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Aerosol Particles and Their Role in the Atmospheric Radiation Balance

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Abstract: *Aerosol particles in the atmosphere have a big impact on the Earth's energy budget, because they can scatter and absorb solar energy (solar radiation) and heat energy from the Earth (terrestrial radiation). Also, they affect the climate indirectly by their interaction with clouds. Both of these factors lead to uncertainty about how much the aerosol particles contribute to human-caused climate change. Research on this topic has been going on for decades. To create a summary of what we currently know about aerosol optical properties, direct and indirect radiative effects due to aerosol particle characteristics, primary sources of aerosols, and regional variations across the globe. Overall, this review shows aerosol particles contribute to a cooling effect (aero-refraction) from the total radiative forcing of aerosol particles (against the greenhouse gases they have). This cooling effect cancels out a large portion of the greenhouse gases, although we still do not know very much about the indirect effect of clouds on aerosol particles (Boucher et al 2013).*

Keywords: aerosols, radiative forcing, radiation balance, direct effect, indirect effect, climate forcing, optical properties

I. INTRODUCTION

Aerosols are made up of solid or liquid particles that are suspended in the atmosphere, with particle sizes that generally range from a few nanometers to several micrometers in diameter. The sources of the different types of aerosols come from both nature (e.g., sea salt, mineral dusts, volcanic activity and biogenic materials) and human activity (e.g., fossil fuel combustion, biomass burning and industrial activity (Charlson et al., 1992). Aerosols will influence the climate through a number of ways that interact with the atmospheric radiation field, making aerosols fundamental to understanding the current and future state of the Earth's energy budget (Kaufman et al., 2002).

The Intergovernmental Panel on Climate Change (IPCC) has continuously ranked aerosol radiative forcing as the largest source of uncertainty in the predictions of human-caused climate change, with the fifth assessment report estimating total aerosol radiative forcing to be approximately -0.9 W m^{-2}, with a range of uncertainty from -1.9 to -0.1 W m^{-2} (Myhre et al., 2013). The wide range of uncertainty is indicative of fundamental deficiencies in our understanding of the physicochemical properties of aerosols, their spatiotemporal variability, and their interactions with clouds (Forster et al., 2007).

This review provides an integrated overview of how aerosol particles interact with radiation, encompassing both the direct radiative effect (DRE) and indirect cloud-mediated effects.

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II. AEROSOL OPTICAL PROPERTIES

The optical characteristics of aerosol solids namely their aerosol optical depth (AOD), single-scattering albedo (SSA) and asymmetry parameter (g) determine how aerosol solids absorb and scatter electromagnetic radiation (EMR) emitted from any object. AOD represents the total extinction of light (EMR) in an atmospheric column at any given point, and is a dimensionless value. Values typically range from 0.3–1.0 over polluted urban areas, and <0.1 over clean ocean regions (Holben et al., 1998).

SSA indicates how scattering and absorption are divided between these two processes. For example, pure scattering (i.e., SSA = 1.0) aerosol types like sulfate and sea salt will serve to cool Earth's radiation by returning some of its shortwave radiation to outer space. In contrast, strongly absorbing (SSA ≈ 0.2–0.4) aerosol types, like black carbon, will serve to increase the temperature of the atmosphere through their absorption of solar radiation (Ramanathan & Carmichael, 2008). The overall impact of an aerosol group will differ depending upon the surface albedo of an aerosol, wherein the same absorbing aerosol will create positive radiative forcing over bright surfaces and negative radiative forcing over dark ocean surfaces (Haywood & Boucher, 2000).

The Ångström exponent (α) provides an assessment of AOD's spectral dependence based on which can classify the dominant particle size mode. High α (> 1.5) would indicate fine-mode accumulation (commonly formed through combustion or photochemical processes) while low α (< 0.5) would indicate coarse-mode (e.g., mineral dust and sea salt) particles (Dubovik et al. 2006). As a result, this spectral dependence is frequently reported in satellite retrievals and supported by ground-based AERONET (Holben et al. 1998) sun photometry networks.

