wildfire smoke injection heights – Two perspectives from space. *Geophys. Res. Lett. 35,* doi:10.1029/2007GL032165.

Kahn, R.A. (2012). Reducing the uncertainties in direct aerosol radiative forcing. *Surv. Geophys. 33, 701–721*, doi:10.1007/s10712-011-9153-z.

Kahn, R.A., & Gaitley, B.J. (2015). An analysis of global aerosol type as retrieved by MISR. *J. Geophys. Res. Atmos. 120*, *4248-4281*, doi:10.1002/2015JD023322.

Kahn, R.A., Berkoff, T., Brock, C., Chen, G., Ferrare, R., Ghan, S., et al. (2017). *SAM-CAAM*: A concept for acquiring systematic aircraft measurements to characterize aerosol air masses. *Bull. Am. Meteoro. Soc. 2215-2228*, doi:10.1175/BAMS-D-16-0003.1.

Kahn, R.A., & Samset, B.H. (2022). Ch. 10, Remote sensing measurements of aerosol properties. In: Aerosols and Climate, K.S. Carslaw, Ed., Elsevier Publications, ISBN: 978-0-12-819766-0, pp. 823.

Kalashnikova, O.V., Garay, M.J., Martonchik, J.V., & Diner, D.J. (2013). MISR Dark Water aerosol retrievals: operational algorithm sensitivity to particle non-sphericity. *Atmos. Meas. Tech., 6, 2131–2154*, doi:10.5194/amt-6-2131-2013.

Kapustin, V.N., Clarke, A.D., Shinozuka, Y., Howell, S., Brekhovskikh, V., Nakajima, T., & Higurashi, A. (2006). On the determination of a cloud condensation nuclei from satellite: Challenges and possibilities*, J. Geophys. Res., 111, D04202*, doi:10.1029/2004JD005527.

Kim, D., M. Chin, M., Yu, H., Diehl, T., Tan, Q., Kahn, R.A., et al. (2014). Sources, sinks, and transatlantic transport of North African dust aerosol: A multi-model analysis and comparison with remote sensing data*. J. Geophys. Res. 119*, *6259-6277*, doi:10.1002/2013JD021099.

Kim, D., Chin, M., Yu, H., Pan, X., Bian, H., Tan, Q.,et al. (2019). Asia and trans-Pacific Dust: A multi-model and multi-remote sensing observation analysis. *J. Geophys. Res. 124*, doi:10.1029/2019JD030822.

Kim, M-H., Omar, A.H., Tackett, J.L., Vaughan, M.A., Winker, D.M., Trepte, C.R., et al. (2018). The CALIPSO version 4 automated aerosol classification and lidar ratio selection algorithm. *Atmos. Meas. Tech. 11, 6107–6135*, doi: 10.5194/amt-11-6107-2018.

Kinne, S., Lohmann, U., Feichter, J., Timmreck, C., Schulz, M., Ghan, S., et al. (2003). Monthly Averages of Aerosol Properties: A Global comparison among models, satellite data and AERONET ground data. *J. Geophys. Res, 108, 4634*. doi: 10.1029/2001JD001253.

Kinne, S., Schulz, M., Textor, C., Guibert, S., Balkanski, Y., Bauer, S. E., et al. (2006). An AeroCom initial assessment – optical properties in aerosol component modules of global models. *Atmos. Chem. Phys., 6, 1815–1834*. doi:10.5194/acp-6-1815-2006.

Kleinman, L. I., Sedlacek III, A. J., Adachi, K., Buseck, P. R., Collier, S., Dubey, M. K., et al. (2020). Rapid evolution of aerosol particles and their optical properties downwind of wildfires in the western US. *Atmos. Chem. Phys., 20, 13319–13341*, doi:10.5194/acp-20-13319-2020.

Koch, D, Schulz, M., Kinne, S., McNaughton, C., Spackman, J.R., Balkanski, Y., et al. (2009). Evaluation of black carbon estimations in global aerosol models. *Atmos. Chem. Phys., 9, 9001–9026*, doi:10.5194/acp-9-9001-2009.

Kokkola, H, T. Kühn, A. Laakso, T. Bergman, K.E.J. Lehtinen, T. Mielonen, A. (2018). SALSA2.0: The sectional aerosol module of the aerosol–chemistry–climate model ECHAM6.3.0-HAM2.3-MOZ1.0. *Geosci. Model Dev., 11, 3833–3863*, doi: 10.5194/gmd-11-3833-2018.

Koren, I., Kaufman, Y.J., Remer, L.A., Martins, J.V. (2004). Measurement of the effect of Amazon smoke on inhibition of cloud formation. *Science, 303, 1342–1345*, doi: 10.1126/science.1089424.

Koren, I., Kaufman, Y.J., Rosenfeld, D., Remer, L.A., & Rudich, Y. (2005). Aerosol invigoration and restructuring of Atlantic convective clouds. *Geophys. Res. Lett.,32, L14828*, doi:10.1029/2005GL023187.

Koren, I., Remer, L.A., Kaufman, Y.J., Rudich, Y., and Martins, J.V. (2007). On the twilight zone between clouds and aerosols. *Geophys. Res. Lett 34, L08805*, doi: 10.1029/2007GL029253.

Kylling, A., Vandenbussche, S., Capelle, V., Cuesta, J., Klüser, L., Lelli, L., et al. (2018). Comparison of dust layer heights from active and passive satellite sensors, *Atmos. Meas. Tech. 11, 2911-2936*, doi:10.5194/amt-11-2911-2018.

Laj, P., Bigi, A., Rose, C., Andrews, E., Myhre, C.L., Coen, M.C., et al. (2020). A global analysis of climate-relevant aerosol properties retrieved from the network of Global Atmosphere Watch (GAW) near-surface observatories. *Atmos. Meas. Tech., 13, 4353–4392*, doi:10.5194/amt-13-4353-2020.

Lau, K. M., Ramanathan, V., Wu, G.-X., Li, Z., Tsay, S. C., and Hsu, C.N. (2008). The joint aerosol-monsoon experiment: a new challenge for monsoon climate research. *Bull. Am. Meteorol. Soc. 89, 369–383*. doi:10.1175/BAMS-89-3-369

Lee, C.M., Cable, M.L., Hook, S.J., Green, R.O., Ustin, S.L., Mandl, D.J., & Middleton, E.M. (2015). An introduction to the NASA Hyperspectral InfraRed Imager (HyspIRI) mission and preparatory activities. *Remt. Sens. Environ. 167, 6-19*, doi:10.1016/j.rse.2015.06.012.

Lee, J.-Y., Marotzke, J., Bala, G., Cao, L., Corti, S., Dunne, J.P., et al. (2021). Future Global Climate: Scenario-Based Projections and Near- Term Information. In Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change [Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S.L., Péan, C., Berger, S., et al. (eds.)]. *Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 553–672*, doi:10.1017/9781009157896.006.

