GREEN, WHITE, AND BLACK POLLUTION: ENGINEERING AND GEO-ENGINEERING OF CLIMATE

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The global mean surface temperature of the Earth warms in response to the net increase in energy absorbed by the Earth. In a simplified or 'Textbook' model of this system, man-made emissions to the atmosphere can be classified in three types: (1) 'green' house gases (GHGs), which absorb infrared energy emitted from the Earth's surface, causing a 'blanketing' or warming effect, (2) 'white' aerosol particles that 'reflect' energy before it reaches the surface of the Earth, thus negating some of the incoming warming or causing a cooling effect, and (3) 'black' aerosol (such as black carbon, or BC) that absorbs energy thereby causing warming of the atmosphere. The latter two components may be mixed, so that some aerosol emissions have components of both energy reflection (cooling) and absorption (heating).

Many natural processes exist that produce 'white' aerosols, from sea salt to some kinds of dust, but most 'black' aerosols are produced by combustion, most of which has occurred since the Industrial Revolution (with the exception of some natural wildfires). However, most combustion processes emit not only BC but also reflecting aerosols and GHGs (primarily CO₂). In fact, natural and early man-made combustion processes emitted all three components roughly proportionally, although the ratios of emission types were dependent on burning conditions. As the Industrial Revolution proceeded, fossil fuel combustion in controlled engine burns began to far exceed wildfire and other biomass burning emissions. This meant that as population and the power-consumption by that population both increased, the amount of CO₂ produced increased, and both reflecting aerosols and BC tracked these increases, as suggested schematically by Fig. 1(A). As an estimate, these increases were approximately proportional, although technology, applications, and usage caused variability in different regions during different periods. The result of these increases of both 'green' and 'black' absorbing emissions along with increases in 'white' aerosol emissions was that the increasing amount of 'white' aerosol masked some of the heating that would otherwise have occurred from the 'green' and 'black' emissions.

In the wake of the 1952 London smog-induced respiratory-health-related deaths and the ensuing legislation in favor of limiting emissions in the United States and Europe, reflecting aerosols and BC were reduced by technology that removed particulates from the emissions of combustion processes. Interestingly, in California it turned out the removal of BC was more effective than of reflecting aerosols: California has thus 'engineered' reductions in diesel and residential biofuel burning emissions of black carbon over the last 20 years. The resulting cooling is -1.4 W m^{-2} (Bahadur *et al.*, 2011). While this result is likely not unique worldwide, it is only documented because the U.S. and California have monitored 'total' aerosols (including both 'white' and 'black' carbon-containing particles) since the 1970s (Bahadur *et al.*, 2011).

The resulting history of 'engineered' emissions for a region like the western U.S. is illustrated schematically in Fig. 1(B). While these emissions were 'engineered' to achieve the desired improvements in local and regional air quality, the impact on global temperatures would be less warming per combustion-generated energy, because less BC is emitted with the CO_2 . The reduction in emissions of reflecting aerosols alone would have led to the opposite, *i.e.* more warming per combustion-generated energy. However, fortuitously technology and controls led to an offset of the reduced cooling by the BC-related reduced warming.

Noting this history of the serendipitous impacts of air quality 'engineering' of combustion emissions on climate, a number of people have suggested that we can use this knowledge of the role of aerosol impacts on temperature to mitigate climate change in the coming years, as suggested by Fig. 1(C). The idea would be to intentionally add 'white' reflecting aerosols to the Earth system (or some other sun-reflecting method, SRM) (Shepherd et al., 2009). (Other ideas for climate engineering such as carbon dioxide removal (CDR) have been suggested also, but they are beyond the scope of this article). The idea of SRM would be to insert reflecting aerosols in an unpopulated area of the atmosphere, so that the additional aerosols will cause cooling but not respiratory disease. These proposed methods of offsetting man-made climate change are broadly referred to as 'geo-engineering' rather than 'engineering,' and the aerosol-related SRM methods are characterized by the purposeful production of reflecting aerosol emissions as an end in themselves rather than as a side effect of energy production. (See Fig. 1, end of document).

