Improved retrievals of cloud condensation nuclei (CCN) number concentration over the ocean to reduce indirect forcing uncertainties in climate models

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We propose that satellite retrieval of CCN number concentrations within a factor of two of the measured values are required to constrain anthropogenic aerosol indirect effect in climate models. Such high accuracy in CCN retrievals can be achieved by the most capable High Spectral Resolution Lidar that is envisioned for the NASA ACE mission.

Introduction

Atmospheric aerosols play a large role in air quality and human-induced climate change. After many decades of research, aerosol–cloud interactions (ACI) remain the largest source of uncertainty in current estimates of global radiative forcing [*IPCC,* 2013]. This is mainly due to the fact that aerosols, clouds and ecosystem elements are intricately linked in the Earth System via multiple interrelated forcings and feedbacks. Therefore, narrowing the uncertainties in ACI will require considering all elements together from both a mechanistic and observational standpoint. The 2007 NRC Decadal Survey Report identified the Aerosol, Cloud, and ocean Ecosystem (ACE) mission as an essential Tier 2 mission that, based on its inter-disciplinary nature, can address challenges associated with ACI. Because effects on climate are estimated from the difference between model simulations with present-day and with preindustrial aerosol and precursor emissions, **accurate representation of number concentrations of cloud condensation nuclei (CCN) associated with both natural background and man-made aerosols separately is critical for better assessment of anthropogenic aerosol effects** [*Wang and Penner*, 2009; *Hoose et al*., 2009; *Meskhidze et al*., 2011; *Carslaw et al*., 2013].

To narrow the uncertainties in representing key processes in climate models, a comprehensive dataset is needed to document daily CCN concentration over different regions (at horizontal and vertical resolution relevant to shallow marine clouds, e.g., near 1 km and 15 m, respectively) and under a range of synoptic conditions. As such observations cannot be achieved globally using in situ measurements, *satellite-based CCN estimates* emerge as an extremely attractive option for narrowing the gap in the current estimates of radiative forcing responsible for the ongoing climate change.

In response to current DS-RFI2 we identify **satellite-based CCN estimates over the ocean for natural background and man-made aerosols (separately) as one of the key challenges for Earth System Science**. Recognizing the observational interdependencies of aerosols, clouds and ecosystem elements, we propose that the ACE mission - with a single set of consistent satellite sensors such as High Spectral Resolution Lidar (HSRL), Ocean Color Sensor, the Polarimeter, and the Dual Frequency Doppler Radar - is best suited to address this challenge. The HSRL (in conjunction with the polarimeter) can provide satellite-based estimates of CCN concentrations at cloud altitudes within a factor of 2 of observed values for the majority of the globe; something that cannot be achieved by currently available sensors [*Shinozuka et al*., 2015; *Stier*, 2015]. Satellite-based CCN estimates for natural and man-made aerosols are cross cutting among the Earth System Science themes identified in the second 2017 Decadal Survey Request for Information (DS-RFI2):

I. Global Hydrological Cycles and Water Resources
II. Weather and Air Quality
III. Marine and Terrestrial Ecosystems and Natural Resource Management
IV. Climate Variability and Change

1. Science and Application Target

About 45 % of the variance in model-predicted aerosol indirect forcing since preindustrial time arises from the uncertainties in natural emissions, with only 34 % of the variance associated with anthropogenic emissions [*Carslaw et al.,* 2014]. The largest portion of this uncertainty, according to Fig. 1, occurs over the oceans, and is related to the estimates of CCN number concentrations for marine stratiform clouds. This large contribution of CCN uncertainty to the mean forcing uncertainty range is associated with high susceptibility (cloud albedo sensitivity to changes in cloud droplet concentrations) for marine stratocumulus [*Twomey*, 1991]. Table 1 shows aerosol sources (both natural and man-made) that are responsible for the uncertainty range in model-predicted aerosol indirect forcing for marine stratiform clouds. Potential feedbacks, i.e., a wide range of stress factors caused by human activities that can incite complex interactions between the land, ocean and atmosphere, can also affect CCN number concentration over the oceans and cause further increase in climate prediction uncertainty.