Lee, S.S., Feingold, G., McComiskey, A., Yamaguchi, T., Koren, I., Martins, J.V., & H. Yu, H. (2014), Effect of gradients in biomass burning aerosol on shallow cumulus convective circulations. *J. Geophys. Res. Atmos., 119, 9948–9964*, doi:10.1002/2014JD021819.

Levy, R. C., Remer, L. A., & Dubovik, O. (2007). Global aerosol optical properties and application to Moderate Resolution Imaging Spectroradiometer aerosol retrieval over land*, J. Geophys. Res. Atmos., 112, D13210*, doi:10.1029/2006JD007815.

Li, S., Kahn, R.A., Chin, M., Garay, M.J., & Liu, Y. (2015). Improving satellite-retrieved aerosol microphysical properties using GOCART data. *Atmos. Meas. Tech. 8, 1157–1171*. doi:10.5194/amt-8-1157-2015.

Li, J., Kahn, R. A., Wei, J., Carlson, B.E., Lacis, A. A., Li, Z., et al. (2020). Synergy of satellite- and ground-based aerosol optical depth measurements using an ensemble Kalman filter approach. *J. Geophys. Res. Atmos. 125, e2019JD031884*, doi:10.1029/2019JD031884.

Li, J., Carlson, B.E., Yung, Y.L., Lv, D., Hansen, J.E., Penner, J.E., et al. (2022). Scattering and Absorbing Aerosols in the Climate System. *Nature Rev. Earth Env.*, doi:10.1038/s43017-022-00296-7.

Lim, H., Choi, M., Kim, J., Kasai, Y.,& Chan, P.W. (2018). AHI/Himawari-8 Yonsei Aerosol Retrieval (YAER): Algorithm, validation and merged products. *Remote Sens., 10, 699*, doi:10.3390/rs10050699.

Liu, X., Penner, J.E., Ghan, S.J., & Wang, M. (2007). Inclusion of ice microphysics in the NCAR Community Atmospheric Model Version 3 (CAM3). *J. Climate 20, 4526-4547*.

Loeb, N.G., & Su, W. (2010). Direct aerosol radiative forcing uncertainty based on a radiative perturbation analysis. *J. Climate 23, 5288–5293*, doi:10.1175/2010JCLI3543.1.

Lu, Z., Wang, J., Xu, X., Chen, X., Kondragunta, S., Torres, O., et al. (2021). Hourly mapping of the layer height of thick smoke plumes over the western U.S. in 2020 severe fire season. *Front. Remote Sens. 2, 766628*, doi:10.3389/frsen.2021.766628.

Lyapustin, A., Wang, Y., Korkin, S., Kahn, R.A., & Winker, D. (2020). MAIAC thermal technique for smoke injection height from MODIS. *IEEE Geosci. Remt. Sens. Lett. 17 (5), 730-734*, doi: 10.1109/LGRS.2019.2936332.

Lyapustin, A., Go, S., Korkin, S., Wang, Y., Torres, O., et al. (2021). Retrievals of Aerosol Optical Depth and Spectral Absorption From DSCOVR EPIC. *Front. Remt. Sens. 2, 645794*, doi:10.3389/frsen.2021.645794.

Malm, W.C, & Hand, J.L. (2007). An examination of the physical and optical properties of aerosols collected in the IMPROVE program. *Atmos. Env. 41, 3407–3427*, doi:10.1016/j.atmosenv.2006.12.012.

Mann, G.W., Carslaw, K.S., Spracklen, D.V., Ridley, D.A., Manktelow, P.T., Chipperfield, M.P., et al. (2010). Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the UKCA composition-climate model. *Geosci. Model Dev., 3*, *519–551*, doi:10.5194/gmd-3-519-2010.

Marinescu, P.J., van den Heever, S.C., Heikenfeld, M., Barrett, A.I., Barthlott, C., Hoose, C., et al. (2021). Impacts of Varying Concentrations of Cloud Condensation Nuclei on Deep Convective Cloud Updrafts – A Multimodel Assessment. *J. Atmosph. Sci. 78, 1147-1172*, doi:10.1175/JAS-D-20-0200.1.

Marshak, A., Herman, J., Szabo, A., Blank, K., Carn, S., Cede, A., et al. (2018). Earth Observations from DSCOVR/EPIC Instrument. *Bulletin Amer. Meteor. Soc. (BAMS), 9, 1829-1850*, doi:10.1175/BAMS-D-17-0223.1.

Marshak, A., Ackerman, A., Da Silva, A., Eck, T., Holben, B.N., Kahn, R.A., et al. (2021). Aerosol properties in cloudy environments from remote sensing observations: review of current state of knowledge. *Bull. Am. Meteorol. Soc. E2177-E2197*, doi:10.1175/BAMS-D-20-0225.1.

Martin, S.T., Artaxo, P., Machado, L., Manzi, A.O., Souza, R.R.F., et al. (2017). The Green Ocean AMAZON experiment (GOAMAZON 2014/2015) observes pollution affecting gases, aerosols, couds, and rainfall of the rain forest. *Bulletin Amer. Meteor. Soc. (BAMS), 981-997,* doi: 10.1175/BAMS-D-15-00221.1.

Martins, J.V. (2016). Airborne Open Polar/Imaging Nephelometer for Ice Particles in Cirrus Clouds and Aerosols Field Campaign Report. *U.S. Dept. of Energy, Office of Science, ARM Climate Research Facility, DOE/SC-ARM-15-063*.

Mather, J. H., & Voyles, J. W. (2013). The ARM Climate Research Facility: A review of structure and capabilities, *Bull. Am. Meteorol. Soc.*, *94*(3)*, 377-392*. doi:10.1175/BAMS-D-11-00218.1.

McCoy, D.T., Tan, I., Hartmann, D.L., Zelinka, M.D., Storelvmo, T. (2016). On the relationships among cloud cover, mixed-phase partitioning, and planetary albedo in GCMs. *J. Adv. Model. Earth Syst., 8*, doi:10.1002/2015MS000589.

McFiggans, G., Artaxo, P., Baltensperger, U., Coe, H., Facchini, M.C., Feingold, G., et al. (2006). The effect of physical and chemical aerosol properties on warm cloud droplet activation. *Atmos. Chem. Phys., 6, 2593–2649*, doi:10.5194/acp-6-2593-2006.

Mechoso, C.R., Wood, R., Weller, R., Bretherton, C.S., Clarke, A.D., et al. (2014). Ocean-Cloud-Atmosphere-Land interactions in the southeastern Pacific: The VOCALS program. *Bull. Am. Meteoro. Soc. 95, 357-375*, doi: 10.1175/BAMS-D-11-00246.1.