III. DIRECT RADIATIVE EFFECT

The direct radiative effect (DRE) includes the composition and radiative forces due to aerosols that cause changes in radiative flux through both their scattering and absorption of radiation, independent of any cloud effects. At the top of the atmosphere (TOA), total global mean aerosol DRE (natural + anthropogenic) is estimated at between (−4.0 to −5.5 W m−²) due to the predominance of scattering aerosols in the current atmosphere. The anthropogenic portion (the direct radiative forcing (DRF)) is much smaller and is estimated to be (−0.35 W m−²) for the present day.

Black carbon (BC) aerosol presents an even more complicated case to assess accurately because, as the primary light-absorbing carbonaceous aerosol, it has a positive forcing that has been recently estimated at between (+0.4 to +0.9 W m−²) and therefore may be the second most important anthropogenic climate forcing agent following CO_2. However, the magnitude of BC forcing is sensitive to its mixing state, and when BC is internally mixed with other materials (i.e. organic or inorganic coatings) BC has enhanced radiative forcing by a factor of 1.5 - 2.5 times when compared to fresh BC through a process known as the "lensing effect". The observational support for this enhancement is still being debated, and field data has provided mixed results.

The global aerosol loading is greatly affected by mineral dust, which produces 1,000-2,000 Tg per year of total shadowing. Via both short and long wavelengths of radiation in the environment, dust produces an extensive amount of scattering and also absorbs energy. Based on thermal IR absorption and re-emission, coarse particles of dust can create positive forcing (Boucher et al., 2013). Depending upon surface albedo and dust mineral compositions and the distribution of particle sizes, dust forcing at the TOA can have positive or negative net forcing (Dubovik et al., 2006).

IV. INDIRECT RADIATIVE EFFECTS

Aerosols serve as cloud condensation nuclei (CCN) and ice nuclei (IN), modifying cloud characteristics and creating two indirect pathways of forcing. The first – the Twomey effect – explains that as the density of CCN increases, smaller and more numerous droplets (given any particular amount of liquid water present) will form, thus increasing the albedo of clouds. Secondly, the lifetime effect explains how smaller droplets (due to their size) would impede the formation of precipitation; therefore, clouds will last longer and increase cloud cover.

The IPCC's Assessment Report 5 (AR5) estimates the total aerosol-indirect warming value at −0.45 W m−²; however, there is a large (5–95% confidence range of −1.2 to 0.0 W m −²) amount of uncertainty present. Variations in aerosol-cloud interaction parameterizations among global climate model results contribute to the variability of indirect aerosol forcing spread associated with these models.

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Field campaign data, together with satellite-based studies, have produced statistically strong Twomey relationships associated with marine stratocumulus clouds, although finding evidence for the lifetime effect is found to be more difficult due to uncontrolled variations in meteorological parameters.

Additionally, absorbing aerosols affect the cloud formation process via semi-direct (SD) effects. Absorbing aerosols like BC, through heating of the atmospheric boundary layer, will provide stability to the lower troposphere and suppress convection, producing less cloud cover. Generally, semi-direct effects produce a smaller positive forcing effect, which partially offsets the negative effects produced by direct and first indirect effects. The magnitude of semi-direct forcing is highly sensitive to the vertical position of the absorbing aerosol layer relative to cloud tops (Hansen et al., 2005).

V. OBSERVATIONAL AND MODELING ADVANCES

The optical properties of aerosols have been mainly constrained through the use of ground-based networks. Remote sensing of aerosols from satellites has changed our knowledge of their distribution and trends globally. Ground-based networks have contributed significantly to the validation of satellite products and global atmospheric models. Global surface networks that measure aerosol chemistry also provide information about how the optical properties of aerosols are related to their chemical composition.

