Executive Summary

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This report, focusing on the influences of atmospheric aerosols on climate and climate change, is part of the 5-year assessment review of activities and progress of research, conducted by the Climate Change Science Program (CCSP) and mandated by the National Research Council (NRC).

Atmospheric aerosols are a suspension of solid and/or liquid particles in the air, which are ubiquitous and are often observable as dust, smoke, and haze. Both natural and human processes contribute to atmospheric aerosols. On a global basis aerosol mass derives predominantly from natural sources (e.g., sea-salt, and dust). Manmade aerosols, arising mainly from a variety of combustion sources (e.g., "smog"), usually overwhelm the natural ones in areas in and downwind of highly populated and industrialized regions, or areas of intense agricultural burning.

Aerosols affect the Earth's energy budget by scattering and absorbing radiation (direct effect) and by modifying the cloud amount, lifetime, and microphysical and radiative properties (indirect effects). Moreover, the direct absorption of radiant energy by aerosols leads to heating of the troposphere and cooling of the surface, which can change the relative humidity and atmospheric stability thereby influencing the clouds and precipitation (semi-direct effect). The addition of manmade aerosols to the atmosphere may change the radiative fluxes at the top-of-atmosphere (TOA), at the surface, and within the atmospheric column. Such a perturbation of radiative fluxes by anthropogenic aerosols is designated as *aerosol climate forcing*, which is distinguished from the *aerosol radiative effect* of the total aerosol (natural plus anthropogenic). The aerosol climate forcing and radiative effect are characterized by large spatial and temporal heterogeneities due to the wide variety of aerosol sources, the spatial non-uniformity and intermittency of these sources, the short atmospheric lifetime of aerosols (relative to that of the gases), and processing (chemical and microphysical) that occurs in the atmosphere.

On a global average the sum of direct and indirect forcing by anthropogenic aerosols at the TOA is almost certainly negative (cooling) and thus likely offsets the positive forcing (warming) due to anthropogenic greenhouse gases on a global-average basis, and taking into account the sum of longwave and shortwave forcings. However, as the forcings are exerted in different spectral regions and exhibit different magnitudes in different locations the offset cannot be considered to be neutral in terms of effects on Earth's climate.

The Intergovernmental Panel on Climate Change Fourth Assessment Report (IPCC AR4) reported on the results of some 20 participating global climate models. These models can reproduce the observed trend in global mean temperature over the twentieth century due to changes in atmospheric concentrations of greenhouse gases and other forcing agents including aerosols. When anthropogenic aerosol forcings are not included, the models tend to generate too much warming. However the ability of

climate models to reproduce the global mean temperature change over the past 100 years appears to be the result of using a "tuned" aerosol forcing. Although different models exhibit a wide range of climate sensitivity (i.e., the amount of temperature increase due to the increase of CO_2), they yield global temperature change, which is similar to the observed change. Apparently this is because the forcing by aerosols differs between models. For example, the direct cooling effect of sulfate aerosol varies by a factor of 6 among the models, because of different extensive aerosol properties (e.g. sulfate amount) and different intensive properties (e.g. scattering efficiency) used in the models. Greater disparity is found in the model treatment of other aerosol types such as black carbon and organic carbon. Even the choice of which aerosol types and which aerosol forcings are treated in a particular model varies. Some models include only the direct aerosol effect, whereas others include an indirect effect in which the aerosols modify cloud microphysics and hence cloud brightness. In addition, the aerosol indirect effect on cloud brightness varies by up to a factor of 9 among models. This situation is in part a consequence of the large uncertainty in the mechanisms and magnitude of climate forcing by aerosols, and in part due to the differences in cloud amounts between models.