Meyer, K., Platnick, S., & Zhang, Z. (2015). Simultaneously inferring above-cloud absorbing aerosol optical thickness and underlying liquid phase cloud optical and microphysical properties using MODIS. *J. Geophys. Res. Atm. 120, 5524-5547*, doi:10.1002/2015JD023128

McComiskey A., Schwartz, S.E., Schmid, B., Guan, H., Lewis, E.R., Ricchiazzi, P., & Ogren, J.A. (2008). Direct aerosol forcing: calculation from observables and sensitivities to inputs. *J. Geophys. Res. 113, D09202*, doi:10.1029/2007JD009170.

McNeill, J., Snyder, G., Weagle, C.L., Walsh, B., Bissonnette, P., Stone, E., et al. (2020). Large global variations in measured airborne metal concentrations driven by anthropogenic sources. *Nature Sci. Rep. 10:21817*, doi:10.1038/s41598-020-78789-y.

Menon, S., Hansen, J., Nazarenko, L., and Y. Luo, Y. (2002), Climate effects of black carbon aerosols in China and India. *Science, 297, 2250–2253*, doi: 10.1126/science.1075159.

Mishchenko, M.I., & Travis, L.D. (1997). Satellite retrieval of aerosol properties over the ocean using polarization as well as intensity of reflected sunlight. *J. Geophys. Res. 102 (D14) 16,989-17,013,* doi:10.1029/96JD02425.

Morales Betancourt, R., & Nenes, A. (2014). Understanding the contributions of aerosol properties and parameterization discrepancies to droplet number variability in a global climate model. *Atmos. Chem. Phys., 14, 4809–4826*, doi: 10.5194/acp-14-4809-2014.

Mortier, A., Gliss, J., Schulz, M., Aas, W., Andrews, E., Bian, H., et al. (2020). Evaluation of climate model aerosol trends with ground-based observations over the last two decades – an AeroCom and CMIP6 analysis. *Atmos. Chem. Phys., 20, 13355–13378*. doi:10.5194/acp-20-13355-2020.

Mülmenstädt, J. & Feingold, G. (2018). The radiative forcing of aerosol-cloud interactions in liquid clouds: Wrestling and embracing uncertainty. *Curr. Clim. Change Rep., 4, 23–40*, doi:10.1007/s40641-018-0089-y, 2018.

Myhre, G., Samset, B.H., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T.K., et al. (2013). Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations. *Atmos. Chem. Phys. 13, 1853–1877,* doi:10.5194/acp-13-1853-2013.

Nakajima, T., Higurashi, A., Kawamoto, K., & Penner, J.E. (2001). A possible correlation between satellite-derived cloud and aerosol microphysical parameters. *Geophys. Res. Lett. 28 (7), 1171-1174,* doi:10.1029/2000GL012186.

1681 Nazarenko, L., Rind, D., Tsigaridis, K., Del Genio, A.D., Kelley, M., & Tausnev, N. (2017).
1682 Interactive nature of climate change and aerosol forcing. *J. Geophys. Res. Atmos., 122, 3457–*
1683 *3480*, doi:10.1002/2016JD025809.
1684
1685 Nelson, D.L., Garay, M.J., Kahn, R.A., & Dunst, B.A. (2013). Stereoscopic height and wind
1686 retrievals for aerosol plumes with the MISR INteractive eXplorer (MINX). *Remote Sens. 5*,
1687 *4593-4628*, doi:10.3390/rs5094593.
1688
1689 Nguyen, H.N., Gudmundsson, A., & Martinsson, B.G. (2006). Design and calibration of a multi-
1690 channel aerosol sampler for tropopause region studies from the CARIBIC platform. *Aerosol Sci.*
1691 *Tech., 40:649–655*, doi:10.1080/02786820600767807.
1692
1693 Nicolae, D., Vasilescu, J., Talianu, C., Binietoglou, I., Nicolae, V., Andrei, S., & Antonescu, B.
1694 (2018). A neural network aerosol-typing algorithm based on lidar data. *Atmos. Chem. Phys., 18,*
1695 *14511–14537*, doi:10.5194/acp-18-14511-2018.
1696
1697 O'Gorman, P.A., Allan, R.P., Byrne, M.P., & Previdi, M. (2012). Energetic constraints on
1698 precipitation under climate change. *Survy. Geophys. 33, 585-608*, doi:10.1007/s10712-011-9159-
1699 6.
1700
1701 Pahlow, M., Feingold, G., Jefferson, A., Andrews, E., Ogren, J.A., Wang, J., et al. (2006).
1702 Comparison between lidar and nephelometer measurements of aerosol hygroscopicity at the
1703 Southern Great Plains Atmospheric Radiation Measurement site. *J. Geophys. Res., 111, D05S15*,
1704 doi:10.1029/2004JD005646.
1705
1706 Pan, X., Chin, M., Gautam, R., Bian, H., Kim, D., Colarco, P.R., et al. (2015). A multi-model
1707 evaluation of aerosols over South Asia: Common problems and possible causes. *Atmos. Chem.*
1708 *Phys. 15, 5903-5928*, doi:10.5194/acp-15-5903-2015.
1709

Papagiannopoulos, N., Mona, L., Amodeo, A., D'Amico, G., Gumà Claramunt, P., Pappalardo, G., et al. (2018). An automatic observation-based aerosol typing method for EARLINET. *Atmos. Chem. Phys., 18, 15879–15901*, doi:10.5194/acp-18-15879-2018.

Pappalardo, G., Amodeo, A., Apituley, A., Comeron, A., Freudenthaler, V., Linn, H.,et al. (2014). EARLINET: towards an advanced sustainable European aerosol lidar network. *Atmos. Meas. Tech., 7, 2389–2409*, doi:10.5194/amt-7-2389-2014.

Persad, G.G., Samset, B.H., & Laura J. Wilcox, L.J. (2022). Aerosols must be included in climate risk assessments, *Nature, 611 (24), 662-664*. doi:10.1038/d41586-022-03763-9.

Petrenko, M., Kahn, R.A., Chin, M., & Limbacher, J.A. (2017). Refined use of satellite aerosol optical depth snapshots to constrain biomass burning emissions in the GOCART model. *J. Geophys. Res. 122,* doi:10.1002/2017JD026693.

Pu, B., Ginoux, P., Guo, H., Hsu, C., Kimball, J., et al. (2020). Retrieving the global distribution of the threshold of wind erosion from satellite data and implementing it into the Geophysical Fluid Dynamics Laboratory land–atmosphere model (GFDL AM4.0/LM4.0). *Atmos. Chem. Phys., 20, 55–81*, doi:10.5194/acp-20-55-2020.

Quaas, J., Arola, A., Cairns, B., Christensen, M., Deneke, H., Ekman, A.M.L., et al. (2020). Constraining the Twomey effect from satellite observations: Issues and perspectives. *Atmos. Chem. Phys., 20, 15079–15099*, doi:10.5194/acp-20-15079-2020.