AERONET, a global network of sun-sky radiometers, provides AOD, SSA and size distribution retrievals with spectral resolution at over 400 permanent sites (Holben et al., 1998). Measurements from AERONET and similar networks around the world have provided valuable datasets for validating satellite aerosol products and global model output.

Satellite aerosol observations have revolutionized our understanding of where aerosols are located globally and how they change over time. The MODIS instrument provides daily global AOD retrievals, while the MISR instrument's multi-angle capability allows us to discriminate between various aerosol types (Kaufman et al., 2002). The CALIOP lidar on CALIPSO offers vertical profiles of aerosol extinction, showing the true three-dimensional shape of aerosol plumes that cannot be captured using column-integrated products. Together, these observations have identified distinct regional patterns such as persistent continental layers of absorbing aerosol in the Southeast Atlantic and transported Saharan dust across the tropical Atlantic (Boucher et al., 2013).

Global aerosol models have progressed significantly, evolving from basic bulk sulfate schemes to multi-component representations that account for BC, organic carbon, dust, sea salt, and nitrate aerosols (Schulz et al., 2006).

VI. REGIONAL VARIABILITY AND SOURCE ATTRIBUTION

Aerosol radiative impact is very different by location and time and is a reflection of where aerosols are picked up (source), where they travel to (transport path), and how they are lost in the atmosphere.

There is a significant contribution of Aerosol Optical Depth (AOD) over eastern and southern Asia from fossil fuel combustion, burning of biomass, and transport of dust (Ramanathan & Carmichael 2008). Before now, AOD measured from satellites showed decreasing AOD over North America and Europe as a result of emission reductions, while in rapidly developing countries like India, there was an increase in AOD (Kaufman et al. 2002).

The Arctic is unique and increasingly important to the study of aerosol-radiation interactions. Aerosol haze created from sulfates and black carbon emitted from industrial regions of Eurasia affects the surface balance of radiation in the late-winter and early-spring months (Koch & Del Genio, 2010). With loss of sea ice in the Arctic, the amount of open ocean will increase creating more marine-derived aerosols, as well as altering boundary layer dynamics surrounding the Arctic. The connection between this change in the Arctic and the overall warming of the climate system is an active area of investigation (Forster et al., 2007).

VII. CONCLUSIONS AND FUTURE DIRECTIONS

Aerosol particles have multiple effects on the balance of atmospheric radiation. Globally averaged, their most significant effect is a negative (cooling) effect, offsetting to some extent the warming caused by greenhouse gases from human activity, although there is a great deal of uncertainty in these estimates that affects the level of accuracy of estimating climate sensitivity (Myhre et al., 2013). The dominant negative direct effect is due to scattering aerosols (e.g. sulfate, sea salt and organic carbon), whereas the dominant positive direct effect is created by absorbing aerosols,

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especially black carbon (Ramanathan & Carmichael, 2008). Indirect aerosol effects on cloud formation and precipitation (specifically the first and second indirect effects) also produce negative forcing, but they remain the most uncertain sources of forcing (Lohmann & Feichter, 2005; Twomey, 1977). Areas to focus on in reducing uncertainty include improved characterization of black carbon (BC) mixing state and optical properties (Hansen et al., 2005), improved estimates of the formation of secondary organic aerosols, and improved representations of aerosol-cloud-precipitation interactions in global models (Schulz et al., 2006). Additionally, expanding ground-based and airborne measurement networks in sparsely populated areas (Holben et al., 1998) and developing next-generation satellite sensors with advanced aerosol typing capability (Boucher et al., 2013) are critical to enhancing climate research and atmospheric research.

As the amount of aerosols in the atmosphere continues to adapt to shifting patterns of emissions, it becomes essential to accurately measure the radiative effects of these aerosols on our climate to provide the best attribution of the climate changes that have been found so far, as well as good projections for any future increases in temperature (Charlson et al., 1992; Forster et al., 2007). Therefore, integrated observational and modelling activities will continue to be necessary to reduce the major source of uncertainty related to the Earth's energy budget (Haywood & Boucher, 2000).

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