Two types of aerosol reflection can be considered. One is simply the injection of aerosols into the stratosphere, similar to that which occurs naturally in a volcanic eruption (Crutzen, 2006). While typically sulfate aerosols are suggested, the amount required is likely too small to have significant effects on deposition in populated or forested areas (Kravitz *et al.*, 2009). The other is to use aerosols over ocean regions to enhance the reflection of the lowlying stratus clouds that cover large areas of the globe (Latham, 1990). These cloud effects of aerosols effectively use the enhancement of naturally-occurring water vapor to increase the amount of light reflected. While the modification of cloud albedo has significant appeal due to its limited lifetime and regional application, it is worth noting that this 'indirect' effect of particles on clouds has significant uncertainties (Rasch *et al.*, 2009). Models use simple parameterizations of these effects to extrapolate behaviors that are well understood qualitatively but for which there is little quantitative understanding.

Uncertainties remain for the 'climate' or 'geo-engineering' effects of aerosols in the lower troposphere because multiple aerosol-cloud interactions are not modeled on the regional scale and because only limited observations are available for comparison (Table 1). One surprising aspect of particle-cloud interactions that has been provided by observations is the role of organic-dominated particles: Leaitch et al. (2010) have shown that non-reflecting organic particles may actually reduce cloud reflection in marine clouds, as a result of the underlying competition between particles of different sizes. To address these limitations in the observations of aerosolcloud interactions, two types of experiments to explore the quantitative nature of aerosol-cloud interactions: (1) one would use controlled aerosol emissions to generate spatially defined perturbations for comparison to regional models (e.g. the proposed Eastern-Pacific Emitted Aerosol-Cloud Experiment, E-PEACE), and the other would use long-term ground-based observations of cloud-particle interactions and droplet composition (e.g. May-Gray Experiments on Black Carbon in Activated Particles in Clouds). Both types of experiments are essential to reduce the uncertainty of aerosolcloud interactions in model simulations.

Without these additional observations, global model representations of aerosol-cloud effects are too simplistic (as summarized in Table 2). To date, they lack: (1) accurate aerosol particle activation, with the resulting implications for the profiles of supersaturation, vertical velocity, liquid water content, and drop distribution; (2) realistic microphysical growth and precipitation processes that control the formation and impacts of drizzle on cloud structure, lifetime, and particle concentration; (3) eddy-based transport processes that control the effects of entrainment on cloud thickness and lifetime as well as the dispersion of aerosol plumes. For similar reasons, the assessment of similar effects by aerosols on climate change is equally uncertain (IPCC, 2007).

Table 1. Summary of relevant observations for aerosol-cloud interactions in marine stratocumulus experiments.

Experiment (Region)	Publications	Key Findings (for aerosol-cloud interactions)
MAST (NE Pacific)	Russell <i>et al.</i> , 1999 Hobbs <i>et al.</i> , 2000 Hoppel and Frick, 2000 Durkee <i>et al.</i> , 2000 Noone <i>et al.</i> , 2000,b Ferek <i>et al.</i> , 2000	Observed changes in drop distributions and LWC profile. Ship emission characterization and size distributions. Case studies of four ship emissions that produce ship tracks. Test of aerosol-induced ship track hypothesis. Case studies illustrating background pollution effects on albedo sensitivity. Drizzle and LWC changes in ship tracks relative to unperturbed clouds.
DYCOMS II (Nocturnal) (NE Pacific)	Stevens <i>et al.</i> , 2003 Twohy <i>et al.</i> , 2005 Petters <i>et al.</i> , 2006 Hawkins <i>et al.</i> , 2008 Faloona <i>et al.</i> , 2005 vanZanten and Stevens, 2005	Characterization of POCs in nocturnal marine boundary layers. CN/CCN/CDN relationships are linear. CCN closure for marine boundary layer particles. Composition-independence of particle activation in the aged boundary layer. Entrainment rates and variability in the nocturnal marine boundary layer. Drizzle in nocturnal boundary layer in intense precipitation pockets.
MASE-I/II (NE Pacific)	Lu et al., 2007 Lu et al., 2009 Sorooshian et al., 2009	Ship tracks had smaller cloud drop effective radius, higher N_c , reduced drizzle drop number, and larger cloud LWC than adjacent clean regions, but trends were obscured by spatial-temporal variability.
EPIC & DECS (NE Pacific)	Stevens <i>et al.</i> , 2005 Sharon <i>et al.</i> , 2006	Rift POCs study; variability in cloud drizzle characteristics due to natural processes and emissions.
VOCALS-REx (SE Pacific)	Bretherton <i>et al.</i> , 2009-AGU* Twohy <i>et al.</i> , 2009-AGU* Feingold <i>et al.</i> , 2009-AGU* Wood <i>et al.</i> , 2009-AGU*	Offshore drizzle not explained by CCN decrease. Higher LWC had larger but fewer droplet concentrations. Aerosol gradients may cause POC formation POCs exhibited greater heterogeneity in drizzle and LWC.