Quaas, J., Hailing, J., Smith, C., Albright, A.L., Aas, W., Bellouin, N., et al. (2022). Robust evidence for reversal of the trend in aerosol effective climate forcing. *Atmos. Chem. Phys., 22, 12221–12239*, doi: 10.5194/acp-22-12221-2022.

Ramaswamy, V., Boucher, O., Haigh, J., Hauglustaine, D., Haywood, J., et al. (2001). Ch. 6, Radiative forcing of climate change. In: Climate Change 2001: The Scientific Basis, J. T. Houghton et al., Eds., Cambridge University Press, 349–416.

Randerson, J. T., Chen, Y., van der Werf, G. R., & Morton, D. C. (2012). Global burned area and biomass burning emissions from small fires. *J. Geophys. Res. 117, G04012*, doi:10.1029/2012JG002128.

Randles, C. A., da Silva, A.M., Buchard, V., Colarco, P.R., Darmenov, A., Govindaraju, R.,et al. (2017). The MERRA-2 aerosol reanalysis, 1980–onward, Part I: System description and data assimilation evaluation. *J. Climate 30, 6823-6850*, doi:10.1175/jcli-d-16-0609.s1.

Reddington, C.L., Carslaw, K.S., Stier, P., Schutgens, N., Coe, H., Liu, D., et al. (2017). The global aerosol synthesis and science project (GASSP). *Bull. Am. Meteoro. Soc. 1857-1877*, doi:10.1175/BAMS-D-15-00317.1.

Redemann, J., Wood, R., Zuidema, P., Doherty, S.J., Luna, B., LeBlanc, S.E., et al. (2021). An overview of the ORACLES (ObseRvations of Aerosols above CLouds and their intEractionS) project: aerosol–cloud–radiation interactions in the southeast Atlantic basin. *Atmos. Chem. Phys., 21, 1507–1563*, doi:10.5194/acp-21-1507-2021.

Reid, J.S., R. Koppmann, R., Eck, T.F., & Eleuterio, D.P. (2005). A review of biomass burning emissions part II: intensive physical properties of biomass burning particles. *Atmos. Chem. Phys., 5, 799–825*, doi:1680-7324/acp/2005-5-799.

Reid, J.S., Reid, E.A., Walker, A., Piketh, S., Cliff, S., Al Mandoos, A., et al. (2008). Dynamics of southwest Asian dust particle size characteristics with implications for global dust research. *J. Geophys. Res., 113, D14212*, doi:10.1029/2007JD009752.

Reid, J.S., Hyer, E.J., Prins, E.M., Westphal, D.L., Zhang, J., et al. (2009). Global monitoring and forecasting of biomass-burning smoke: Description of and lessons from the Fire Locating and Modeling of Burning Emissions (FLAMBE) program. *IEEE J. Topics Applied Earth Obs. & Remt. Sens. 2 (3), 144-162*, doi:1939-1404/26.00.

1772 Remer, L.A., Kaufman, Y.J., Tanre, D., Mattoo, S., Chu, D.A., Martins, J.V., et al. (2005). The
1773 MODIS aerosol algorithm, products, and validation. *J. Atmos. Sci., 62, 947–973*,
1774 doi:10.1175/JAS3385.1.
1775
1776 Reutter, P., Su, H., Trentmann, J., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H., et al. (2009).
1777 Aerosol- and updraft-limited regimes of cloud droplet formation: influence of particle number,
1778 size and hygroscopicity on the activation of cloud condensation nuclei (CCN), *Atmos. Chem.*
1779 *Phys., 9, 7067–7080*, doi:10.5194/acp-9-7067-2009.
1780
1781 Rissman, T., Nenes, A., & Seinfeld, J.H. (2004). Chemical amplification (or dampening) of the
1782 Twomey effect: Conditions derived from droplet activation theory. *J. Atmos. Sci., 61(8), 919-*
1783 *930*, doi:10.1175/1520-0469.
1784
1785 Rose, C., Coen, M.C., Andrews, E., Lin, Y., Bossert, I., Myhre, C.L., et al. (2021). Seasonality of
1786 the particle number concentration and size distribution: a global analysis retrieved from the
1787 network of Global Atmosphere Watch (GAW) near-surface observatories. *Atmos. Chem. Phys.,*
1788 *21, 17185–17223*, doi:10.5194/acp-21-17185-2021.
1789
1790 Rosenfeld, D., Andreae, M.O., Asmi, A., Mian Chin, M., de Leeuw, G., Donovan, D.P., et al.
1791 (2014). Global observations of aerosol-cloud-precipitation-climate. *Rev. Geophys*. *52,*
1792 doi:10.1002/2013RG000441.
1793
1794 Rosenfeld, D., Zheng, Y., Hashimshoni, E., Pöhlker, M.L., Jeferson, A., Pöhlker, C., et al.
1795 (2016). Satellite retrieval of cloud condensation nuclei concentrations by using clouds as CCN
1796 chambers. *Proc. Nat. Acad. Sci. 113 (21)*, doi:10.1073/pnas.1514044113.
1797
1798 Rubin, J.I., Reid, J.S., Hansen, J.A., Anderson, J.L., Holben, B.N., Xian, P., et al. (2017).
1799 Assimilation of AERONET and MODIS AOT observations using variational and ensemble data
1800 assimilation methods and its impact on aerosol forecasting skill. *J. Geophys. Res. Atmos. 122,*
1801 *4967–4992*. doi:10.1002/2016JD026067.
1802

1803 Russell, P.B., Kacenelenbogen, M., Livingston, J.M., Hasekamp, O.P., Burton, S.P., Schuster,
1804 G.L., et al. (2014). A multiparameter aerosol classification method and its application to
1805 retrievals from spaceborne polarimetry. *J. Geophys. Res. Atmos. 119*,
1806 doi:10.1002/2013JD021411.
1807
1808 Samset, B. H., G. Myhre, M. Schulz, Balkanski, Y., Bauer, S., Berntsen, T.K., et al. (2013).
1809 Black carbon vertical profiles strongly affect its radiative forcing uncertainty. *Atmos. Chem.*
1810 *Phys. 13(5), 2423–2434*, doi:10.5194/acp-13-2423-2013.
1811
1812 Samset, B., Myhre, G. & Schulz, M. (2014). Upward adjustment needed for aerosol radiative
1813 forcing uncertainty. *Nature Climate Change 4, 230–232*. doi:10.1038/nclimate2170.
1814
1815 Sand, M., Samset, B.H., Balkanski, Y., Bauer, S., Bellouin, N., Berntsen, T.K., et al. (2017).
1816 Aerosols at the poles: an AeroCom Phase II multi-model evaluation. *Atmos. Chem. Phys. 17,*
1817 *12197–12218*. doi:10.5194/acp-17-12197-2017.
1818
1819 Sayer, A.M., Hsu, N.C., Lee, J., Kim, W.V., Burton, S., Fenn, M.A., et al. (2019). Two decades
1820 observing smoke above clouds in the south-eastern Atlantic Ocean: Deep Blue algorithm updates
1821 and validation with ORACLES field campaign data. *Atmos. Meas. Tech., 12, 3595–3627*,
1822 doi:10.5194/amt-12-3595-2019.
1823
1824 Sayer, A.M., Govaerts, Y., Kolmonen, P., Lipponen, A., Luffarelli, M., Mielonen, T., et al.
1825 (2020). A review and framework for the evaluation of pixel-level uncertainty estimates in
1826 satellite aerosol remote sensing. *Atmos. Meas. Tech. 13, 373–404*, doi:10.5194/amt-13-373-2020.
1827
1828 Schill, G.P., Froyd, K.D., Bian, H., Kupc, A., Williamson, C., Brock, C.A., et al. (2020).
1829 Widespread biomass burning smoke throughout the remote troposphere. *Nature Geosci.*,
1830 doi:10.1038/s41561-020-0586-1.
1831