Over the past decade there has been substantial improvement in measurement of the amount, geographical distribution, and physical and chemical properties of atmospheric aerosols, the controlling processes, and the direct and indirect radiative effects of these aerosols. Key research activities have been:

- Execution of intensive field experiments examining aerosol processes and properties in various aerosol regimes around the globe;
- Establishment and enhancement of ground-based networks measuring aerosol properties and radiative effects;
- Development and deployment of new and enhanced instrumentation, importantly aerosol mass spectrometers examining size dependent composition, and;
- Development and implementation of new and enhanced satellite-borne sensors examining aerosol effects on atmospheric radiation.

These efforts are beginning to provide the needed inputs and constraints necessary to improve the accuracy of representation of aerosol effects in climate models. In addition they are allowing for a shift in estimates of aerosol radiative effect and climate forcing from largely model-based to an increasing level of measurement-model synthesis. The new observational capabilities together with dedicated aerosol modeling efforts have led to a better understanding of the aerosol system, and to smaller uncertainties in estimates of aerosol direct radiative effect and climate forcings. The resulting improved understanding and quantification of these aerosol effects have contributed to the finding in the 2007 assessment report of the Intergovernmental Panel on Climate Change that human activities "very likely" have been contributing to the global change that has been observed over the last 100 years.

In short, direct aerosol climate forcing is understood now much better than 10 years ago. However the tools to reduce uncertainties are still urgently needed. The improvement in measurement-based systems is necessary to identify remaining outstanding issues and improve quantification of aerosol effects on climate. Improvement in modeling is necessary to confidently extend estimates of forcing to prior times and to project future emissions. Achieving these capabilities will require a synergistic approach between observational systems and modeling.

1. Measurement of aerosol properties and their evolution

1.1 *In situ and surface remote sensing measurements.*

Over the past two decades, more than a dozen large-scale intensive field experiments have been conducted to study the physical, chemical, and optical properties of aerosol, and the processes that govern aerosol amounts, types and the effects aerosols have on clouds in a variety of aerosol regimes around the world. Ground-based networks including both comprehensive sites with in-situ and remote sensing instrumentation (e.g., NOAA Global Monitoring Division - GMD sites) and simpler ground-based remote sensing networks (e.g., the NASA Aerosol Robotic network - AERONET) provide the long-term context for intensive field campaigns and ground-truth for satellite validation. Widespread application of aerosol mass spectrometers has shown the large contribution of organic substances to total aerosol mass concentration over much of the Northern Hemisphere and the increasing preponderance of secondary organic aerosols with increasing distance from urban source regions. Laboratory studies of aerosol formation and evolution processes provide the fundamental basis for representing aerosols in models.

1.2 *Satellite measurements.*

A measurement-based characterization of aerosols on regional to global scales can be realized only through satellite remote sensing, because of the large spatial and temporal heterogeneities of aerosol distributions. Over the past decade, satellite aerosol retrievals have become increasingly sophisticated. From these observations, retrieved aerosol products include spectral optical depth, particle shape and effective particle size over both ocean and land, as well as more direct measurements of polarization and phase function. Cloud screening is much more robust than before, and onboard calibration is now widely available. Active remote sensing is also making promising progress by collecting essential information about aerosol vertical distributions. These measurements are essential to evaluate the performance of aerosol transport and transformation models used to determine aerosol forcing.

1.3 *Synergy.*

The best strategy for characterizing aerosols has been to integrate measurements from different satellite sensors with airborne and ground-based measurements and models. While we have made much progress toward measurement-based estimates of aerosol effects and forcing, models still provide information lacking in the observations, such as aerosol composition, characteristics, and four-dimensional (4-D) distributions over wide regional areas in situations difficult to observe (i.e., cloudy scenes, complex surface types, different times of day) by satellite or other platforms. Models also provide critical links among different observations, and can simulate the past and project into the future.

1.4 *Looking Forward*

- Long term networks such as the NOAA GMD sites, AERONET and emerging ground-based lidar networks provide essential information on decadal and mulit-decadal trends and on aerosol properties that are vital for satellite validation, model evaluation, and climate change assessment. Progress in aerosol-climate science is dependent on the continuity of these networks.