*Abstracts from 2009 American Geophysical Union (AGU) Fall Meeting are accessible at http://agu-fmo9.abstractcentral.com/.

Model Type	Publications	Key Findings (for aerosol-cloud interactions)
ACP using observations	Russell <i>et al.</i> , 1999	Feedback effects of particles on supersaturation and LWC profile.
ACP with LES trajectories	Feingold <i>et al.</i> , 1998	Sensitivity of cloud properties to variability in trajectories.
ACP at Smax distribution	Hsieh <i>et al.</i> , 2008	Importance of maximum supersaturation distribution.
LES-Nocturnal (NE Pacific)	Hill et al., 2009	Inhomogeneous mixing less important than particles; results are insensitive to spatial resolution.
LES	Ackerman 2003	LWP is reduced as CDN increases.
(Pacific/Atlantic)	Ackerman 2004	Nighttime CDN increases will suppress drizzle.
LES-Diurnal	Lu and Seinfeld, 2005	Giant CCN increase drizzle in some conditions.
(Pacific/Atlantic)	Lu and Seinfeld, 2006	Relative dispersion increases apparent indirect effect.
LES-Nocturnal (NE Pacific)	Savic-Jovcic and Stevens, 2008	Reduction in cloud albedo associated with drizzle.
LES (SE Pacific)	Caldwell and Bretherton, 2009	Diurnal cycle controls drizzle and LWP.
Mixed-layer	Wood, 2009	Drizzle decreases cloud height and entrainment and CDN increases.
LES	Sandu <i>et al.</i> , 2009	Vertical stratification affects LWP; diurnal transition effects on LWP.

Table 2. Summary of recent MBL cloud responses to aerosol perturbations included in models.

In addition to scientific uncertainties that affect whether SRM methods would cool climate, what do we need to consider before starting climate engineering? There are four types of considerations: (1) Scientific uncertainties associated with implementation *i.e. Will it 'work'?;* (2) Costs and benefits (social, environmental, political), *i.e. If it works, who will pay and who will benefit?;* (3) Local and global governance, *i.e. Who will decide?;* (4) Ecosystem impacts and unintended consequences, *i.e. What if we are wrong about what will happen?* Research is needed to address each of these questions, from climate scientists and others, despite the controversial nature of research related to 'geo-engineering'.

There are also substantial ecosystem impacts of SRM that should be considered. These can be divided into: (1) Physico-chemical climate aspects:

temperature, precipitation, sunlight; (2) Ecosystem components: Community structure and taxonomic diversity, Biomass and productivity, and Biogeochemical cycling; and (3) Ecosystem services: Supporting (NPP, soil, nutrient cycling), Provisioning (fuel, fiber, food), Regulating (water quality, climate regulation), and Cultural (aesthetic, educational, spiritual). Current research suggests the ecosystem impacts may not be worse than expected from current climate change emission scenarios, unless SRM is undertaken for several years then stopped abruptly

(Russell *et al.*, *Ambio*, submitted 20 June 2011). While geo-engineering may lessen some of the climate changes of the foreseeable future, there is significant uncertainty about unintended consequences.

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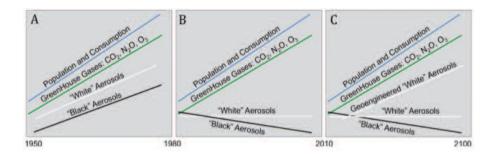


Figure 1. Schematic representation of relative changes in the emissions of fossil fuel combustion for three scenarios: (A) Fossil fuel combustion emissions without particulate, NO_{χ} , or VOC controls, similar to the United States before the implementation of Clean Air Act regulations from 1950 to 1980; (B) Fossil fuel combustion emissions with particulate, NO_{χ} , and VOC controls, similar to California after the implementation of Clean Air Act and state regulations from 1980 to 2010; (C) Same as (B) but with the addition of aerosols for SRM, as has been proposed for future amelioration of climate warming.