For a first-order estimate of the relationship between uncertainty in CCN and uncertainty in aerosol indirect radiative forcing, it is assumed that CCN concentration scales in direct proportion with anthropogenic emissions [*Carslaw et al*., 2014]. Simple calculations shown on Fig. 2 suggest that **to reduce the uncertainty in anthropogenic aerosol radiative forcing below ~1.5 W m^{-2}, CCN number concentration over the oceans needs to be constrained to better than a factor of 2**.

2. Geophysical Variables Needed to Constrain CCN Number, Sources, and Impacts on Cloud Properties Over The Oceans

Satellites are, and will likely remain, the dominant players for improved characterization of ACI in a changing climate, because they provide global, long-term information about the spatiotemporal variability of aerosols and other parameters affecting CCN concentrations over the oceans. Satellite-based CCN estimates are typically derived by using a relationship between the number concentration of CCN and light extinction. There is **a range of past, existing and planned remote sensing instruments supported through U.S. and international programs** such as MODIS, MISR, AATSR, PARASOL, MERIS, SeaWiFS, CALIPSO, GPM, SAGE-III/ISS, CATS, and PACE and ground-based systems including the MAN, a ship-borne data acquisition initiative complementing island-based AERONET measurements that can be used for characterization of CCN over the oceans. However, **none of these sensors can achieve coincident (in time and space) retrievals of cloud properties, vertically-resolved aerosol information, ocean sub-surface data, and ocean biological parameters**, i.e., parameters essential for quantitative characterization of both natural and anthropogenic ACI. Moreover, current satellites either do not provide the data or provide them at low signal-to-noise ratio (SNR) not high enough for retrieval of many ocean ecosystem processes and aerosol speciation and loadings over the oceans. Therefore, **uncertainty in CCN concentration derived through passive remote sensing remains larger than a factor of 2** [*Shinozuka et al*., 2015; *Stier*, 2015], imposing a fundamental limit on the ability to constrain aerosol-cloud interactions in state-of-the-science climate models. The primary product from passive remote sensing of aerosol properties is aerosol optical depth (AOD). Many previous studies analyzing satellite observations of aerosol and cloud properties have used AOD as a proxy for CCN number, and explored the relationships between AOD and cloud properties. However, the passive satellite-derived CCN–AOD relationship is complicated by several factors. First, passive sensors lack the ability to vertically resolve the aerosol or CCN number: whereas the CCN most relevant to ACI are located at the cloud base altitude, the AOD is defined for the entire vertical column. Second, passive satellites depend on daylight and have difficulty in making retrievals over bright surfaces. Making CCN retrievals over the polar regions is fundamental for improved characterization of high-latitude clouds. Third, clouds interfere with passive sensor observations, and so AOD can be accurately measured only under clear-sky conditions; yet the air mass interacting with clouds may be kilometers away from, or hours after, clear-sky satellite measurements of AOD. These differences matter because aerosol spatiotemporal distribution is generally inhomogeneous [*Shinozuka et al*., 2015]. Fourth, most CCN are smaller aerosol particles (with diameters between 0.05 and 0.15 μm), while most aerosol-related light extinction (when vertically integrated is the AOD) at mid-visible wavelengths is caused by larger aerosol particles (with diameters between 0.5 and 10 μm), making AOD a poor proxy for CCN number in many circumstances. Over most of the globe, the correlation coefficients between AOD and CCN at a supersaturation of 0.2% at cloud base are below 0.5, and AOD variability explains only 25% of the CCN variance globally [*Stier*, 2015]. For CCN concentrations below 100 cm^{-3} (i.e., clean regions where clouds are most susceptible to changes in CCN), AOD-estimated CCN could be within a factor of 10 of measured CCN [*Shinozuka et al*., 2015]. This uncertainty in passive satellite derived CCN number concentration must be reduced considerably in order to improve constraints on CCN number and further advance climate change research.