Schroeder, P., Brewer, W.A., Choukulkar, A., Weickmann, A., Zucker, M, Holloway, M.W., & Sandberg, S. 2020. A compact, flexible, and robust micropulsed Doppler lidar. *J. Atmosph. Ocean Tech. 37, 1387-1402*, doi:10.1175/JTECH-D-19-0142.1.

Schulz, M., Textor, C., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., et al. (2006). Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations. *Atmos. Chem. Phys. 6(12), 5225–5246,* doi:10.5194/acp-6-5225-2006.

Schuster, G.L., & Trepte, C. (2021). Models, In situ, and Remote sensing of Aerosols (MIRA). https://science.larc.nasa.gov/wpcontent/uploads/sites/147/2021/11/MIRA_newsletter_20211027.pdf (last accessed 05/02/2022)

Schutgens, N.A.J., Bellouin, N., Bian, H., Boucher, O., Chin, M., Ghan, S., et al. (2013). Host model uncertainties in aerosol radiative forcing estimates: results from the AeroCom Prescribed intercomparison study. *Atmos. Chem. Phys., 13, 3245–3270*, doi: 10.5194/acp-13-3245-2013.

Schutgens, N., Tsyro, S., Gruspeerdt. E., Goto, D., Weigum, N.,, Schultz, M., & Stier, P. (2017). On the spatio-temporal representativeness of observations. *Atmos. Chem. Phys., 17, 9761-8780*, doi:10.5194/acp-17-9761-2017.

Schutgens, N., Sayer, A.M., Heckel, A., Hsu, N.C., Jethva, H., de Leeuw, G., et al. (2020). An AeroCom–AeroSat study: intercomparison of satellite AOD datasets for aerosol model evaluation. *Atmos. Chem. Phys. 20, 12431–12457*, doi:10.5194/acp-20-12431-2020.

Schutgens, N., Dubovik, O., Hasekamp, O., Torres, O., Jethva, H.,Leonard, P.J.T., et al. (2021). AEROCOM and AEROSAT AAOD and SSA study – Part 1: Evaluation and intercomparison of satellite measurements. *Atmos. Chem. Phys., 21, 6895–6917*, doi:10.5194/acp-21-6895-2021.

Seidel, F., & C. Popp (2012). Critical surface albedo and its implications to aerosol remote sensing. Atmos. Meas. Tech., 5, 1653–1665, doi: 10.5194/amt-5-1653-2012.

1863 Seinfeld, J.H., Bretherton, C.S., Carslaw, K.S., Coe, H., DeMott, P.J., Dunlea, E.J., et al. (2016).
1864 Improving our fundamental understanding of the role of aerosol-cloud interactions in the climate
1865 system. *Proc. Nat. Academy Sci*. *113 (21),* 5781–5790, doi:10.1073/pnas.1514043113.
1866
1867 Sekiyama, T. T., Tanaka, T. Y., Shimizu, A., & Miyoshi, T. (2010). Data assimilation of
1868 CALIPSO aerosol observations. *Atmos. Chem. Phys., 10, 39–49*. doi:10.5194/acp-10-39-2010.
1869
1870 Shen, Y., Virkkula, A., Ding, A., Luoma, K., Keskinen, H., Aalto, P. P., et al. (2019). Estimating
1871 cloud condensation nuclei number concentrations using aerosol optical properties: role of
1872 particle number size distribution and parameterization, *Atmos. Chem. Phys., 19, 15483–15502*,
1873 doi:10.5194/acp-19-15483-2019.
1874
1875 Sheridan, J.P., Andrews, E., Ogren, J.A., Tackett, J.L., & Winker, D.M., (2012). Vertical profiles
1876 of aerosol optical properties over Central Illinois and comparison with surface and satellite
1877 measurements. *Atmos. Chem. Phys., 12, 11695–11721*, doi:10.5194/acp-12-11695-2012.
1878
1879 Shindell, D.T., Lamarque, J.-F., Schulz, M., Flanner, M., Jiao, C., Chin, M., et al. (2013).
1880 Radiative forcing in the ACCMIP historical and future climate simulations. *Atmos. Chem. Phys.,*
1881 *13, 2939–2974*, doi: 10.5194/acp-13-2939-2013.
1882
1883 Shinozuka, Y., Clarke, A.D., Nenes, A., Jefferson, A., Wood, R., McNaughton, C.S., et al.
1884 (2015). The relationship between cloud condensation nuclei (CCN) concentration and light
1885 extinction of dried particles: indications of underlying aerosol processes and implications for
1886 satellite-based CCN estimates. *Atmos. Chem. Phys., 15, 7585-7604*, doi:10.5194/acp-15-7585-
1887 2015.
1888
1889 Smirnov, A, Holben, B.N., Giles, D.M., Slutsker, I., O'Neill, N.T., Eck, T.F., et al. (2011).
1890 Maritime aerosol network as a component of AERONET – First results and comparison with
1891 global aerosol models and satellite retrievals. *Atmos. Meas. Tech. 4*, 583-597, doi:10.5194/amt-4-
1892 583-2011.
1893

Snider, G., Weagle, C.L., Martin, R.V., van Donkelaar, A., Conrad; K., Zwicker, M., et al. (2015). SPARTAN: A global network to evaluate and enhance satellite-based estimates of ground-level aerosol for global health applications. *Atmos. Meas. Tech. 8, 505-521*, doi:10.5194/amt-8-505-2015.

Soden, B., and Chung, E.-S. (2017). The Large-Scale Dynamical Response of Clouds to Aerosol Forcing. *J Clim. 30(21), 8783–8794*, doi:10.1175/JCLI-D-17-0050.1.