- Enhancing long term stations to include accurate measurements of spectral aerosol absorption would add an essential characteristic needed for estimates of aerosol forcings, both direct and indirect.
- The modern satellite sensors have a relatively short life time. Global long-term aerosol trends cannot be detected without equivalently-calibrated replacement sensors that at least match current capabilities.
- Current satellite capabilities, while good, still lack accurate information on aerosol size distribution, absorption, type and extinction above clouds. New sensors with enhanced capabilities and different satellite orbit configurations including geostationary and Lagrange points will better constrain models and narrow uncertainties.
- Outstanding questions on aerosol scattering and absorption across the solar spectrum, and on aerosol roles in cloud nucleation will only be answered by development of new measurement techniques applied in the laboratory and during field campaigns.

2. Aerosol representations in models

Aerosol simulations in chemical transport models and climate models have been extensively evaluated using observations. However, comparison of model parameters with observations is problematic because of strong disagreements among the different satellite instruments, between satellite and ground-based sensors, and between remote-sensing and in-situ measurements.

2.1 *General evaluation of aerosol representation in models.*

Differences in simulated aerosol composition between climate models are large. These differences affect calculated aerosol properties and radiative effects. In addition, aerosol models tend to underestimate total aerosol optical depth (AOD) compared with observations, but they are doing better currently than they were a few years ago. The Aerosol Comparisons between Observations and Models (AEROCOM) project report on a set of models that produced global aerosol optical thicknesses at 0.55 μm ranging between 0.11 and 0.15. In comparison the average AOD obtained at a large number of sites comprising the AERONET network is 0.135, and the satellite composite is 0.15. However, even though the models' global mean AOD values are converging, the convergence is obtained despite large differences in aerosol composition, particle size and atmospheric residence times. This implies improvements in model representation of AOD will not translate directly into model improvement of calculated forcing, which depends on AOD as well as other aerosol parameters. An additional concern is that aerosol forcing tends to exhibit large spatial and temporal variability so that global-mean comparisons may not be meaningful.

2.2 *Looking forward*

- Progress requires effort on the observational side to reduce uncertainties and disagreements between different observational data sets.
- Agreement in aerosol optical depth among models and between models and observations can mask large differences that exist in modeled aerosol types and representations of aerosol processes. Comparison of additional aerosol properties (e.g., aerosol chemical composition, spectral variation of optical depth, size parameter, and absorption properties) is expected to

yield information on model differences and begin to constrain representations of the several aerosol component species.

- Better estimates of source strength and location of both primary aerosol particles and precursor gases are needed. Improvements can be achieved by observational techniques including satellite measurements of emissions from biomass burning and improved inversion techniques. However developing emission inventories of primary particles and of aerosol precursor gases is ultimately based on measurements of emission factors for pertinent processes together with detailed inventorying of the intensity of the pertinent activities as a function of location and time.
- Wet/dry removal and transport processes require enhanced understanding by field measurements together with further model evaluation and development. Continued satellite and in situ measurements are needed for model evaluation.

3. Aerosol direct radiative effect and climate forcing

3.1 *Cloud-free aerosol direct radiative effect (DRE).*

Aerosol direct radiative effect denotes the change in radiative flux at top of the atmosphere or at the Earth's surface due to the presence of aerosols. Present measurement capabilities permit determination of the global annual average cloud-free aerosol DRE for solar radiation over oceans as -5.5 ± 0.2 W m^{-2} net flux at the top of the atmosphere (TOA) and -8.8 ± 0.7 W m^{-2} net flux at the surface. Deriving the aerosol direct effect over land from flux measurements such as from CERES is complicated by a large and highly heterogeneous surface reflection. A hybrid of satellite retrievals and model simulations yields a global (land and ocean) cloud-free DRE of solar radiation of -4.9 ± 0.7 W m^{-2} and -11.8±1.9 W m^{-2} at the TOA and surface, respectively. Model simulations result in estimates that are 30-50% weaker (less cooling) than the measurement-based estimates. The model-measurement differences relate to model underestimates of AOD over oceans and tropical land, where dark surfaces allow aerosols to produce big differences in radiative effects.