3. Key Requirements For Reducing CCN Number Uncertainty Over The Ocean

To reduce uncertainty in satellite-based CCN estimates, the CCN number concentration needs to be retrieved:

1. At airmasses located at the cloud base altitude instead of the entire vertical column of air. This cannot be achieved with passive remote sensing and will require vertically resolved extinction measurements. One way to achieve this is a combination of CALIPSO-type backscatter lidar (with aerosol backscatter (β) and depolarization ratio (δ) at 1064 and 532 nm with measurements from polarimeter or radar-SODA (Synergized Optical Depth of Aerosols) [*Dawson et al*., 2015] method to get extinction. Due to the potentially large (and unknown) difference between CALIPSO-measured attenuated backscatter and true backscatter signal, errors in the aerosol backscatter can accumulate as the signal penetrates through the column when using CALIPSO-type elastic (e.g., 2β+1δ) lidar. A better way to achieve vertically resolved extinction measurements with improved accuracy is with HSRL.
2. Separately for different aerosol types, because cloud droplet activation for a given size of aerosol depends on its hygroscopicity and therefore can be very different for common aerosol types over the ocean, i.e., mineral dust, urban pollution, and sea-salt. Achieving qualitative information on vertical distribution of aerosol type will require HSRL-type lidar with aerosol backscatter and depolarization ratio at 1064 and 532 nm and aerosol extinction (α) at 532 nm (e.g., 2β+1α+2δ).
3. Separately for anthropogenic and natural aerosols so anthropogenic aerosol effects on clouds can be disentangled from the background "preindustrial" conditions. Such information can be obtained only for very limited cases using passive remote sensing and will require HSRL-type lidar with 2β+1α+2δ for quantitative vertical distribution of aerosol type.
4. With uncertainty within a factor of 2 of the measured values, to constrain poorly characterized CCN number over the ocean and reduce the uncertainty in model-predicted annual mean indirect radiative forcing below 1.5 W m^{-2}. The ACE satellite mission aims to produce a comprehensive set of vertically and horizontally resolved aerosol number concentration, effective variance, and effective radius over the 0.1–1-μm radius range under humidified ambient conditions with uncertainties of 100%, 50%, and 10%, respectively [*Fridlind and Ackerman,* 2011], by including a lidar instrument with HSRL capability at both 355 nm and 532 nm, plus an elastic backscatter channel at 1064 nm (i.e., 3β+2α+3δ). Uncertainties in CCN retrievals by such HSRL correspond to a low limit of 0.7 W m^{-2} in global equivalent

uncertainty in diurnal shortwave indirect forcing for marine clouds [*Fridlind and Ackerman,* 2011], which is lower than the current value of 0.5 to 1.2 W m^{-2} [*Carslaw et al.*, 2013]. The addition of the 355 nm wavelength adds sensitivities to aerosols at sizes smaller than ones allowed by 532 and 1064 nm wavelengths alone. As aerosol number over the 0.1–1-μm radius range can be adopted as a proxy for CCN in marine clouds, this example can be used as a quantitative illustration that proposed aerosol property retrievals for the ACE mission could provide CCN number within a factor of 2 of observed values and help climate models to constrain the global indirect radiative forcing.

Studies are also in progress to derive chemical composition for predicted aerosol types such as smoke, fresh smoke, urban, polluted maritime, maritime, dusty mix and pure dust as derived by *Burton et al*. [2012] that will help to constrain changes in the aerosol scattering coefficient with relative humidity.

In addition to aerosols, global-characterization of other geophysical variables is also needed:

A. Ocean sub-surface vertical structure and community composition in phytoplankton biomass and particulate carbon, because ocean-derived aerosols with diameters between 0.05 to 0.15 μm are thought to be primarily organic in nature. Such information can also help in constraining the contribution of secondary organic aerosol (through phytoplankton-derived trace gases) to the CCN budget over the oceans. Passive remote sensing can only provide ocean surface *chlorophyll a* concentration without information on vertical structure. Such information will require a HSRL-type lidar with ocean-profiling capabilities.