Sogacheva, L., Popp, T., Sayer, A. M., Dubovik, O., Garay, M. J., Heckel, A., et al. (2020). Merging regional and global aerosol optical depth records from major available satellite products, *Atmos. Chem. Phys., 20, 2031–2056*, https://doi.org/10.5194/acp-20-2031-2020.

Sorooshian, A., Anderson, B., Bauer, S.E., Braun, R.A., Cairns, B., et al. (2019), Aerosol-Cloud-Meteorology interaction airborne field investigations: Using Lessons Learned from the U.S. West Coast in the Design of ACTIVATE off the U.S. East Coast. *Bull. Am. Meteor. Soc. 100, 1511–1528*, doi:10.1175/BAMS-D-18-0100.1.

Sorooshian, A., Atkinson, J., Ferrare, R., Hair, J., & Ziemba, L. (2021). Taking flight to study clouds and climate. *Eos,102,* doi:10.1029/2021EO158570.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., et al. (2005). The aerosol-climate model ECHAM5-HAM. *Atmos. Chem. Phys., 5, 1125–1156*, doi: 10.5194/acp-5-1125-2005.

Stier, P., Schutgens, N.A.J., Bellouin, N., Bian, H., Boucher, O., Chin, M., et al. (2013). Host model uncertainties in aerosol radiative forcing estimates: results from the AeroCom Prescribed intercomparison study. *Atmos. Chem. Phys., 13, 3245–3270*, doi: 10.5194/acp-13-3245-2013.

Stier, P. (2016). Limitations of passive remote sensing to constrain global cloud condensation nuclei. *Atmos. Chem. Phys., 16, 6595–6607*, doi:10.5194/acp-16-6595-2016.

1925 Sullivan, S.C., Lee, D., Oreopoulos, L., & Nenes, A. (2016). Role of updraft velocity in temporal
1926 variability of global cloud hydrometeor number. *Proc. Nat. Acad. Sci. 113 (21), 5791-5796*,
1927 doi:10.1073/pnas.1514039113.
1928
1929 Tao, W.-K., Chen, J.-P., Li, Z., Wang, C., & Zhang, C. (2012). Impact of aerosols on convective
1930 clouds and precipitation. *Rev. Geophys., 50, RG2001*, doi:10.1029/2011RG000369.
1931
1932 Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., et al. (2006). Analysis
1933 and quantification of the diversities of aerosol life cycles within AeroCom, *Atmos. Chem. Phys.,*
1934 *6*, 1777–1813.
1935
1936 Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., et al. (2007). The effect
1937 of harmonized emissions on aerosol properties in global models – an AeroCom experiment.
1938 *Atmos. Chem. Phys., 7, 4489–4501*, doi:10.5194/acp-7-4489-2007.
1939
1940 Thomas, G.E., Poulsen, C.A., Sayer, A.M., Marsh, S.H., Dean, S.M., Carboni, E., et al. (2009).
1941 The GRAPE aerosol retrieval algorithm. *Atmos. Meas. Tech., 2, 679–701*, doi:10.5194/amt-2-
1942 679-2009.
1943
1944 Thorsen, T.J., Winker, D.M., & Ferrare, R.A. (2021). Uncertainty in observational estimates of
1945 the aerosol direct radiative effect and forcing. *J. Climate 34, 195-213*, doi:10.1175/JCLI-D-19-
1946 1009.1.
1947
1948 Tsigaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., et al.
1949 (2014). The AeroCom evaluation and intercomparison of organic aerosol in global models.
1950 *Atmos. Chem. Phys., 14, 10845–10895*, doi:10.5194/acp-14-10845-2014.
1951
1952 Toon, O. B., Maring, H., Dibb, J., Ferrare, R., Jacob, D.J., Jensen, E.J., et al. (2016). Planning,
1953 implementation, and scientific goals of the Studies of Emissions and Atmospheric Composition,
1954 Clouds and Climate Coupling by Regional Surveys (SEAC4RS) field mission. *J. Geophys. Res.*
1955 *Atmos., 121,* doi:10.1002/2015JD024297.

Torres, O., Jethva, H., & Bhartia, P.K. (2012). Retrieval of aerosol optical depth above clouds from OMI observations: sensitivity analysis and case studies. *J. Atmos. Sci. 69, 1037–1053*, doi:10.1175/JASD-11-0130.1.

Twomey, S. (1974). Pollution and the planetary albedo. *Atmos. Env. 8, 1251-1256*, doi: 10.1016/0004-6981(74)90004-3.

Twomey, S. (1977). The influence of pollution on the short wave albedo of clouds. *J. Atmos. Sci., 34, 1149–1152*, doi:10.1175/1520-0469(1977)034.

Val Martin, M. Kahn, R.A., & Tosca, M. (2018). A global climatology of wildfire smoke injection height derived from space-based multi-angle imaging. *Remote Sensing 10, 1609*, doi:10.3390/rs10101609.

Vandenbussche, S., Kochenova, S., Vandaele, A.C., Kumps, N., & De Maziere, M. (2013). Retrieval of desert dust aerosol vertical profiles from IASI measurements in the TIR atmospheric window. *Atmos. Meas. Tech., 6, 2577–2591*, doi:10.5194/amt-6-2577-2013.

van der Werf, G.R., Randerson, J.T., Giglio, L., van Leeuwen, T.T., Chen, Y., et al. (2017). Global fire emissions estimates during 1997–2016. *Earth Syst. Sci. Data, 9, 697–720*, doi: 10.5194/essd-9-697-2017.

Van Donkelaar, A., Martin, R.V., Brauer, M., Kahn, R.A., Levy, R.C., Verduzco, C., & Villeneuve, P. (2010). Global estimates of average ground-level fine particulate matter concentrations from satellite-based aerosol optical depth. *Environ. Health Perspect. 118, 847-855*, doi:10.1289/EHP.0901623.

Van Donkelaar, A., M. Hammer, M., Bindle, L., Brauer, M., Brook, J., Garay, M., et al. (2021). Monthly global estimates of fine particulate matter and their uncertainty. *Environ. Sci. Tech. 55, 15287−15300*, doi:10.1021/acs.est.1c05309.

Vernon, C.J., Bolt, R., Canty, T., & Kahn, R.A. (2018). The impact of MISR-derived injection-height initialization on wildfire and volcanic plume dispersion in the HySPLIT model. *Atmos. Meas. Tech*. *11*, *6289–6307*, doi:10.5194/amt-11-6289-2018.

Vicent, J., Verrelst, J., Sabater, N., Alonso, L., Rivera-Caicedo, J.P., Martino, L., et al. (2020). Comparative analysis of atmospheric radiative transfer models using the Atmospheric Look-up table Generator (ALG) toolbox (version 2.0). *Geosci. Model Dev., 13, 1945–1957*, doi:10.5194/gmd-13-1945-2020.