3.2 *Cloud-free aerosol direct climate forcing (DCF).*

DCF denotes only the effects of *anthropogenic* aerosols. The measurement-based estimate of cloud-free TOA DCF by anthropogenic aerosols ranges from -1.1 to -1.6 W m^{-2} over ocean, stronger than model simulated values of -0.3 to -0.9 W m^{-2}. Including the less certain land component, on global average, the measurement-based estimate of TOA DCF ranges from -1.1 to -1.9 W m^{-2}, again stronger than the model-based estimates of -0.4 to -1.0 W m^{-2}. The range in model estimates is due to having different aerosol components plus different properties for the particular aerosol type. Overall, satellite-based estimates of DCF exhibit much greater uncertainties than estimates of DRE.

3.3 *Total sky aerosol direct radiative effect and climate forcing.*

Aerosols staying above or below clouds can scatter and absorb solar radiation, creating an aerosol effect on the radiation field or a direct climate forcing if the aerosols are anthropogenic. This not an indirect effect when aerosols modify cloud properties. The total sky aerosol direct radiative effect and climate forcing is a sum of cloud-free and cloudy DRE/DCF weighted respectively by cloud-free fraction and

cloud fraction. However the aerosol and its direct radiative effect and climate forcing are difficult to observe in the presence of clouds. As a result, it is only model estimates of the total sky forcing that are generally reported. The total sky aerosol direct climate forcing at the top of the atmosphere shows a wide disparity among models, varying from slight warming (+0.04 W m^{-2}) to cooling (–0.63 W m^{-2}). This is primarily the result of models having different aerosol components plus different properties for the particular aerosol type and relative heights of aerosol and cloud layers.

3.4 *Looking Forward*

- Uncertainties in measurement-based estimates over land can be reduced with more accurate measurements of aerosol absorption and improved understanding of the reflectance from heterogeneous land surfaces.
- Active remote sensing observations can constrain relative heights of aerosol layers and cloud layers in order to estimate the total sky DRE and DCF from measurements. These observations can also constrain the vertical distributions of aerosol absorption in the atmosphere that are essential to understanding how the atmosphere will respond to the aerosol radiative effect.
- Improvements in measurement-based estimates of DCF depend on developing techniques to better determine the anthropogenic component of the aerosol in the measurements. For example, using in situ measurements of composition and optical properties to constrain satellite techniques have not yet been fully explored.
- Ultimately the improvements in measurement-based estimates can be expected to better constrain model results and narrow the range of model estimates of both DRE and DCF.

4. Aerosol indirect effects

4.1 *Observations of aerosol indirect effects*.

Remote sensing estimates of aerosol indirect forcing are still very uncertain. Current estimates of global average aerosol indirect forcing based on remote observations range from -0.6 to -1.7 W m^{-2}. Few observational systems measure cloud liquid water path, even though this quantity is necessary to quantify aerosol indirect forcing. Basic processes still need to be understood on regional and global scales. Uncertainties will likely increase before they decrease as new processes and their feedbacks become known. Remote sensing observations of aerosol-cloud interactions and aerosol indirect forcing are based on simple correlations between variables, from which cause-and-effect are inferred. However, such inferences are not proven. The most difficult aspect of inferring aerosol effects on clouds from the observed relationships is separating aerosol effects from meteorological effects when aerosol loading itself is often correlated with the meteorology. As with the case of direct forcing, the regional nature of indirect forcing is especially important.

4.2 *Representations of aerosol indirect effects in General Circulation Models (GCMs)*.