B. Concurrent retrievals of aerosol and cloud optical properties with simultaneous measurements of drizzle and precipitation rates. Obtaining such information will require advanced sensors such as HSRL and Duel-frequency Doppler Radar.

4. The Likelihood Of Affordably Achieving The Required Measurement(s) In The Decadal Timeframe

Table 2 below provides a rating for five measurement characteristics similar to those used in the 2015 report prepared by the National Academies of Sciences, Engineering, and Medicine [*National Academies of Sciences*, 2015] for different geophysical measurements and instrument types. Discussion for the scientific importance of the quantified objective (I), the utility of a geophysical variable record for achieving a quantified objective (U), the quality of a measurement for providing the desired geophysical variable record (Q), the success probability of achieving the measurement and its associated geophysical variable (S), and the affordability of providing the measurement and its geophysical variable record (A) is given below. The analytical values of each parameter were prescribed based on the best estimate as discussed below and the benefit (B) and the final value rating (V) were calculated using the following formulas:

$$B = I \times U \times Q \times S$$

$$V = B \times A = I \times U \times Q \times S \times A$$

More detailed discussion for each of the parameters can be found in other white papers, i.e., *Mace et al.* [2016], *Ferrare et al*. [2016], *Hostetler et al*. [2016], and *Behrenfeld and Meskhidze* [2016] submitted to this call.

Scientific importance

Having a lidar in space is critical for resolving CCN number concentration at the cloud base altitude and at horizontal distances from the clouds that are much less than the aerosol decorrelation scale.

Different instruments summarized in Table 2 can provide satellite based CCN concentration estimates. However, only 3β+2α+3δ HSRL instrument will be able to retrieve CCN number within a factor of 2 of the observed values for the majority of the globe. This uncertainty in CCN would roughly correspond to 0.7 W m^{-2} in global equivalent uncertainty in diurnal shortwave indirect forcing for marine clouds, and is expected to lead to improved assessments of human-induced climate change. With current satellite instruments, uncertainty in CCN estimates cannot be reduced below a factor of 2 and will often remain as high as a factor of 10 for some specific regions and/or aerosol types [*Shinozuka et al*., 2015; *Stier*, 2015].

The utility of a geophysical variable

The ACE mission in its pre-formulation phase has made significant progress regarding proposed science objectives and instrument requirements. Table 2 shows the increasing usefulness of a spaceborne lidar for addressing the QESO. A simple elastic backscatter lidar is useful only in that it is able to differentiate aerosols at the surface and cloud base from those in the free troposphere, with the former being most relevant for CCN. The HSRL is required to measure (rather than retrieve) aerosol extinction, which can crudely be related to CCN number through empirical correlations [e.g., *Shinozuka et al*., 2015]. The high information content measured by the most capable HSRL type (i.e., 3β+2α+3δ) sensor enables application of advanced aerosol microphysical property retrievals suitable for meeting the stated QESO by directly retrieving aerosol number, size distribution and refractive index [*Sawamura et al*., 2016]. Since CCN depend on aerosol size and chemical composition, this advanced 3+2 retrieval provides the information needed to satisfy the QESO. Finally, the greatest utility (and information content) is achieved through the combination of the advance HSRL with a hyperspectral polarimeter.

The quality of a measurement

The quality of each geophysical measurement increases in tandem with its utility in addressing the QESO. The extensive series of cross-cutting applications have common requirements for global vertical retrievals of CCN number over the ocean. While several instruments are shown in Table 2, the primary driver is the tradeoff between complexity of the instruments and retrieval uncertainty. Despite a slight increase in complexity, the HSRL represents a significant and important advance over a simple CALIPSO-type lidar. Signal to noise in both channels will be much higher vertical and horizontal resolution will be better allowing for profiling aerosols in partially cloud scenes right up to the edge of marine stratocumulus. Simply put, the higher measurement quality of an HSRL system is fundamental to addressing this QESO!