Vogelmann, A., G. M. McFarquhar, J. A. Ogren, D. D. Turner, J. M. Comstock, G. Feingold, et al. (2012). RACORO extended-term, aircraft observations of boundary-layer clouds, *Bull. Amer. Meteor. Soc., 93, 861-878*, doi:10.1175/BAMS-D-11-00189.1.

Wang, J., Aegerter, C., Xu, X., & Szykman, J.J. (2016). Potential application of VIIRS Day/Night Band for monitoring nighttime surface PM2.5 air quality from space. *Atmosph. Environ. 124, 55-63*, doi:10.1016/j.atmosenv.2015.11.013.

Wang, S., Wang Q., & Feingold, G. (2003). Turbulence, condensation, and liquid water transport in numerically simulated nonprecipitating stratocumulus clouds. *J. Atmos. Sci., 60, 262–278*, doi:10.1175/1520-0469.

Watson-Parris, D., & Smith, C.J. (2022). Large uncertainty in future warming due to aerosol forcing. *Nat. Clim. Chang. 12, 1111–1113*, doi:10.1038/s41558-022-01516-0.

Wells, K.C., Martins, J.V., Remer, L.A., Kreidenweis, S.M., & G. L. Stephens G.L. (2012). Critical reflectance derived from MODIS: Application for the retrieval of aerosol absorption over desert regions, *J. Geophys. Res., 117, D03202*, doi:10.1029/2011JD016891.

Welton, E.J., Campbell, J.R., Spinhirne, J.D., & Scott, V.S. (2001). Global monitoring of clouds and aerosols using a network of micro-pulse lidar systems. *Proc. SPIE 4153, 151-158,* doi:10.1117/12.417040.

Wen, G., Marshak, A.,Cahalan, R.F., Remer, L., & Kleidman, R.G. (2007). 3D aerosol-cloud radiative interaction observed in collocated MODIS and ASTER images of cumulus cloud fields. *J. Geophys. Res., 112, D13204*, doi:10.1029/2006JD008267.

Werdell, P.J., Behrenfeld, M.J., Bontempi, P.S., Boss, E., Caorms. B. et al. (2019). The plankton, aerosol, cloud, ocean ecosystem mission: Status, science, advances. *Bull. Am. Meteorol. Soc. 1775-1794*, doi:10.1175/BAMS-D-18-0056.1.

Wiggins, E.B., Soja, A.J., Gargulinski, E., Halliday, H.S., Pierce, R.B., et al. (2020). High Temporal Resolution Satellite Observations of Fire Radiative Power Reveal Link Between Fire Behavior and Aerosol and Gas Emissions. *Geophys. Res. Lett. 47, e2020GL090707*, doi:10.1029/2020GL090707

Williams, A.I.L., Stier, P., Dagan, G., & Watson-Parris, D. (2022). Strong control of effective radiative forcing by the spatial pattern of absorbing aerosol. *Nature Climate Change (12), 735-742*, doi:10.1038/s41558-022-01415-4.

Wilson, J.C., Lafleu, B.G., Hilbert, H., Seebaugh, W.R., Fox, J., Gesler, D.W., Brock, C.A., Huebert, B.J., & Mullen, J. (2004) Function and performance of a low turbulence inlet for sampling supermicron particles from aircraft platforms. *Aerosol Sci. Tech., 38(8), 790-802*, doi:10.1080/027868290500841.

Winker, D.M., Vaughan, M.A., Omar, A., Hu, Y., & Powell, K.A. (2009). Overview of the CALIPSO mission and CALIOP data processing algorithms. *J. Atmos. Ocean Technol. 26, 2310–2323*, doi:10.1175/2009JTECHA1281.1.

Witek, M.L., Garay, M.J., Diner, D.J., Bull, M.A., & Seidel, F.C. (2018). New approach to the retrieval of AOD and its uncertainty from MISR observations over dark water. *Atmos. Meas. Tech. 11, 429–439*, doi:10.5194/amt-11-429-2018.

Xian, P., Reid, J.S., Hyer, E.J., Sampson, C.R., Rubin, J.I., Ades, M. et al. (2019). Current state of the global operational aerosol multi-model ensemble: An update from the International Cooperative for Aerosol Prediction (ICAP). *Q. J. Royal Meteoro. Soc. 145 (Suppl.), 176–209*, doi:10.1002/qj.3497.

Xue, H., Feingold, G., & Stevens, B. 2008. Aerosol effects on clouds, precipitation, and the organization of shallow cumulus convection. *J. Atmos. Sci., 65, 392–406*, doi:10.1175/2007JAS2428.1.

Yang, C.K., Chiu, J.C., Marchak, A., Feingold, G., Varnai, T., Wen, G., et al. (2022). Near-cloud aerosol retrieval Uusing machine learning techniques, and implied direct radiative effects. *Geophys. Res. Lett. 49, e2022GL098274*, doi:10.1029/2022GL098274.

Yokelson, R., Crounse, J. D., DeCarlo, P. F., Karl, T., Urbanski, S., Atlas, E., et al. (2009). Emissions from biomass burning in the Yucatan. *Atmos. Chem. Phys., 9, 5785-5812,* doi:10.5194/acp-9-5785-2009.

Yorks, J.E., McGill, M.J., Palm, S.P., Hlavka, D.L., Selmer, P.A., Nowottnick, E.P., et al. (2016). An overview of the CATS level 1 processing algorithms and data products. *Geophys. Res. Lett., 43, 4632–4639*, doi:10.1002/2016GL068006.

Zamora, L.M., & Kahn, R.A. (2020). Saharan dust aerosols change deep convective cloud prevalence, possibly by inhibiting marine new particle formation. *J. Climate 33, 9467-9477*, doi:10.1175/JCLI-D-20-0083.1

Zender, C.S., Bian, H., & Newman, D. (2003). Mineral Dust Entrainment and Deposition (DEAD) model: Description and 1990s dust climatology*, J. Geophys. Res., 108(D14), 4416*, doi:10.1029/2002JD002775, 2003.

Zhang, H., Kondragunta, S., Laszlo, I., & Zhou, M. (2020a). Improving GOES Advanced Baseline Imager (ABI) aerosol optical depth (AOD) retrievals using an empirical bias correction algorithm. *Atmos. Meas. Tech. 13, 5955–5975*, doi:10.5194/amt-13-5955-2020.

Zhang, J., Spurr, R.J.D., Reid, J.S., Xian, P.,Colarco, P.R., Campbell, J.R., et al. (2020b). Development of an Ozone Monitoring Instrument (OMI) aerosol index (AI) data assimilation scheme for aerosol modeling over bright surfaces – a step toward direct radiance assimilation in the UV spectrum. *Geosci. Model Dev, 14, 27–42*, doi:10.5194/gmd-14-27-2021.