Most GCMs do not address aerosol indirect effects. Approximately only one-third of the models used for the IPCC 20th century climate change simulations incorporated an aerosol indirect effect. Generally, though not exclusively, this effect was associated with sulfate-only aerosol. The results varied strongly among models. The IPCC estimate of the net radiative forcing associated with the cloud

albedo effect given in the Chapter 2 Executive Summary of the IPCC AR4 ranges from +0.4 to –1.1 W m^{-2}, with a 'best-guess' estimate of –0.7 W m^{-2}. Most models did not incorporate forcing associated with aerosol effects on cloud fraction and amount of condensate. Comparison of the indirect effect in various models showed that differences in cloud dynamics and microphysics play a strong role in inducing differences in the indirect effect(s).

4.3 *Coupling GCMs with cloud resolving models.*

A primary difficulty in representing aerosol cloud interactions in GCMs is that GCMs do not resolve convection on their large grids (order several hundred km), and that their treatment of cloud microphysics is rather crude, as is their representation of aerosols. Until GCMs are able to represent cloud scales, it is questionable what can be obtained by adding microphysical complexity to poorly resolved clouds. Superparametrization efforts (where standard cloud parameterizations in the GCM are replaced by resolving clouds in each grid column of the GCM via a cloud resolving model) could lead the way for the development of more realistic cloud fields. However these are just being incorporated in models that resolve both cloud and aerosols. Global cloud resolving models, with grid sizes on the order of 4 km are in their infancy but represent another possible path forward. They are, however, restricted to shorter integration times. The coupling of aerosol and cloud modules to dynamical models that resolve the large turbulent eddies associated with vertical motion and clouds (large eddy simulations or LES, with grid sizes of ~ 100 m and domains ~ 10 km) has proven to be a powerful tool for representing the details of aerosol-cloud interactions together with feedbacks. Such models, together with observations at similar scales, enable improved understanding of aerosol-cloud processes, and represent a foundation for work at larger scales.

4.4 *Looking forward:*

- All progress in estimating aerosol indirect effects requires a better understanding of the basic processes of aerosol-cloud interaction.
- A methodology for integrating observations (ground-based, airborne and satellite) and models at the range of relevant temporal/spatial scales is crucial, as is separating meteorological effects from aerosol effects on clouds.
- Coupling GCMs with cloud resolving models is the long term solution, but a better overall understanding of the processes must be achieved first.

5. Long term trends of aerosol and radiation

5.1 *Multi-decadal change of solar radiation reaching the surface.*

Systematic changes in observed global solar radiation (a sum of direct and diffuse solar radiation) reaching the surface (so-called dimming or brightening) have been reported in the literature. Speculation suggests that such trends result from multi-decadal changes of aerosol emissions. However, the lack of reliable long-term observations of aerosol trends over both land and ocean during this time period makes it difficult to assess the role aerosols have played in the multi-decadal change of surface solar radiation. Therefore, long-term global aerosol observation in conjunction with high-quality surface radiation measurements is strategically necessary. In addition to the aerosol optical depth,

changes in aerosol composition due to changes in industrial practices, environmental regulations, and biomass burning emissions are required. There are also important ramifications for cloud formation since radiative balance requires that reduced incoming surface radiation results in reduced surface sensible and latent heat fluxes, and therefore weaker convection.

5.2 *Dynamic quality of the aerosol system.*

The global aerosol system is a moving target. Ten years ago the spatial distribution of aerosol production was different than it is today, and ten years from now, it will be different again. This is unlike the observations of the relatively constant increase in greenhouse gases.

5.3 *Looking Forward*:

- Aerosol trend analysis requires long-term consistent data records. Practically this will require multiple sensing systems over decades and careful testing the consistency of the measurements as the record passes from one sensing system to the next. An improved understanding of aerosol indirect and semi-direct effects is particularly needed to make a robust attribution of the observed dimming or brightening to the aerosol changes.
- Simulating long-term aerosol variations with global models can link long-term trends of emissions, aerosol loading, and radiative effects.