Success Probability

The lidar and polarimeter technology that comprises ACE's core measurement suite is expected to continue advancing to a technological readiness level that will permit this ACE component to go in full formulation phase by the time the 2017 Decadal Survey Report is published (ACE Science Study Team, 2016.)

Affordability

Years of technology development, airborne instrument development, and algorithm development have put lidars of this type on a path to affordable implementation within a timely schedule. A team of engineers and instrument scientists at NASA Langley developed a detailed instrument concept in January 2016. The cost of that instrument has been estimated using the SEER parametric cost estimation tool and a bottom up approach. Those cost estimates, and the information on which they are based, can be supplied to the NRC Decadal Survey Panel upon request.

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Table 1. Global annual mean forcing uncertainty due to specific parameters

CCN Type	Uncertainty ($W\ m^{-2}$)*	Reference
Sea-salt	0.76 (-1.34 to -2.1)	*Ma et al*. [2008]; *Carslaw et al*. [2013]
Primary and secondary marine organics	0.09 (-0.009 to - 0.15)	*Meskhidze et al*. [2011]
Volcanic SO_2	0.2 (−0.86 to −1.16)	*Schmidt et al*. [2012]; *Carslaw et al*. [2013]
DMS emissions	0.15 (+0.02 to -0.16)	*Boucher et al*. [2003]; *Carslaw et al*. [2013]
Biogenic secondary organic aerosol	0.03 (-1.12 to -1.15)	*Carslaw et al*. [2013]
Biomass burning	0.02 (-1.13 to -1.15)	*Carslaw et al*. [2013]
Anthropogenic SO_2	0.17 (-1.05 to -1.22)	*Carslaw et al*. [2013]
Anthropogenic secondary organic aerosol	0.07 (-1.11 to -1.15)	*Carslaw et al*. [2013]
Total over the ocean	-0.35 to -1.8	*Hoose et al*. [2009]

*Values in parentheses show 9% – 91% confidence interval

Table 2. Summary of Subjective Method Ratings

Geophysical Measurement	Instrument Type	Microphysical Retrieval Type	Importance (I)[1]	Utility (U)[2]	Quality (Q)[3]	Success Probability (S)[4]	Synergistic Multiplier (M)[5]	Total Benefit B = IxUxQxSxM	Affordability (A)[6]	Total Value (BxA)	Adjectival Value Score
Aerosol backscatter (β) and depolarization ratio (δ) at 1064 and 532 nm	CALIPSO-type backscatter lidar (but with higher SNR)	$2\beta+2\delta$	5	0.4	0.4	0.9	1.0	0.7	5	**3.6**	**Fair**
The above + aerosol extinction (α) at 532 nm	HSRL-type lidar	$2\beta+1\alpha+2\delta$	5	0.8	0.7	0.8	1.0	2.2	5	**11.2**	**Good**
The above + aerosol backscatter (β), depolarization ratio (δ), and extinction (α) at 355 nm	More Capable HSRL	$3\beta+2\alpha+3\delta$	5	0.9	0.9	0.8	1.0	3.2	5	**16.2**	**Very Good**
The above + hyperspectral polarimetric radiances	More Capable HSRL + PACE Polarimeter	Advanced Active-Passive Retrieval	5	1	1	0.8	1.2	4.8	5	**24.0**	**Excellent**

[1] Having a lidar in space is critical for resolving the vertical distribution of CCN.

[2] Utility increases with increasing information content.

[3] Placing a premium on the increased accuracy/precision of the HSRL, and even further on the 3+2 microphysical retrieval.

[4] CALIPSO-type lidar is space-proven; both HSRLs are traceable to extensive airborne characterization/laboratory risk-reduction studies; EarthCare will provide important HSRL performance information. Individual instrument retrieval work is on-going and proven. The combined active-passive (lidar+polarimeter) retrieval is funded and under development, but has yet to been demonstrated.