Zhang, J., Campbell, J.R., Reid, J.S., Westphal, D.L., Baker, N.L., Campbell, W.F., & Hyer, E.J. (2011). Evaluating the impact of assimilating CALIOP-derived aerosol extinction profiles on a global mass transport model. *Geophys. Res. Lett., 38, L14801*, doi:10.1029/2011GL04773.

Zhu, L., Val Martin, M., Hecobian, A., Deeter, M.N., Gatti, L.V., Kahn, R.A., & Fischer, E.V. (2018a). Development and implementation of a new biomass burning emissions injection height scheme for the GEOS-Chem model. *Geosci. Model Develop*. 11, 4103–4116, doi:10.5194/gmd-11-4103-2018.

Zhu, Y., Rosenfeld, D., & Li, Z. (2018b). Under what conditions can we trust retrieved cloud drop concentrations in broken marine stratocumulus? *J. Geophys. Res. Atmos. 123, 8754–8767*, doi:.

Zieger, P., Fierz-Schmidhauser, R., Weingartner, E., & Baltensperger, U. (2013). Effects of relative humidity on aerosol light scattering: results from different European sites. *Atmos. Chem. Phys., 13, 10609–10631*, doi:10.5194/acp-13-10609-2013

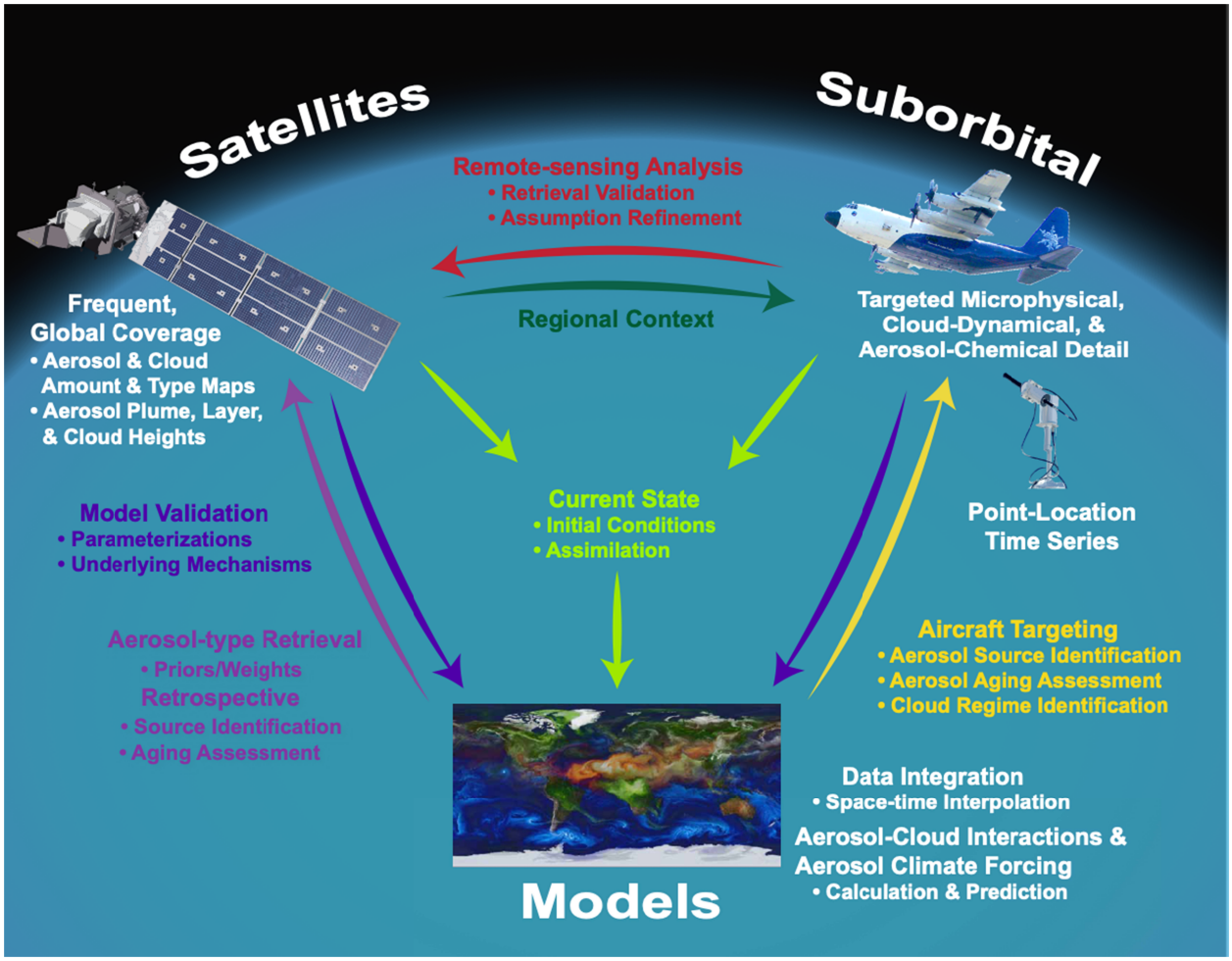

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Figure 1.

Satellites

Frequent, Global Coverage
- Aerosol & Cloud Amount & Type Maps
- Aerosol Plume, Layer, & Cloud Heights

Suborbital

Targeted Microphysical, Cloud-Dynamical, & Aerosol-Chemical Detail

Point-Location Time Series

Remote-sensing Analysis
- Retrieval Validation
- Assumption Refinement

Regional Context

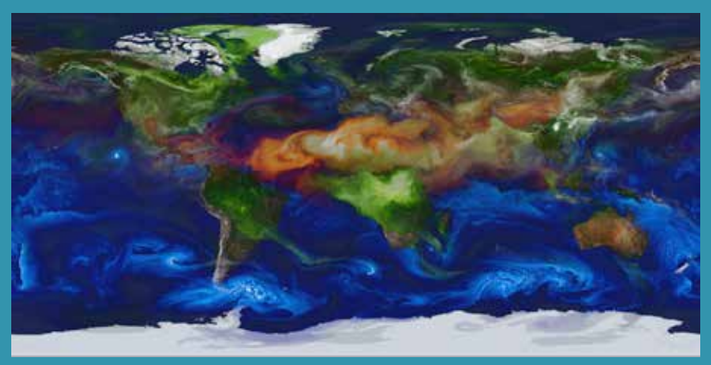
Current State
- Initial Conditions
- Assimilation

Model Validation
- Parameterizations
- Underlying Mechanisms

Aerosol-type Retrieval
- Priors/Weights

Retrospective
- Source Identification
- Aging Assessment

Aircraft Targeting
- Aerosol Source Identification
- Aerosol Aging Assessment
- Cloud Regime Identification



Models

Data Integration
- Space-time Interpolation

Aerosol-Cloud Interactions & Aerosol Climate Forcing
- Calculation & Prediction