6. Climate response to aerosol forcing

6.1 *Aerosol forcing compared with greenhouse gas forcing.*

Although the nature and geographical distribution of forcings by greenhouse gases and aerosols are quite different, it is often assumed that to first approximation the effects of these forcings on global mean surface temperature are additive, and thus that the negative forcing by anthropogenic aerosols has offset the positive forcing by incremental greenhouse gases over the industrial period. The IPCC AR4 estimates the total global average TOA forcing by incremental greenhouse gases (long-lived GHGs, tropospheric and stratospheric ozone, and stratospheric water vapor from methane oxidation) to be 3.0 (2.8 to 3.4) W m^{-2}, where the range in parenthesis is meant to encompass the 5-95% probability that the actual value will be within the indicated range. The corresponding value for aerosol forcing (direct plus enhanced cloud albedo effects only) is –0.6 (0 to -1.8) W m^{-2}. The total forcing, 1.6 (0.6 to 2.4), reflects the offset of GHG forcing by aerosols; the uncertainty in the total forcing is dominated by the uncertainty in aerosol forcing.

6.2 *Implications of aerosol forcing and its uncertainty on GCM calculations of temperature change over the industrial period.*

A key requirement for forcing over the industrial period is as input to GCM calculations, necessary to evaluate the performance of these models in calculating climate change over the industrial period by comparison with observations. In such a comparison Hansen et al. (2007) explicitly took note of the wide range of possible forcings resulting from uncertainty in aerosol forcing and the implications on determining climate model sensitivity, acknowledging that "an equally good match to observations probably could be obtained from a model with larger sensitivity and smaller net forcing, or a model

with smaller sensitivity and larger forcing". This balance between the magnitude of the forcing and the sensitivity of the model thus makes it difficult to determine climate sensitivity. Studies suggest that the climate responses to greenhouse gases and sulfate aerosols are correlated, and separation is possible only occasionally, especially at global scales and during summer when the aerosol effect on solar absorption in the Earth-atmosphere system is likely to be bigger. The conclusions concerning this appear to be model and method-dependent: using time-space distinctions as opposed to trend detection may work differently in different models.

Even distinguishing between the effect of different aerosol types is difficult. Overall, the similarity in response to all these very different forcings is undoubtedly due to the importance of climate feedbacks in amplifying the forcing, regardless of its nature.

6.3 *Absorbing versus reflective aerosol forcing.*

Distinctions in the climate response do appear to arise in the vertical, and these could be used to help quantify aerosol forcing. Absorbing aerosols produce warming that is exhibited throughout the troposphere and into the stratosphere, whereas reflective aerosols cool the troposphere but warm the stratosphere. In the ocean the cooling effect of aerosols extends to greater depths than the warming due to greenhouse gas increases, because of the thermal instability associated with cooling the ocean surface. Hence the temperature response at levels both above and below the surface may provide an additional constraint on the magnitudes of each of these forcings.

6.4 *Looking forward:*

The response of climate models to aerosol forcing is dependent on aerosol forcing in the models. Continued progress in evaluation of climate models will depend on improved estimates of aerosol forcing. There is potential for further improvement by examination of the relative forcing due to absorbing and reflective properties of different aerosols.

7. Aerosol interaction with precipitation and weather

The most important improvement of understanding of aerosols and their role in global change is that the aerosol effect extends much beyond radiative forcing in the Earth's energy budget. Aerosols absorb sunlight and create differential heating within the atmosphere that affects atmospheric circulations and weather at many scales. Aerosol affects clouds by modifying cloud brightness, cloud cover, precipitation and severe weather. Clouds and precipitation, in turn, affect aerosol.

7.1 *Looking forward:*

- Aerosols, clouds, precipitation, weather and climate must be studied as a holistic system, with both observations and models, and with an emphasis on long term monitoring of a system that exhibits a strong degree of regional variability and is continually changing.
- Improved measurement capabilities coupled with more sophisticated treatment of aerosols and clouds in models suggest that more accurate and constrained aerosol forcings are expected to be available for future intercomparisons of general circulation models.