[5] Synergistic multiplier accounts for increased accuracy/precision that are expected to be associated with the combined active-passive retrieval. The swath of the PACE imager/polarimeter combined with the vertical component from the lidar enables a 3-dimensional view of the atmospheric state that is much more than just the sum of its parts (this latter increase in "sum of parts" is reflected in the increased U and Q scores).

[6] Years of technology development, airborne instrument development, and algorithm development have put lidars and polarimeters of this type on a path to affordable implementation within a reasonable schedule. International collaboration/leveraging on lidar is likely.

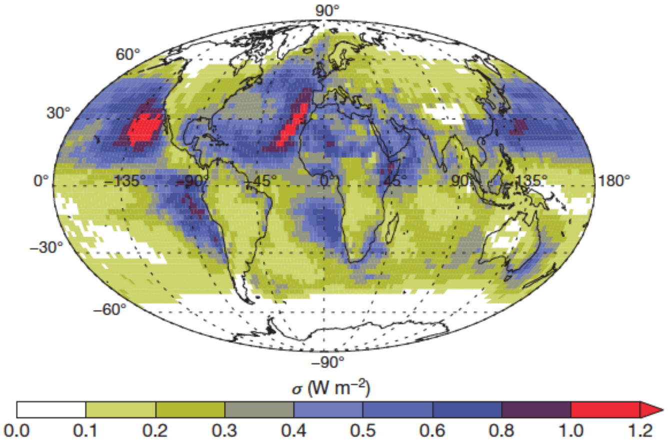

Figure 1. Standard deviation of the global annual mean aerosol first indirect forcing. Figure adapted from *Carslaw et al.* [2014].

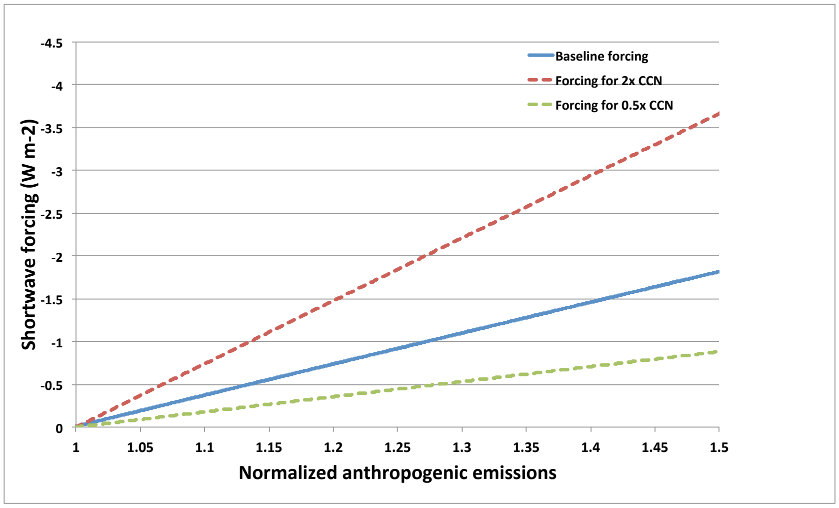

Figure 2. Calculated effect of the uncertainty in derived CCN number over the ocean on shortwave radiative forcing. The solid line shows the calculated forcing, while dotted lines show forcing associated with a factor of 2 uncertainty in CCN. The initial CCN concentration is 50 cm^{-3}, rising to a maximum of 80 cm^{-3}. The cloud droplet number concentration is calculated as $CDNC = 375\times(1 - e^{0.0025\times CCN})$. The albedo of the baseline cloud is assumed to be 0.5 and the albedo versus CDNC is $dA/d\ln(CDNC) = A\,(1 - A)/3A$ [*Twomey*, 1991]. $\Delta F = -dA/d\ln(CDNC) \times F_T/4\times T_a^2\times A_m\times\Delta\ln\,(CDNC)$ where F_T is the solar constant (assumed to be 1368 Wm^{-2}), Ta is the transmission of the atmosphere (assumed to be 0.75), and A_m is the fraction of the earth covered by maritime clouds (assumed to be 0.3).